Project No. SN-01/18

MuSA

Integrating Multiple Scale Impact Assessment on Ecosystems for Contaminated site management

Final Research Report

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This report is the result of work jointly funded by the following SNOWMAN partners:

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ABSTRACT

In regard to the SNOWMAN research program in the field of soil-water-groundwater systems with respect to contamination, the main objective of the MuSA project is to propose methodological improvement for a decision support tool bringing together Environmental Risk Assessment (ERA) and Life Cycle Assessment (LCA) for contaminated soils management.

In order to achieve this aim the work was divided into (1) Improving multiple scale ecological risk assessment and Life Cycle Assessment of contaminated land, (2) Improvement of the understanding of the modelling of the mobility and bioavailability of metals in soils, and (3) the Ronde-Venen Case Study in order to (i) ensure the applicability of the developments; and (ii) proposing an illustration of the use of the propose sustainable land management in order to facilitate the understanding of users.

The development of a decision support tool especially responds to a need of soil resource management authorities for scientifically sound methods permitting the ecological risk assessment of contaminated soil at different spatial and temporal scales in order to have a better perception of the environmental impacts associates with the management strategy of the contaminated soils.

RESUME

Au regard des attentes du programme de recherche SNOWMAN dans le domaine de la contamination des sols, de l'eau et des nappes, le principal objectif du projet MuSA est de proposer des améliorations méthodologiques afin de permettre l'utilisation de l'Analyse de Cycle de Vie (ACV) et de l'Analyse de Risque Environnemental dans un même outil d'aide à la décision en ce qui concerne la gestion des sites contaminés.

Afin de réaliser ces objectifs, l'objectif du travail est divisé en trois parties, (1) améliorer l'évaluation des impacts à différentes échelles de temps et d'espace pour les sites contaminés ; (2) d'améliorer la compréhension dde la modélisation de la mobilité et de la biodisponibilité des métaux dans le sol ; et (3) de mettre en application des observations dans le cadre d'un cas d'application sur le site de Ronde Venen, d'une part pour s'assurer de l'applicabilité des observations mentionnées, et d'autre part pour disposer d'une illustration qui permette de mieux présenter les enjeux relatifs à la gestion durables des sites contaminés.

Le développement d'un outil d'aide à la décision repond essentiellement à un besoin des organismes en charge de la réglementation concernant la gestion des sites contaminés qui souhaitent faire appel à des méthodes permettant d'intégrer différentes échelles de temps et d'espace dans la gestion des sites contaminés afin d'avoir une meilleures quantification des impacts associées aux stratégies de gestion.

EXECUTIVE SUMMARY

The focus of the MuSA project (Integrating Multiple Scale Impact Assessment on Ecosystems for Contaminated Land Management) is the development of a decision support tool combining the complementary analyses of Ecological Risk Assessment (ERA) and Life Cycle Analysis (LCA).

As a first step the report identifies the similarities and main differences between the ERA and LCA approaches focusing on the impact of contaminated site on ecosystems.The notion of the functional unit and the implication of the choice of the system boundaries in the LCA of contaminated sites, are key discrepancies between LCA and ERA situated before the impact assessment step. At the next stage, even if the two approaches cover the same questions, the scope of the tools differs since LCA is used for comparison purpose while ERA is applying for conservative assessment. However, a tendency of the two approaches getting closer and closer can be identified. This development includes the identification of limits to integration, and the required methodology development to increase the coherence between the two approaches in addressing the complex question of the impact on ecosystems, especially the change in biodiversity.

A further result of this project is the presentation of the key ecological parameters for multiple scale assessment, the selection of the most relevant indicator for the assessment of soil quality, and presentation of the minimum required dataset for contaminated soil characterisation for the purpose of a realistic characterisation of the behaviour and impact of trace metals on soil ecosystems to give answers on soil contamination by trace elements. Following the research results, recommendations include the use of a system of bioindicators that include information at the levels of communities of soil invertebrates, plant communities, and soil ecosystem processes. The use of these parameters in site characterisation provides the framework for quantifying the ecological impact of soil contamination. In this vein, suggestions are given for onsite data collection that will facilitate comparison between contaminated sites once all data are combined into appropriate databases.

Another important aspect of contaminated sites are metals and their transfer from soil to water. Hence results of the MuSA project include the modelling of impacts of metals on ecosystems, and the theory.A review of available models to identify the intermedia transfer and to quantify the mobility of metals in soils especially considering their ability to be applied in LCA showed coherent results: the high mobility of Cadmium and Zinc is highlighted, while Cu and Pb appear to be more sensitive to the media conditions. For these well studied metals four key parameters could be identified and the modelling of impacts is currently feasible. However, impact assessments of other metals such as anions are to date not feasible using these approaches and it will require further investigations.

For cations the report gives a description of the key parameters for biological and chemical availability and their relation with time. The complexity associated with the soil matrix and the terrestrial ecosystem it supports are illustrated using examples of how the influences of multiple scales of time, space, and biological organization will modify metal behaviour in soil over both short and long time periods.

In the context of Sustainable Land Management, and in the special situation of Risk-Based Land Management, the concepts related to metal solubility and mobility are illustrated using a case study which show the use of the above described results. The historically metal-contaminated Demmerik polder site located in the Dutch municipality of de Ronde Venen is used to illustrate an approach to the assessment of contaminated site management options based on management scenario evaluations. Three main management scenarios are evaluated by two environmental assessment methods: Life Cycle Assessment (LCA) and Ecological Risk Assessment (ERA). The results of both assessments are combined to provide recommendations for site management strategies that incorporate concerns for sustainability into the decision-making process. However, interpretation of the results is not strictly a scientific question, and the choice of the most appropriate management strategy ultimately depends on the priorities of the decision-making group. Due to the interdisciplinary nature of this multiple-scale combined assessment, efforts have been made to include involvement of researchers, regulators, and industry members in the development of an innovative decision-support framework in order to give valid recommendations.

Finally, an interactive workshop provided the opportunity for interested parties to come together and collaborate and to publicly discuss the outcomes of MuSA. Special focus was placed on efforts to examine existing knowledge regarding four principle themes, which are the spatial differentiation of environmental impact assessment, biodiversity, ecotoxicology, and the bioavailability and mobility of metals in soils. The examination of each of these themes from a perspective of both LCA and ERA resulted in the identification of areas where these tools overlap, areas where they complement each other, and areas where they each need improvement.

Overall, the MuSA project has been able to provide some insight into how the simultaneous consideration of local and global impacts of contaminated site management can inform decision-makers and stakeholders as to the short and long term effects of management strategies. The current study recognizes the need for decision-support tools that can quantitatively evaluate the ecological impacts of site management strategies. Further research efforts are indicated, with strong recommendations to continue the fruitful exchanges that result from an interdisciplinary approach.

ACKNOWLEDGEMENT

MuSA is a trans-national research project jointly funded by the partners of the soil and groundwater ERA-Net SNOWMAN (http://www.snowman-era.net) (SN-01/18 MuSA).

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I USE OF GRANTS

In the frame of the SNOWMAN ERA-NET project, the MuSA project was initially planned with an overall budget of 141972 Euros, among which 43% was funded by ADEME, 28.5% by SETEMIP, 20% by SKB and 8.5% by VU Universiteit Amsterdam. Within the initial budget, 90% was allocated to the staff cost and the rest has to cover the travelling fees associated with the project. Nevertheless, the short duration of the project requires to have many opportunities to discuss the advancement of the project between partners. Furthermore, a strong effort was made with the dissemination dissemination of the project outcomes, it was espacially the case with the presentation of three posters at the SETAC EUROPE meeting in may 2008 in Warsaw, and with the organisation of a workshop with 36 participants in september 2008 in Amsterdam. These constraints lead both partners to invest more money in the project compared to the initial plan.

II BACKGROUND

The overall objective of the project is to develop an innovative decision support tool for Sustainable Land Management providing ecological risk estimate at the local and short-term scale but also at a broader time and space scale. This purpose will be reached using two axis of work: the use in parallel of Ecological Risk Assessment and Life Cycle Assessment tools in a one assessment of contaminated land and management strategy; and the acquisition of new knowledge related to chemical and biological availability of metals in soil.

The project aims at identifying the gaps of existing methods for impact assessment of soil contamination on ecosystems and proposing an innovative multiple scale assessment method at the interface between Ecological Risk Assessment (EcoRA) and Life Cycle Assessment (LCA). This new method aims at supporting the development of a decision support tool for Sustainable Land Management easily applicable to contaminated land and enabling the identification of the optimum trade off between impacts of contaminants and impacts of the remediation strategy. Current approaches in EcoRA and LCA needs to better address impacts of metals and metalloids. The project will focus on these substances, even if the proposed methods will also be applicable to organic substances.

The key issue of the project is to enable a multiple scale assessment of the ecological risk associated with a contaminated land with the integration of the impacts over time and the estimation of the risk at the landscape (or the watershed) level.

The second axis of developments is related to the availability of metals in soil. Biological and chemical availability of contaminants is considered as a major parameter determining the risk on a short and long-term scale. Therefore a strong emphasis will be put on modelling of bioavailability, ageing and speciation of metals in soils. This will enable the clarification of the stabilization and remobilisation processes of metals in contaminated soils and the estimation of the inter-media transfer in risk assessment at a broad scale.

These two axis of research will enable the identification of (i) the key ecological parameters that are necessary in the risk assessment procedure for contaminated sites; (ii) the most relevant indicators enabling the determination of the soil quality; and (iii) the minimum dataset required to enable the characterization of the contaminated land.

Final aims is an innovative decision support tool a Sustainable Landscape Management strategy using both EcoRA and LCA knowledge in order to understand implication of the contaminated land management at different time and space scale.

In order to ensure the applicability of the proposed strategy, the findings of the project will be applied to the “Ronde Venen” case study. This well known large contaminated land is polluted since several centuries. Furthermore, this area also presents a large variability of substance concentrations. Therefore, the management strategy of the landscape requires both an efficient and reliable estimate of the risk and the prioritisation of the remediation strategy depending on the level of contamination, the stability of the substances in soil, and the land use organisation on the long term. The screening tools developed in the project will enable the integration of these three aspects in the remediation strategy.

Beyond the project itself, the communication of the results is a priority. Therefore, several ways of dissemination will be used such as web sites (Project website and EUGRIS web site), scientific publications, and training of stakeholders and university students using output of the project in Ecological Risk Assessment training.

III AIMS AND COMPARISON WITH PREDETERMINED OBJECTIVES

The main objective of the project is to build up a decision support tool bringing together Environmental Risk Assessment (ERA) and Life Cycle Assessment (LCA) for contaminated soils management.

The following actions were achieved:

WP1: Project Management and coordination

Main actions:

- Kick off meeting took place an VU-Amsterdam the 10th of October 2008.

- Leaders of the two partners have regular meeting.

- Working meeting took place at VU at least ones per month

- A workshop was planned at the end of the project

- Final deliverables was finalized with end of October.

WP2: Multiple scale ecological risk assessment of contaminated land

Objective: enabling a multiple scale assessment of the ecological risk associated with a

contaminated land with the integration of the impacts over time and an estimation of the risk

at the landscape (or the watershed) level.

Main action:

- Bibliography review of existing studies on the topic of multiple scale assessment in LCA

- Bibliography review of existing studies exploring the interface between LCA and ERA

- Comparison between LCA and ERA in terms of modelling, assumptions and concepts and identification of similarities and discrepancies between tools.

- Review and description of key parameters which could be used for describing contaminated site and for providing data both for LCA and ERA

- Identification of the data and models needed to manage at the same time LCA and ERA.

WP3: Speciation, ageing and bioavailability

Objective: Modelling of the bioavailability, ageing and speciation of metals and metalloids in

soils and the assessment of the transfer from the soil to the other environmental compartment.

Main actions:

- Review of parameters required for assessing mobility and bioavailability of metals in the site characterisation process, selection of the key parameters and description of the key parameters influencing the mobility and bioavailability of metals.

- First estimate of the influence of water percolation and organic matter in the transfer of metals from soil to surface water.

- Presentation of the indicator which can be used for the assessment of soil quality and the minimum requirement dataset for contaminated soil characterization

WP4: Ronde-Venen Case Study

Objective: Application of the developed decision support tool in a case study in order to (i) ensure the applicability of the developments; and (ii) proposing an illustration of the use of the propose sustainable land management in order to facilitate the understanding of users.

Main actions:

- Data availability has been checked to perform at the same time the Environmental Risk Assessment of the site and the Life Cycle Assessment.

- Literature review of sustainable land management strategies for Ronde Venen

- LCA of the Ronde Venen was started, including several alternative scenarios: the natural attenuation, an in situ phyto-remediation, and ex-situ thermal treatment of the soil.

- Development of a regional LCA study for Ronde Venen that can be used as an illustration of what can possibly be done to provide LCA study closer to ERA.

- Deviation from the project objectives, if any: state reason.

1- Finally the project mainly focuses on cations because Zn, Cu, Pb and Cd are the substances that pose problem in the Ronde Venen case study.

2- Due to the substances selection, the applicability of the HSAB concept for soils was not relevant since the substances considered are borderline substances that require the use of other models. Therefore, we have focused on the review of existing models for assessing the mobility of metals in soils and the improvement of existing models to make it applicable for Zn, Cu, Pb and Cd, metals that are considered in the project.

3- It seems feasible to have a partial integration of LCA and ERA for contaminated site management, nevertheless, the developments are still at the research level and it is not yet possible to have a guideline presenting a method for achieving integration of LCA and ERA approaches. Instead of producing a guideline, the project comes out with a list of recommendations and a demonstration case study of spatialized LCA that can be used to manage the interface between LCA and ERA.

4- At the time of the duration of the project, is was not feasible to provide a guideline for using at the same time LCA and ERA for contaminated site management, due to the lack of important data. However, in order to meet the predetermined objectives as best as posible and to make use of the project resources, the project partners elaborated a compendium of recommendations plus a demonstration case study for the use of LCA and ERA for contaminated site management. The realisation of the comprehensive compendium and the extra demonstration case study presented an extensive task, demanding more personnel time and more budget as predicted for a drafting of a guideline. Nonetheless, thanks to the effort of all partners, and to the accorded additional budget by SETEMIP, this recommendation and demonstration documents have been realised, in order to compensate the lack of data.

• Deliverables

Work Package 1:

Deliverable	Title	Planned deadline	Delivery
D1.1	Mid term report	10-04-07	10-04-07
D1.2	Final report	10-10-08	25-10-08

Work package 2:

Deliverable	Title	Planned deadline	Actual deadline
D2.1	Comparison between ERA and LCA	30-4-08	10-06-08
D2.2	Presentation of the key ecological parameters for multiple scale assessment and selection of the most relevant indicator for the assessment of the soil quality	30-04-08	10-04-08

Work package 3:

Deliverable	Title	Planned deadline	Actual deadline
D3.1	Description of the key ecological parameters for biological and chemical availability of cations and oxyanions and their relation with time	30-09-08	10-10-08
D3.2	Presentation of the minimum requirement dataset for contaminated soil characterization	30-03-08	10-04-08
D3.3	Determination of the potential Hazard of metals	30-09-08	25-10-08

Work package 4:

Deliverable	Title	Planned deadline	Actual deadline
D4.1	Literature of sustainable land management strategy applicable to Ronde Venen	30-04-08	10-05-08
D4.2	Application of the Sustainable land management approach to the Ronde Venen	10-10-08	10-10-08

Work Package 5:

Deliverable	Title	Planned deadline	Actual deadline
D5.1	Guideline for the sustainable management of soil and groundwater pollution using LCA and ERA	30-07-08	25-10-08
D5.2	Scientific publication presenting the output of the project- Synthesis of the MuSA workshop-	10-10-08	25-10-08

- Deviations from the project work programme, and corrective actions taken/suggested: identify the nature and the reason for the problem, identify contractors involved.

1- Change in the content of the workpackages

Two aspects were requiring a redefinition of some deliverable contents. It especially concerns the list of the substances identified as key substances in the Ronde Venen case study, and the need of further work for achieving integration of LCA and ERA approaches. This lead to the following redefinition of the deliverables D33 and D51.

D 33: Due to the substances selection, the applicability of the HSAB concept for soils was not relevant since the substances considered are borderline substances that require the use of other models. Therefore, we have focused on the review of existing models for assessing the mobility of metals in soils and the improvement of existing models to make it applicable for Zn, Cu, Pb and Cd, metals that are considered in the project.

D51: It seems feasible to have a partial integration of LCA and ERA for contaminated site management, nevertheless, the developments are still at the research level and it is not yet possible to have a guideline presenting a method for achienving integration of LCA and ERA approaches. Instead of producing a guideline, the project comes out with a list of recommendations and a demonstration case study of spatialized LCA that can be used to manage the interface between LCA and ERA.

2- Change in the deadline of the workpackages activities

The deliverable D2.2 initially planned for the 30 of April was delivered earlier the 10 of April at the same time as the deliverable D 3.2.

The workshop organisation in September was time consuming during summer and did not allowed us to write the final reports. Therefore, reports due at the end of the MuSA project where delayed by 2 weeks. It especially concerns the deliverable D33 and D51. The deliverable D52 is now available but not yet submitted as an article since it requires the review of the partners.

IV RESULTS

Deliverable 2.1:

Similarities and differences between EcoRA and LCA

Date of preparation: June 2008

Start date of project: October 10th, 2007 **End date of project:** **October, 10th 2008**

Report author: Jerome Payet

Project coordinator name/author: Jerome Payet

Project coordinator organisation name: SETEMIP-Environnement

Date: Octobre, 22nd, 2008

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1 Abstract:

Life Cycle Assessment (LCA), and its impact assessment component Life Cycle Impact Assessment (LCIA), can be used to assess impacts due to contaminated sites. Nevertheless, so doing, the assessment can be compared to the assessment performed by Ecological Risk Assessment (EcoRA) for contaminated site. Furthermore, LCIA is know addressing the impact on ecosystems considering a change in biodiversity due to a change in environmental pressure in the media in a different way than what is done with ERA but with a similar question. It is the purpose of this report to first identify the similarities between the two approaches, but also the main differences. Nevertheless, due to the vast area of applicability of LCA, the work especially focuses on the impact of contaminated site on ecosystems. After describing the key properties of the two methods, the work is highlighting the common point between LCA and ERA for impact assessment on ecosystems. Afterward, the main sources of discrepancies are discussed under the perspective of the needs and objectives of each method. This discussion starts with the presentation of the notion of functional unit, and the implication of the choice of the system boundaries in the LCA of contaminated site. These two aspects are key discrepancies between LCA and ERA since they take place before the impact assessment step. Beyond this stage, even if the two approaches covered the same questions, the scope of the tools differs when LCA is used for comparison purpose while ERA is applying for conservative assessment. Afterwards, the work is discussing issues linked with the LCIA and ERA such as the time scale and the space scale issue, or the notion of biodiversity and the link with the biological organisation level and the “midpoint” and “endpoint” commonly used in LCA. As a conclusion, the work is discussing the tendencies of the two approaches in getting closer and closer, identifying possible limits to integration, and the methodological development that can be needed to increase the coherence between the two approaches in addressing the complex question of the impact on ecosystems, especially the change in biodiversity.

2 Introduction

Context

Life Cycle Assessment (LCA) and Environmental Risk Assessment (ERA) can be used in parallel for selecting the most appropriate land management strategy. Nevertheless, even if both approaches can now address impact on ecosystems and human health, the methodological background is very different and results can differ considerably. Furthermore, since both tools can not currently be applied in the same framework, some situation can occur where the environmental burden of the remediation can be higher than the impact of the site itself. It is the purpose of this work to identify the similarities and discrepancies of the methods and to propose a coherent common framework for applying LCA and ERA at the same time for selecting the most appropriate strategy in a contaminated land management perspective for terrestrial and aquatic ecosystems

Ecological Risk Assessment for contaminated site management

Ecological risk assessment is a process of collecting, organising and analysing environmental data to estimate the risk of contamination for ecosystems. Ecological Risk Assessment for contaminated site is rather different that EcoRA for substances regulation since it is focusing on pollutions already present in the environment. It is therefore more an impact assessment than a risk assessment. EcoRA for contaminated site is typically perform in phases or tiers. The tiered assessment for ERA starts with a simple screening at tier one, followed by a refined screening at tier 2 and then a detailed assessment at tier 3.

Ecological Risk Assessment method can be presented in 4 steps:

1- Hazard identification : identification of the capacity of the chemical to cause adverse effects
2- Exposure assessment: determination of the emission volume and the level of exposure
3- Hazard Assessment: determination of the dose response relationship for the chemical, estimating a level of effect for different exposure concentration
4- Risk characterization: comparison of the exposure and the effect assessment

These four steps are organised as presented below.

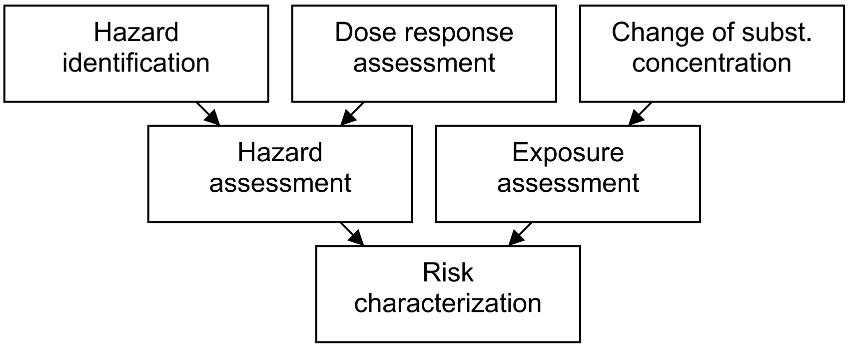

Figure 1*: Illustration of the risk assessment method (after Flemström et al. 2004)*

Life Cycle Assessment and Life Cycle Impact Assessment

Life Cycle assessment (LCA) is a method that aims at compares the relative environmental burden of different products (or services) satisfying the same service. The comparison is performed regarding different impact categories. The key point of LCA is the notion of equivalence of service which requires defining a functional unit, expressing the unit of service that as to be fulfil by the different products compared in the study. The functional unit is the basis of the comparison and is the basis of all environmental assessment "product oriented" such as LCA or eco-design. As describe in the international standard ISO 14040-44, Life Cycle Assessment is performed following 4 stages.

A- Definition of the scope and goal : build up the framework of the evaluation, defining the product or service evaluated, the alternatives compared and the purpose and limits of the evaluation
B- Life Cycle inventory: estimate the amount of resources that are consumed for achieving the product and the amount of substances emitted in the air, water and soil
C- Life Cycle Impact Assessment (LCIA): quantifies the environmental burden associated with the inventory emissions, covering different impact categories related for example to resources consumptions, global warming, human health and ecosystems quality. The LCIA can be done at the midpoint level considering a large number of impact categories somewhere in between the emissions and the final environmental burden, or at the damage (endpoint) level, considering a very limited number of impact categories (commonly four) that are very close to the Area of Protection or the Safeguard Subject that is of interest in LCA.
D- Interpretation: addresses the level reliability of the results of the LCA.

Among the 4 phases of LCA describes above, the Life Cycle Impact Assessment phase is very close to the environmental risk assessment procedure especially concerning impact categories such as impact on ecosystems or human health. Within this part of LCIA, several steps of the assessment can be distinguished:

1- Classification: identifying the substances that are likely to cause an effect in an impact category. This stage of the LCIA was very important when only a very small number of substances were characterized but now the tendency is to address the potential toxicity of all substances present in the inventory, considering very large substances dataset.
2- Fate modelling of the substances listed in the inventory: using an intermedia fate model, has the purpose of quantifying the fraction of the substance transferring from the media where the emission takes place to the target media (it therefore averages the concentration of substances over area

covered by the space scale); and integrating the concentration of the substances overtime making it compatible with a steady state modelling.

3- Exposure modelling: Estimating the fraction of the substances that is causing adverse effects within the total amount of substance that is present in the compartment.

4- Effect assessment: based on a dose-effect relationship relating the increasing stressor occurrence and a increase in the intensity of the effects expected.

5- Damage modelling: That aims at facilitating the interpretation of the results; (1) Integrating together different stressors related to a single safeguard subject, it therefore ranks the different impact categories in terms of priority and reduce the number of categories considered typically from 15-20 to 4 damage categories; and (2) Facilitating the interpretation of the results, enabling the understanding of the impact in terms of absolute value (such as the DALY) and not in a relative assessment value (Like the substance equivalent).

The different stages of the life cycle impact assessment are presented below.

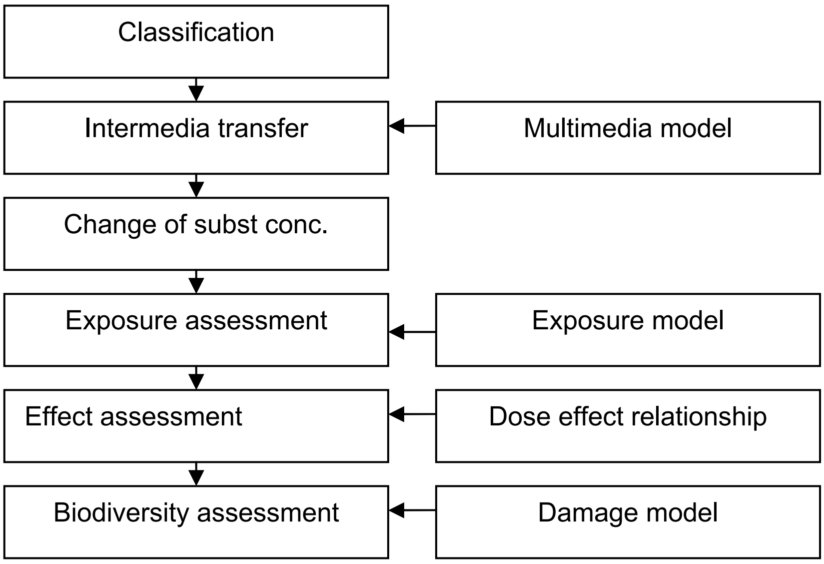

Figure 2: Illustration of the Life Cycle Impact Assessment method

As we can see in Figure 2: Illustration of the Life Cycle Impact Assessment method, the structure of the Life Cycle Impact Assessment process is rather different than the ERA. Furthermore, the terminology such as “exposure” does not cover the same reality in the two tools, and at the end, ERA comes out with the characterisation of a risk while LCA provide the quantification of a damage.

3 Comparison Ecological Risk Assessment and Life Cycle Assessment

Main similarities between the tools

Looking at the schematic description of the two approaches in Figure 1 and Figure 2 it helps understanding some differences in terminology between the two approaches. It must be first noted that the “classification” mention in LCIA have strong similarities with the “Hazard identification” of EcoRA. Then the “Exposure assessment” in EcoRA aims at quantifying an exposure concentration. This is therefore rather similar to the “Life Cycle Inventory & Fate modelling” of LCA that aims at calculating a change of concentration of a chemical in the environmental media for each inventory emission. Then the “Hazard assessment” mentioned in EcoRA can be viewed as the “exposure & effect modelling” of LCIA, since if enable the calculation of the impact on ecosystems, and then the “risk characterisation” step of EcoRA presents similarities with the “Damage modelling & interpretation” of LCIA. Even if the tools does not present similar structure, they appears to be quite coherent as soon as we consider carefully the term of “exposure” assessment which covers different things whether we are in LCIA or EcoRA.

In terms of management of the tools, EcoRA for contaminated site management is a tiered approach commonly applied in three tiers. LCA is commonly presented as a one tier assessment, nevertheless, LCA is commonly manage with different level of details which presents some similarities with the EcoRA procedure as presented below.

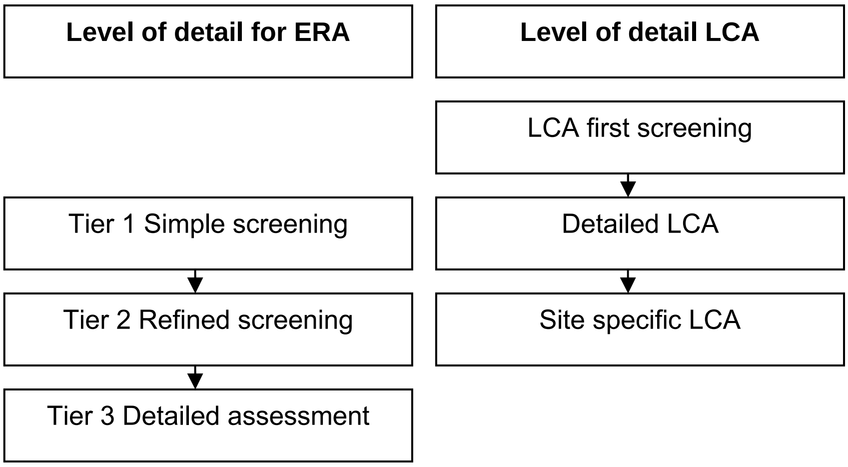

Figure 3: Comparison of the tiered ERA approach and the different level of details of LCA

As presented above, both EcoRA and LCA can be considered as tiered approaches. This is commonly accepted for EcoRA, but LCA is not thus describes. Nevertheless, efficient management of LCA studies requires to start with a “First Screening”, enabling the identification of the key environmental aspects, and then to focus on those points for the detailed LCA. Nevertheless, “Detailed LCA” is typically performed with tools already available in commercial softwares that are generic assessment tools. The development of a site specific LCA requires the collection of environmental data form the site itself and therefore requires a considerable effort. This can therefore be considered as a refined LCA study. Furthermore, in between the generic LCA and the site specific, a regional LCA can be performed with a spatial resolution at the watershed scale.

Finally, the main similarity between the two approaches is their overall purpose to quantify environmental burden of human activities. Concerning impact on ecosystems, questions addressed by the Life Cycle Assessment and Ecological Risk Assessment are aiming at quantifying the impact on biodiversity. Nevertheless, beyond this similarity, numerous discrepancies can be observed. It is therefore important to check if the tools are compatible. Indeed the two tools will be more and more used in parallel and we shall ensure they will not give opposite conclusions as presented in figures 4 and 5.

After considering the similarities between tools, discrepancies can now be addressed.

Comparative LCA versus absolute EcoRA

LCA was first designed for comparative assessment purpose. The tool enables a comparison between several alternative and identify their relative impact for each impact category considered. EcoRA is applied to assess if a site is at risk or not. This can be assessed considering threshold of effects which corresponds to an acceptable level of effect. For site remediation, threshold are defined in a first tier assessment (such as Target or Intervention values), but detailed assessment therefore has to consider a reference site. This specificity of EcoRA has some implication for LCA. Indeed, LCA is working in a comparative way to assess impact associated with different scenarios. This strategy can be viewed as a “less is better” where the environmental optimum is the zero emission level. ERA is commonly working with a threshold level (typically for substance regulation) or with a reference site (for contaminated site remediation). Dealing with contaminated site management, LCA has to consider the site itself as a part of the technosphere tending to a zero level emission. The alternative would be to consider the site itself as a part of the ecosphere, thus taking into account its own intrinsic value; nevertheless, this is not compatible with a 0 level emissions for all substances concerned by a natural background. In that case, the reference site used in ERA can be also used as a reference scenario in LCA, therefore the downstream impacts associated with this scenario could be considered as residual impacts. This could be of major importance for contaminated sites occurring in areas affected by a high level of background concentration of pollutants. In that case, the residual impact could be high and the environmental efficiencies of the remediation strategies shall be put in perspective with this residual background.

The Functional Unit, a key difference between the tools

The unique fundamental difference between LCA and ERA is the notion of functional unit which is a key aspect of LCA and where no equivalent can be found in ERA. All the other specificities mentioned below are due to the models, databases or the framework of the methods and are likely to change with the evolution of methods. It must be noticed that all part of the methods that corresponds to EcoRA is the Life Cycle Impact Assessment (LCIA). We will therefore now focus on that part of LCA.

System boundaries, in-between technosphere and ecosphere

In the Definition of the Scope and Goal of the LCA, the boundaries of the system under study are defined. For contaminated site remediation, it is important to decide whether the site itself belongs to the technosphere or the ecosphere.. In the first case, the site is considered as a reservoir of pollutants (such as a landfill) but not as an ecosystem itself, in the second case, it is considered as an ecosystem. Nevertheless, in that case, LCIA need to be adapted to take it into account especially in terms of site specific differentiation for fate and effect modelling. As presented in the table below, as soon as the contaminated site has an intrinsic value as a landscape, ecosystem, or species composition, it has to be included in the ecosphere and not the technosphere.

Tableau 1: Intrinsic value of the site

Site with intrinsic value	Site without an intrinsic value
Landscape or ecological value must be considered in the management strategy	Impact resulting from the site has to be considered disregarding the modifications of the site itself
The site is to be considered as a terrestrial ecosystems and its value shall be considered in the corresponding impact category in LCA	The site is only considered in LCA as an industrial system releasing pollutants and transferring it to other media
Ronde Venen is used in the MuSA project to represent this sort of sites. This site is a polder composed by a peat soil contaminated mainly by Cd, Cu, Pb, Zn.	Recent industrial contaminated area without historical values and strong contamination corresponds to this sort of sites

It is important to distinguish from the early evaluation of contaminated site those that have an intrinsic value such as landscape or ecological value and the sites that simply require a remediation. The first group will be addressed as terrestrial ecosystems and the possible remediation perspectives will have to take into account their intrinsic value, while the other group will just be addressed as a storage or pollutant progressively released in the other environmental media.

Characterization factors in LCA versus dose/effect relationship

LCA is using characterization factors for quantifying the impact of a given substance on the ecosystem. The impact is obtained simply multiplying the emission of a substance in an environmental media (such as air, water or soil) by the characterization factor corresponding to the substance, the emission media, and the target ecosystem. The overall impact on ecosystems in LCA is resulting from the association of a life cycle inventory table and a characterisation factors table. In its experimental perspective, ERA is mainly developing or using concentration/effect curves experienced in the field or in laboratory with an interpretation of the possible consequences of this relation considering the concentration in the field and the sensitivity of the ecosystem or of some of its components. Thus, while LCA is simply using one value to assess the impact of a substance emitted in a media or present in it, ERA is dealing with more qualitative information that are relevant for the site and which could not necessarily be extrapolated to other site.

Differences in assessment scale between the EcoRA and LCIA

Historically, LCA was addressing impact at a large scale such while ERA was mainly focusing at small scale. It was the case for time and spatial scale. This issues of spatial scale, time scale, and biological organisation level are addressed in more details below.

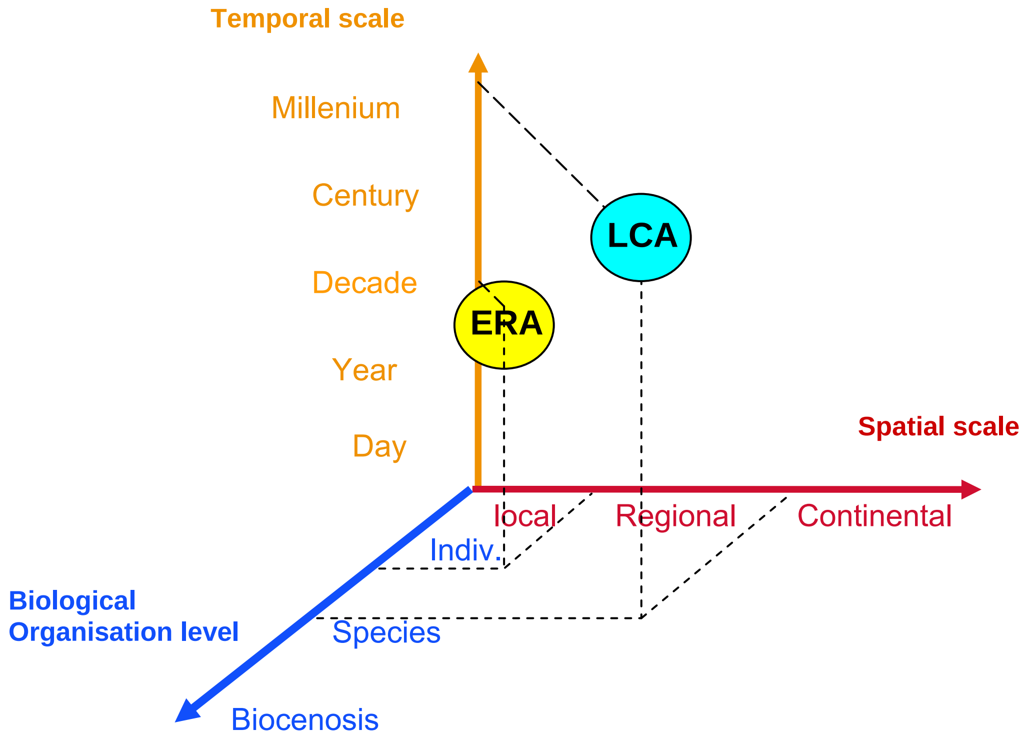

Figure 4: Comparison the EcoRA and LCA in terms of assessment scales

As presented in the figure above, ERA is designed for assessing hazard at a local scale while LCA deals mainly with continental scale. Concerning Time scale, ERA aims at estimating the hazard in a short term perspective, while LCA mainly addresses a long term perspective (100 to 1000 years). And finally concerning biological indicator, ERA is working on a small dataset of well known biological indicators, LCA estimates a percent of change of biodiversity in the ecosystem on the basis of laboratory test species.- Reference value: ERA is based on the notion of an acceptable risk define by a threshold value (the target value) while LCA is based on the “less is better” method. These questions of spatial and time scales are discussed in more details below.

3.1.1 Spatial scale and spatial differentiation

The spatial scale issue is a key problem for the integration of LCA and ERA. Using spatially differentiated models in LCA is a possible response to help resolving this problem.

Managing spatially differentiated modelling in LCA requires to apply it at three different level in the Life Cycle assessment process; at the inventory level (considering spatially differentiated emissions); at the fate model level (including site parameters for a given location in the multimedia transfer model); at the exposure and effect level (considering the actual bioavailability of substances and the actual specie biodiversity composition of the site).

Spatially differentiated Life Cycle Inventory

Practically site remediation is interesting in LCA since we can consider among scenarios the alternative of a “Doing nothing” scenario. In that case, all the emissions are from the site itself and we have therefore a highly spatially differentiated inventory. In all other situations in LCA, working with spatially differentiated emissions has been done for some illustrative studies (such as paper mills for example) but can not be done as routine studies.

Spatially differentiated multimedia modelling

As presented in the table below, ERA is mainly working at a local scale and LCA has been first developed for impact categories that produce impact at a global scale.

Tableau 2: Presentation of different spatial scale for LCIA and ERA

Scale	Local	Regional	Continental
Indic. Km^2	0.0001	2 500	25 000 000
LCA	Site	Watershed	Generic
ERA	Site	Intermedia	No

Generic LCA gives results at the continental or global scale while ERA mainly works at the site scale. Regionalization of LCA models gives a promising perspective in assessing impact at a coherent scale with ERA.

Spatially differentiated exposure and effect models

Generic exposure and effects models are based on an average exposure at a continental level, and effect modeling is based on the calculation of the HC50 (Hazardous Concentration of substances affecting 50% of the organisms above their chronic EC50) for laboratory organisms that are compatible with the area considered. The use of spatially differentiated models of exposure and effects requires the calculation of exposure concentration based on local or regional media parameters, and calculation of HC50s compatible with the organisms presents in the site, or some groups of organisms that can be of specific interest in the considered area.

3.1.2 Time scale and dynamic modelling

Tableau 3: Comparison of time scale issue for impact assessment in LCA and ERA

Years	10	100	1 000	10 000	60 000 to inf.
Persp.	Management	Human life	Civilisation	Ecosystem	Geological
ERA	Yes	No	No	No	No
LCA	No	LCI Short term	No	No	LCI-LCIA

Developing a single decision support tool for LCA and ERA requires working at a coherent time scale. It is possible to develop impact assessment methods for ecosystems working at 100, 1000 and 10000 years for

LCA. Nevertheless, the highest time scale considered in ERA is 10 years (for considering intermedia transfer of pollutants for example) while in LCA, the shortest time scale manageable with current models is the century. It clearly demonstrates the gap we have in terms of time scale between the tools. This problem is mainly due to the underlying assumption of steady state modeling that we are using in LCA. Under this assumption the integration of the concentration over time suggests an exposure to constant concentration of chemicals. Promising works have been done for the development of dynamic modeling in LCIA, nevertheless, such modeling requires considering a considerable number of media parameters which makes it unfeasible at this time in an industrial case study.

Notion of midpoint and endpoint in LCA

The notion of midpoint and endpoint is a key issue in LCIA. Indeed, LCIA was first focussing on global impact such as resources consumption and global warming, and therefore midpoint indicators were developed to address these impacts. Nevertheless, the evolution of the tool lead to address now impact categories such as human health or ecosystems, and it was therefore necessary to assess impacts closer to the area of protection level (AoP). For that purpose, endpoint (also called damage) modelling has been proposed by developers in order to better support the understanding of the impact. Practically, climate change is currently considered as an endpoint indictor even if impact is assessed as the midpoint level. The main reason for that is the limitation of the impacts assessment methods which does not allowed to provide a link between the midpoint level and the consequential impacts on ecosystems and human health.

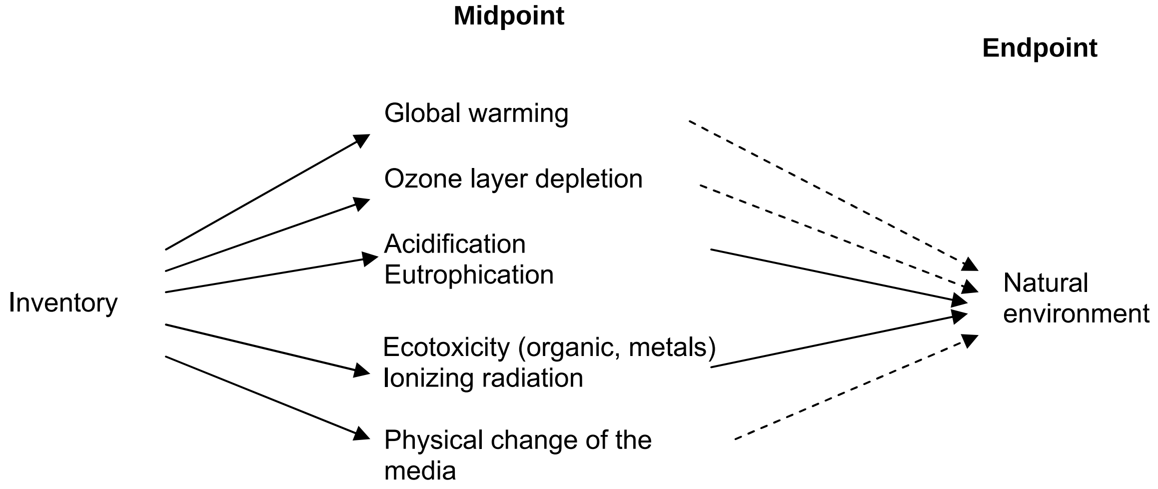

Figure 5 : Integration of environmental stressors in LCA

ERA does not address this issue of midpoint and endpoint. Indeed, the method is assessing damage occurring at the site scale regarding an existing biodiversity. Therefore, midpoint level assessment is not relevant for ERA methods. Furthermore, ERA approaches are assessing anthropogenic stressors separately linking the stressors with the different target that can be of concern in the site. In comparison, LCA is using endpoint modelling to add impact of each stressor on a single final target.

Biodiversity issue and related parameters

Both LCIA and ERA aims at assessing impact on ecosystems, and both tools can provide information related to the quantification of impact on biodiversity due to the presence of toxic substances in the field. Nevertheless, the impact indicator for biodiversity is the fraction of affected species (PAF) for LCIA while ERA can address individual species in the field. At the same time, LCIA can address impacts in soil and water considering intermedia transfer of pollutants, but change in biodiversity occurring in the site itself due to the presence of toxics is not considered. As a contrary, ERA is addressing only (or mainly) impacts occurring in the site itself and not the impacts in the other media.

A second point in biodiversity is the definition of biodiversity addressed. LCA is addressing first population diversity (under the assumption that each population which is not present in the field can recolonized the area as soon as the stressor disappears. ERA can focus on species diversity, considering for example the question of endangered species, or can also use indicators of ecosystems functioning.

As a third important aspect of considering biodiversity, LCA is addressing damage on biodiversity under the assumption of a direct concentration/effect relationship between each substance concentration and a certain level of fraction of diseappeared species. ERA can go far beyond, considering aspects such as trophic chain, foodweb structure, keystones species, etc.

Differences in input data

Fate Modelling

Substances parameters considered in a first tier assessment of EcoRA and a LCA study are mainly Kow, half life of substances, Henry’s constant, solubility, Kds, BAF and BCF. Nevertheless, media parameters such as the fraction of clay, organic matter, the pH, etc can be considered in the EcoRA but are not considered in the generic LCA.

Exposure modelling

Bioavailability for metals needs to be considered in both cases when dealing with site contaminated with metals.

Effect modelling

Until now, first tier studies ERA where mainly using ecotoxicity data based on No Observed (or Lowest observed) Effects Level instead of Effect Concentration levels, while LCA has decided to use EC50s (Effect concentration affecting 50% of the individuals in a test) as data input for impact calculation. Nevertheless, in both cases chronic data from laboratory testing are used in priority.

Differences between data collection for impact assessment

In terms of data collection, the tools also differ considerably. EcoRA is historically based on field observations and field or laboratory experiments. As a difference, LCA is always based on modelling, for quantifying the intermedia transfer, the exposure, and the effect. Furthermore, models used in LCA can not be validated as a whole, it is only possible to validate some part of each model. Nevertheless, for such a purpose, connection with EcoRA works can be of strong help since we should be able to compare the prediction of impact performed with LCA models, with the impact assessment based on EcoRA experiments antd the observations performed in the field.

4 Conclusions:

Possible evolution of LCA and ERA

Historically, even if LCA and ERA have been developed separately, there was always a tendency to become closer and closer as soon as the impact categories addressed were similar. It was also for this reason what impact assessment methods used for LCA were directly derived from ERA methods such as the PNEC method or the HC5 method.

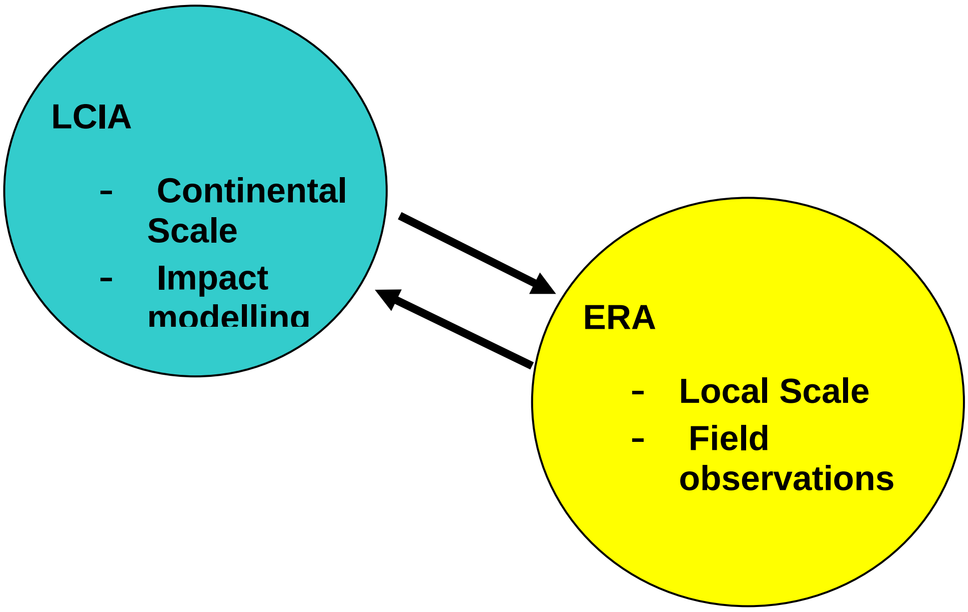

Figure 6: LCIA and ERA can be considered completely different tools

Further these developments, LCA as comparative methods was affirming its specificity developing its own impact indicators, but the tools tend nevertheless to be closer and closer especially concerning the time and space scale addressed.

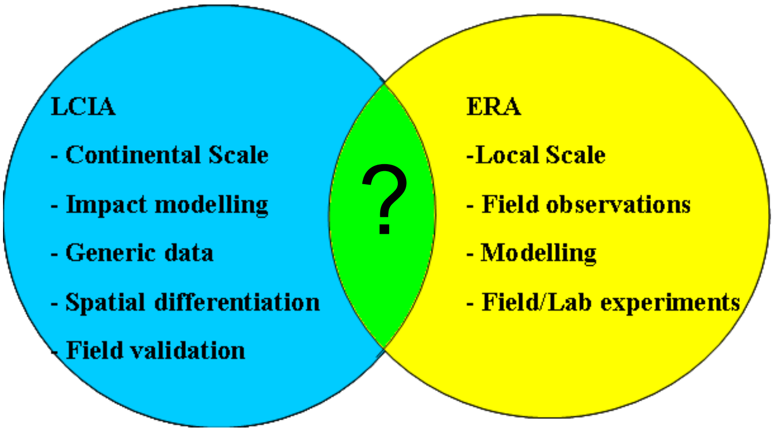

Figure 7: A partial integration of ERA and LCIA

Addressing now partly the same question but with a different view, the question of how far it is possible to integrate the two tools is open. Even if the complementarities of the tools is obvious, it is important to consider the issues that are covered by the two tools on a similar basis in order to ensure we will not have opposite response to the same question.

Ensuring coherence between tools

As highlighted by Udo de Haes, it is possible to ensure the coherence of the methods while considering their specificities at three different levels, the mathematical formula level (use in the impact modelling), the input data and the application (case-study). In the area of site remediation, it is important to bear in mind that LCA is mainly based on models while ERA is based on experimental data (only little modelling is used for intermedia transfer for example). Therefore for impact assessment on ecosystems, we have to ensure the coherence between tools at two levels: (1) the assumptions supporting the models such as the time or spatial scale needs to be coherent with the field observations; and (2) the input data in the model must be coherent with the data observed (or tested) in the field. It is the case for data such as substances concentration, Kd values, Kow, soil composition, biodiversity composition, etc). Considering the coherence at these two levels, it will be possible to ensure the outcomes of the LCA are coherent with the one from ERA. Furthermore, in terms of research perspectives, it would be then possible to make a partial validation of the models, relating for example the predicted change in biodiversity with the one observed in the field, or validating the intermedia transfer of substances.

Perspectives in the improvement of LCIA and ERA

The MuSA project highlights some clear discrepancies between LCA and ERA at the conceptual level and also in terms of input data. Nevertheless, both decision support tools address similar targets and shall give coherent results. The development of regional model in LCIA is a promising perspective. Nevertheless, the uncertainty on the data input in the model are important and measured (or observed) data from the site itself should be compared with model output in order to ensure a reliable modeling at least for the first results obtained with regional models. In terms of perspectives, the priority issues are (1) the variability of the soil composition shall be addressed for developing more accurate soil model and (2) target organisms and biodiversity indicators used in LCA must be consider in parallel with organisms diversity from the site or this area to ensure a coherent assessment at the local, regional and continental scale.

The learning from ERA highlights the need of tiered approach for costly environmental evaluations. For contaminated site management, it is always the case for the site characterisation, and therefore, the tiered approach is now well described in TRIAD for example. LCA is usually seen as a simplified assessment tools which could be considered as a one shot assessment tool. Nevertheless, even in a typical LCA study, 2 step assessments are preferred with a first screening followed by a detailed study. Based on our experience in the comparison between LCA an ERA applied to contaminated site, it seems a third step with a site scale LCA would be relevant for complex systems in order to ensure the coherence between the conclusions of the ERA and the LCA studies. Learnings for Ronde Venen case study highlight that the data collected for a second tier assessment in ERA are rather similar to those required for a site specific LCA, therefore there is no important over cost expected to such an assessment, as soon as the LCIA models enabling site specific assessment for fate, exposure and effect modelling are available. It is therefore a priority to develop such models for LCA in order to support coherent use and partial integration of LCA and ERA. Furthermore, due to their ambiguous status between the ecosphere and the technosphere, and their scientific interest, it seems contaminated site assessment is a good basis for developing case studies on that point.

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Deliverable 3.3:

Metals transfer from soil to water, models and theory

Date of preparation: April 2008

Start date of project: October 10th, 2007 **End date of project: October, 10th 2008**

Report author: Francesca Gambazzi & Jerome Payet

Project coordinator name/author: Jerome Payet

Project coordinator organisation name: SETEMIP-Environnement

Date: Octobre, 22nd, 2008

INDEX

1 Abstract

Metals are often associated with contaminated sites, and one part of the MuSA project was to focus on the modelling of impacts of metals on ecosystems, and initially, the focus was put on the use of the HSAB concept (Hard and Soft Acid and Bases) to model the impact of metals, as what was done for terrestrial ecosystems. Nevertheless, as presented by Helen Beauchamp in the Deliverable 3.1, the MuSA Project considers first substances of concern in the Ronde Venen case study, and the metals that are present in high concentration in this case study are border line metals which can not necessarily be well covered by this concept. Therefore, the focus was put in the review of available models to identify the intermedia transfer and to quantify the mobility of metals in soils especially considering their ability to be applied in LCA. After comaring three models comparing the mobility of Cd, Cu, Pb and Zn, the results are coherent, highlighting the high mobility of Cadmium and Zinc afterward, while Cu and Pb appears to be more sensitive to the media conditions. Four key parameters are identified such as fraction of metals in soil, fraction of organic matter, Redox-potential and pH. This is currently feasible because the four metals here mentioned are very well studied and easy to model. Impact assessment of anions are not feasible using these approaches and it will require further investigations.

2 Introduction

Context

Metals are often associated with contaminated sites, and one part of the MuSA project was to focus on the modelling of impacts of metals on ecosystems, and initially, the focus was put on the use of the HSAB concept (Hard and Soft Acid and Bases) to model the impact of metals, as what was done for terrestrial ecosystems by Haye et al (2006). Nevertheless, as presented by Helen Beauchamp in the Deliverable 3.1, the MuSA Project considers first substances of concern in the Ronde Venen case study, and the metals that are present in high concentration in this case study are border line metals which can not necessarily be well covered by this concept. Therefore, the focus was put in the review of available models to identify the intermedia transfer and to quantify the mobility of metals in soils especially considering their ability to be applied in LCA.

The first part of the deliverable below is describing the framework the model has to fit in. It especially concerns the time and space scale issue and the units and methods that we have to take into account. In a second part, the work compares three models that can be used for assessing mobility of metals and intermedia transfer from soil to water. In a third part, the key aspects needed in building up the models are presented, especially with the description of the main soil parameters influencing the mobility of metals.

Time and space scale in LCA

Assessing the impact of substances involves assessing both their behaviour and interactions in the environment, as well as their evolution in space and during time. Transfer of pollutants in the different environmental compartments (air, water, soil) is a fundamental problem in LCA.

In LCA, the spatial dimension in IMPACT2002+ is represented by a one-box model for all Europe, inserted in a bigger box that stands for the whole planet (referred to as the a-spatial model). The model depicts environmental compartments in interaction with each other (Figure 1). The transfer of a substance from one compartment to another is based on mass balance equations. Improvement to this model in the last years has given new information that allows better predictions of impacts on human health (carcinogenic and non-carcinogenic damages) and aquatic ecotoxicity (referred to as the regionalised model).

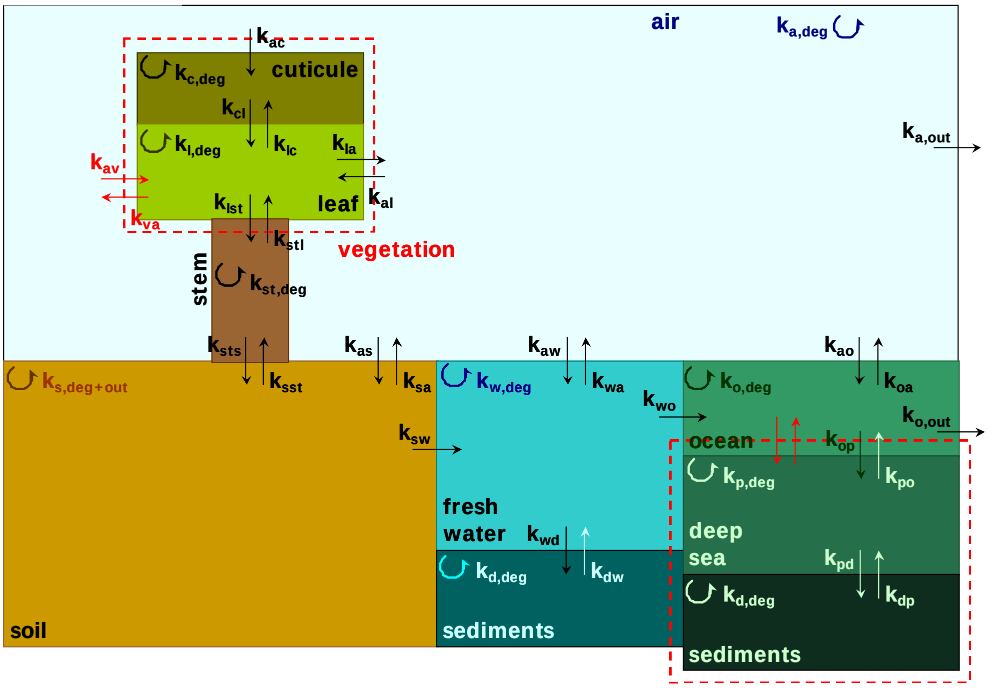

Figure 8 : IMPACT2002 scheme to derive the fate of a substance among the modelled environment compartments (Margni, 2006).

Nowadays, the concept of “global thinking” is more and more present in our society, fact that helps in the understanding of the global spatial distribution of the impact of pollutants. It is possible to understand that something happening in one part of the world can have a concrete influence in other regions, even in remote areas.

The extent of the perception of time is complicated. The impact generated by a substance in 1000 years is not (or very difficultly) “utilizable” as information for the people living now. If this impact of pollution is considered on an infinite scale, it could lack meaning.

One of the aims of this study is to derive a method to evaluate impact on shorter time steps, such as 100 years, which is considered understandable by the human reasoning that makes decisions about pollution management (it can be considered to be the sensitive range of time during which a person feels personally concerned). Another interesting time frame is the millennium, indeed, time horizon between 2 and 10 thousand years can be considered as the installation time of an ecosystem (Jackson, 2008).

Only a few studies treat the problem of quantitative evolution in time of the emission of pollution from the soil. In the first part of this report, these emissions are analysed and compared in order to find significant correlations or discrepancies. A practical example with measured concentrations is also mentioned and analysed more in detail in the second part of the report. In the models presented the considered time lapse is 100 years, which is also the period during which landfill leaching has to be controlled by monitoring. In Doka (2003) two time frames were outlined: short term (< 100 years) and long term (> 100 years). The Ecoinvent dataset follows the same logic, but it limits the long-term landfill emissions at 60 000 years for geological reasons.

This short/long term division is kept in this study, as the focus is on the behaviour of polluted soil. More steps will be evaluated during these two phases as the evolution of the impacts is not necessary linear.

Life Cycle Impact Assessment method: IMPACT

In LCA the IMPACT2002+ dataset covers a wide range of effects on the environment. The midpoint categories are expressed in mass of substance equivalent, which is a reference substance that affects the category with a weighted impact. For example, 1 kg of CH_4 emitted in the atmosphere will correspond to 7 kg of CO_2 for the impact category "climate change". A substance can damage more than one category. For water three endpoint are calculated: ecotoxicity (expressed in kg equivalent of triethylene glycol into water “kg TEG water”), acidification (kgeq SO_2 into air “kg SO_2”) and eutrophication (kg equivalent PO_4^{3-} into a P-limited water “kg PO_4 P-lim”). In addition to this, 9 other impact categories are evaluated, such as emission of carcinogenic substance (in kg C_2H_3Cl equivalent) or global warming impact (kg CO_2 equivalent). Further, the reference-substance equivalent amounts are grouped in endpoint categories, to compare the damage provoked. In the IMPACT2002+ method, the damage categories express the impact on human health, ecosystem quality, climate change and resources.

Table 4 : IMPACT2002+ midpoint categories and standard damage factors

Damage category	**Human Health**	**Unit :DALY**
Carcinogens	2.8E-06	DALY / kg C2H3Cl
Non-Carcinogens	2.8E-06	DALY / kg C2H3Cl
Respiratory inorganics	0.0007	DALY / kg PM2.5
Ionizing radiation	2.1E-10	DALY / Bq C-14
Ozone layer depletion	0.00105	DALY / kg CFC-11
Respiratory organics	2.13E-06	DALY / kg ethylene
Damage category	**Impacts on Water**	**Unit : PDF*m^2*yr**
Aquatic ecotoxicity	5.02E-05	PDF*m^2*yr / kg TEG water
Aquatic acidification	0.065	PDF*m^2*yr / kg SO2
Aquatic eutrophication	3.11E-07	PDF*m^2*yr / kg PO4 P-lim
Damage category	**Climate Change**	**Unit : kg CO2**
Global warming	1	kg CO2 / kg CO2
Damage category	**Resources**	**Unit : MJ primary**
Non-renewable energy	1	MJ primary / MJ primary
Mineral extraction	1	MJ primary / MJ surplus

Impacts on the different categories depend on the amount of the element or substance that will be transferred to water, air or soil. For instance, soil organisms would not be seriously affected by a very volatile compound, as it would almost entirely be transferred to the gaseous phase in a very short time. On the other hand, other compounds can persist in the soil and not be volatilised at all, as is the case for most heavy metals. In this last case the impact on the soil ecosystem would be larger.

In LCA the functional unit is defined as an equivalent service or a product that is compared between multiple options. The comparison allows the discovery of the scenario with the most impact. The problem is: how to estimate the impact of a polluted (or contaminated) site, if no action is taken? Assuming that there is no impact because there is no action (thus no emission) would not be correct. The term contaminated itself implies the presence of substances affecting the environment.

The first part of this project will thus treat the evolution of the emission of substances from soils, trying to give a time scale, which will help describe the evolution of the impact with time. The idea is to look at the contaminated soil as a “pollutant reservoir” releasing pollutants to the surroundings; this approach is chosen following the current LCA way of reasoning. The additional question to solve is the temporal evolution of the emissions.

In the LCA literature several models treating the transfer of heavy metals (but also other pollutants) in soils (and landfills) can be found. The aim of this work is first to give an overview of these models and to search for information to determine metal emissions from soils over a time period of 100 years. Longer term metal behaviour in and emission from soils will be further addressed by considering pedological notions.

3 Examples and Models for Metal Emissions

The problem considered here derives from the question “how to consider and quantify the impact of emitted heavy metals in the long-term time scale?”.

In the Ecoinvent database, short term emissions are evaluated over a period of 100 years, while the long term is 60 000 years. This decision was taken based on Swiss climatic conditions, and supposing that after 60 000 years most of Switzerland will be covered by glaciers of the next ice age, which will thoroughly erode and redistribute matter present in today's surface soil (Doka et al. (c) 2005). In fact, when a glacier moves forward, it perfectly “cleans” the ground, removing the soil with the rocks that are carried at its base (Legros, 2007). This assumption is not valid for Europe as a whole because the extent of the glaciers in the last ice age was not total for the European continent, but this period of 60 000 years will be taken as a reference time, in order to evaluate the evolution of emissions.

Evidently, field data on such long term periods are impossible to collect. Many leaching tests have measured the leachable fraction of the concerned metal, but it must be stressed that such tests give good estimations of the leached fractions at a certain pH, but do not allow the prediction of the intensity and kinetics of metal release in the field (Tack et al., 1999). In addition, for long time periods weathering processes play a role in the chemistry of the soil, and can thus change the soil reactivity. Legislative leaching tests for landfill in Switzerland and in the European Union are performed on fresh, not weathered samples and are therefore merely a test for short-term behaviour of those wastes (Doka (d), 2007).

In the literature, many authors have focused on the fraction of a metal that can be leached from a particular type of soil and on identifying soil parameters that correlate with these fractions. Sauvé et al. (2000) elaborated a mathematical model to estimate the fraction of heavy metals leached from a soil, based on the study of previous models found in the literature (K_d linear coefficient, Freundlich isotherm and the competitive adsorption model). It is concluded that three leading parameters in the calculation of the solid-solution partitioning of metals are: pH, soil organic matter and total metal concentration in soil.

Gray and McLaren (2006) found that the most important factor affecting soluble metal concentrations is the total heavy metal content, whilst soil pH, total carbon content and Fe and Al oxides showed varying degrees of importance depending on the metal.

Thinking about the determination of long-term metal impact, the notion of time has to be integrated in these models. In fact, in laboratory experiments it is often assumed that the soil would reach a precise pH value within an estimated time frame due to local specific conditions. The evaluation of leaching consists of bringing the system to the foreseen pH, which normally corresponds to the worst case scenario for a given situation.

In this work a comparison between models described by different authors, treating emissions of the metals of interest in this study (cadmium, copper, lead, zinc) from the soil to the aquatic compartment. The vector of the movement is water; this is why the term leaching can be used. Emission would otherwise be a more general term to describe the lost of metal from a soil.

Four studies are illustrated:

- Ronde Venen measurements by Alterra (2000)
- ROAD-RES model done by Birgisdóttir et al (2007)
- Landfill model done by Obersteiner et al. (2007) based on empirical data
- Ecoinvent Waste treatment and Assessment of Long-term emissions by Doka (2005)

The comparison between these four cases is not totally rigorous because of the nature of the data presented. Absolute soil values are not always available. The aim here is to look for a trend that dominates the leaching (or emission) process of metals to the aquatic compartment. Where possible, the leaching ratio (concentration (or mass) present in the solid phase / concentration (or mass) leached after 100 years) is given, in order to have a visual support in finding the eventual trends.

Ronde Venen, Demmerik polder

The “toemaakdek” activities have been practiced for centuries in Dutch polders, and for this reason the treatment of data requires attention. Total concentration of metals in the soil and concentration in the pore water were measured by Alterra (Alterra, 2000). To evaluate the relation between the total concentration of each metal and its annual leaching, the pore water concentration [mg/l] was multiplied by the total amount of water flowing through the soil every year [l/yr]. The derived leaching [mg/l] is compared with the total metal content in figure 2.

The conditions used to describe the polder in the mathematical model are as follows: it has a surface of 1625 square metres, a polluted layer 10 centimetres deep, and an annual net infiltration in the soil[1] of 250 mm/m^2. The concentrations of metals are presented in table 2.

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[1] The net infiltration is the fraction of rainfall that contributes to the groundwater table.

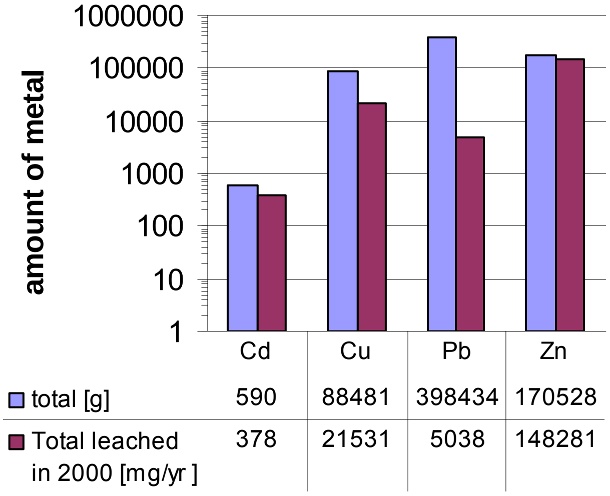


Figure 9 : Metal content [g] and metal leached in 2003 [g/yr] from the Demmerik polder, in Ronde Venen site (Rutgers, in press)

The comparison is between the total metal content of the site and the quantity of metal that has been leached during the same year. It shows that, compared to the other metals, lead is leached in very small amounts despite its high presence in the soil.

Looking at the results of the extraction by 0.01 M $CaCl_2$ in Table 2 (RIVM, 2000), the tendency of cadmium to be extractable is evident. In fact, the fraction released was about 8 % for this element, while for copper and it was under 0.6%. The value for zinc is 2.34 %, which is also high compared to lead and copper.

Table 5 : Metal concentration, pore water concentration, 0.01 M CaCl2 extraction and CaCl2 extractable fraction. Demmerik polder, Ronde Venen site (data by Alterra, 2003)

	Total destruction average [mg/kg]	Porewater concentration [mg/l]	$CaCl_2$ extraction [mg/kg]	Extractable fraction [%]
Cd	1.09	0.00093	0.09	8.26
Cu	165	0.053	0.844	0.51
Pb	743	0.0124	1.653	0.22
Zn	318	0.365	7.441	2.34

The available data show the important relative leaching of cadmium and then zinc, and the resistance to this same process of lead and copper.

ROAD-RES Model

Birgisdóttir et al. (2007) described two disposal methods for 4400 tons of Municipal Solid Waste Incinerator (MSWI) bottom ashes in a life cycle assessment: the first one describes the landfilling of bottom ashes in a coastal landfill in Denmark, and the second one studies their recycling as subbase layer in an asphalt secondary road (the amount corresponds to the quantity of bottom ashes needed as subbase material beneath the lane of 1 kilometre of Danish traditional secondary road). Leaching phenomena are described as being controlled by two main factors: pH and redox potential. The differences between the starting concentrations of heavy metals in road water and in road sections with MSWI bottom ashes or with crushed rock as granular base were trivial, except for lead (Birgisdóttir et al. 2007). The functional unit is one kilometre of Danish secondary road.

To model the landfill, four time periods were considered: the first three during which the leachate of the landfill goes to the waste water treatment plant. It was assumed that on average 20% of the leachate would reach the environment due to defects in the impermeable liner layer at the bottom of the landfill and collection system. Leaching of heavy metals in the road scenario, during 100 years, represents less than 1% of the total amount in the bottom ashes (table 3).

Table 6 : ROAD-RES study, fraction of metals leached from landfill or road after 100 years (Birgisdóttir et al., 2007)

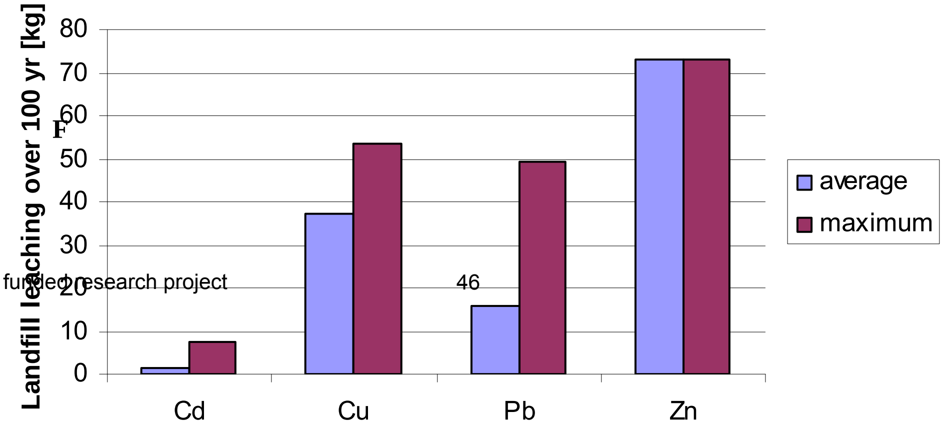
Element	Landfill %	Road %
Cd	0.0240	0.2000
Cu	0.0118	0.0314
Pb	0.0005	0.0045
Zn	0.0006	0.0049

The road material is more sensitive to the leaching of the metals, present as bottom ashes in the road structure. Especially for cadmium, the amount leached from the road is much more important than from the landfill; it is about 10 times bigger. Proportionally, cadmium is leached more easily in road scenario, while in the landfill copper is the metal most sensitive to be leached, even if its leached fraction is very small.

It is important to notice that all the values are equal or under 0.2 %, which means that after 100 years, a very small amount of metals could be released from the stabilisation that occurs in the creation of bottom ashes and in the inclusion of metals in the road subbase material.

Landfill model

Obersteiner et al. (2003) compare data of different kinds of landfills to show the evolution of leachates over a time period of 100 years. Three main phases are distinguished: from 1 to 5 years (called "operational period", supposed to have the highest values), 6-30 years ("aftercare period") and 31-100 years (medium time period, where active aftercare has stopped). Unfortunately, these results depend on the waste composition that is not given in the article. The Figure 3 shows absolute amount of leached metal (average and maximum) for sanitary landfills for the averaged data from different countries (Austria, Germany and Switzerland). Figure 4 weights the results with the fraction of waste composed by each specific metal.



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e 10 : Sum of leachate after 100 years. Obersteiner, 2007

Zinc shows only a very small difference between average and maximum leaching. It can thus be deduced that this metal is weakly dependent on local landfill conditions. At the opposite, lead, cadmium and copper seem to vary more between the landfills, as the maximum values are about 1.5 times higher than the averaged ones (extreme case for cadmium, where the maximum is 4.7 times higher).

Supposing that Germany, Austria and Switzerland domestic wastes have similar composition, and taking limit values for waste composition in sanitary landfill in Switzerland (OTD, Swiss waste ordinance, 1991) it is possible to normalise the plot.

The limit for zinc is 1000 mg/kg, while lead and copper have a limit of 500 mg/kg (the half) and 10 mg/kg for cadmium.

It means that if the landfill is at limit values of composition, the amount of cadmium is 100 times less than the amount of lead. With this hypothesis, the weighted data were plotted. Taking Zinc as a relative reference, Cd was multiplied by 100, Cu and Pb by 2 in order to have a relative “Swiss” standard waste composition.

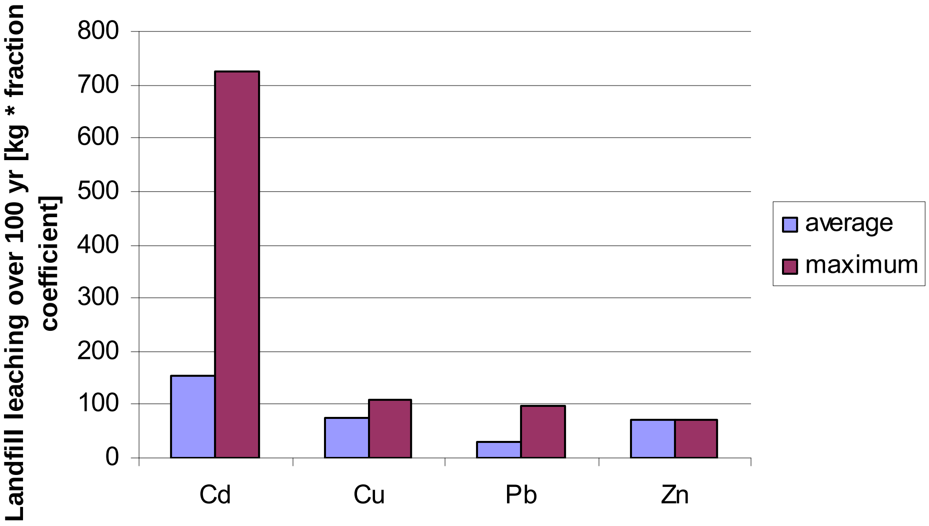

Figure 11 : Weighted sanitary landfill emissions.

The results underline the variability of cadmium leaching (notice the important difference between average and maximum leaching amounts), and also its higher proportional capacity to be emitted by the sanitary landfill compared to the other three metals. Lead seems to be the metal that proportionally remains more in the soil in long-term periods, but the difference between average value and the maximum is still considerable.

Ecoinvent waste treatment and metal emissions

Doka ((a) 2003, (b) 2004) elaborated the system used to determine transfer coefficients (TK) in the Ecoinvent database for the emission of several waste types from different landfills. Until 1996 the modelling was based on thermodynamics and laboratory leaching tests (with some refinements). The waste material was placed during 24 hours at pH 4, and the total leached amount was assumed to represent the long term available fraction. In Ecoinvent 1996, 50% of the total amount was used for landfills with little acidification potential and 100% for landfills with large acidification potential (sanitary landfills) (Doka, 2004).

Emissions from landfills used for calculations in SimaPRo (the software used to calculate impacts in LCA) are split up into emissions that occur in the first 100 years after waste deposition (short term transfer coefficients) and emissions that occur afterwards (long term transfer coefficients, until 60 000 years).

The carbonate phase represents the period during which the pH is buffered by acid neutralising materials present in the landfill. Doka uses a landfill-waste specific transfer coefficient to calculate the emissions.

$$Emission\,media_{phase,i}[kg\;i/kg\,waste]=TK\;media_{phase,i}\cdot waste\;composition_i[kg\,i/kg\,waste]$$

The emissions from the waste to specific media are calculated from the transfer coefficient (TK) for the element *i*, for the concerned media (water or gas) and the landfill phase (short-term or long-term), multiplied by the concentration of the element in the waste.

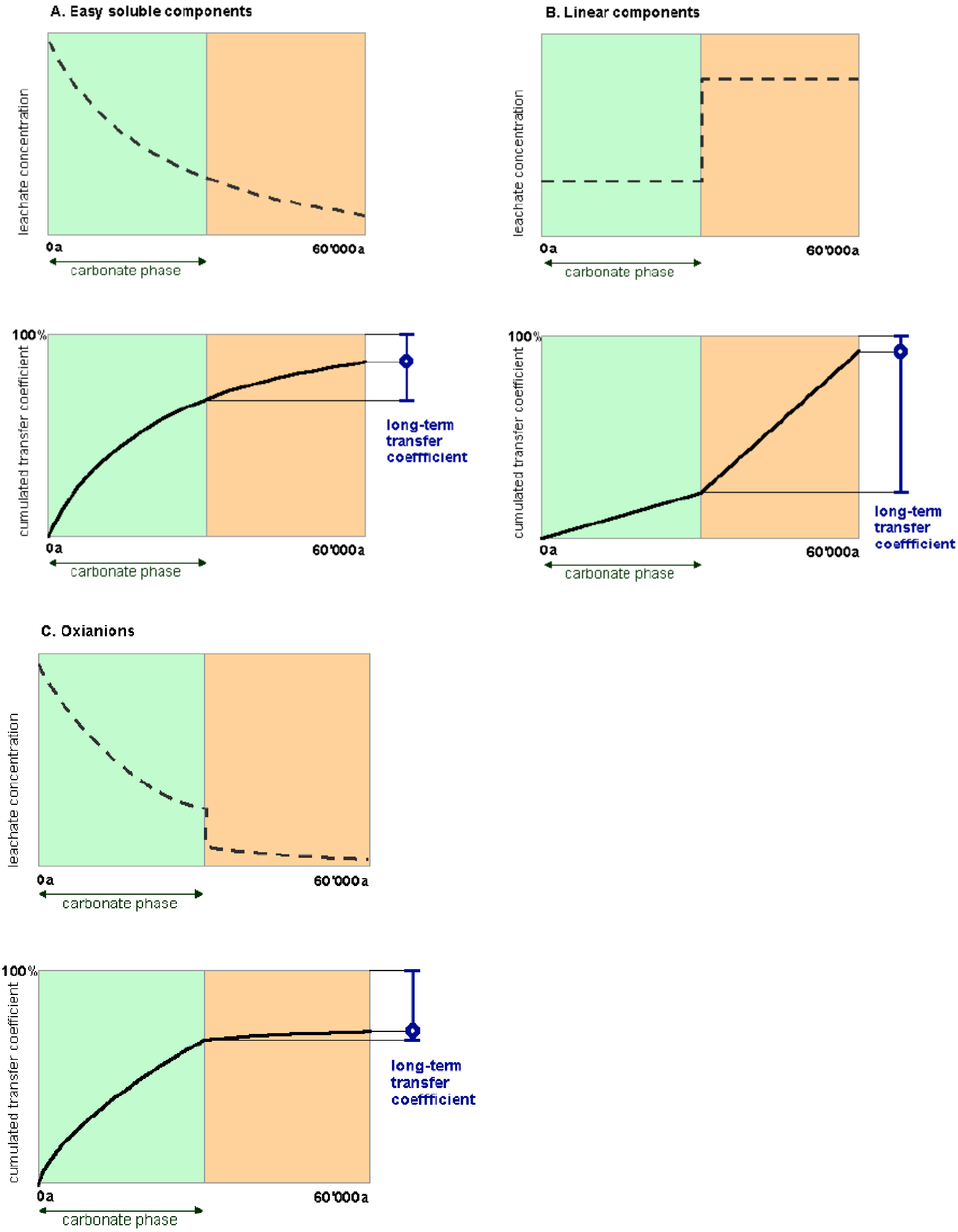

Figure 12 : Behaviour of Easily soluble components, Linear components and Oxyanions by Doka, 2003. Calculations scheme for long-term transfer coefficients for different elements: A.) Exponential decline for soluble elements with no change after the carbonate phase (left), B.) Constant concentrations for metal, Exponential decline of concentrations for metal oxianions with a decrease after the carbonate phase (right). The range indicated at the right side of each panel represents the derived uncertainty range for transfer coefficients

In the Doka model easily soluble elements (monovalent ions Na+, K+, Cl-, F-, Br-, I-) and oxyanion-forming elements (WO4^{2-}, HVO4^{2-}, Cr(VI)O4^{2-}, MoO4^{2-}, HBO3^{2-}-, HSeO4^{-}, SbO4^{3-}, HAsO4^{2-}) and also nitrogen (as nitrate NO_3^-) are supposed to decrease qualitatively in the same way (figure 5)

To have a general idea of the leaching of the four metals studied in this report, an extract from Appendix I is summarised in table 4.

Table 7 : Long term-transfer coefficient (Doka, 2007 (a)). Fraction of metal that will be leached after 60 000 years

Long term-transfer coefficient (%)						
	Sanitary Landfill		Residual Landfill		Slag compartment	
	Short term*	Long term	Short term	Long term	Short term	Long term
Cd	11.89	100	0.001133	0.68	0.0514	100
Cu	0.24	100	0.006426	3.86	0.003472	100
Pb	0.34	100	0.000866	0.52	0.001825	100
Zn	1.75	83	0.002046	1.23	0.002977	100

* The short term emission of a sanitary landfill depends strictly on the length of the carbonate phase, which keeps the pH in neutral-high values

Sanitary landfills are supposed to receive “all” kinds of waste (untreated municipal or household waste, building waste or wastewater treatment sludge)[2]. Residual landfill has low-carbon wastes, like incineration residues or inorganic building materials. In Switzerland the total organic carbon in residual, inert and slag compartment landfill has to be lower than 5 % weight (3 % weight for slag compartments).

Residual landfills receive wastes that are not chemically or biologically reactive in water. They mainly receive MSWI filter ashes solidified with cement. This composition explains the huge difference between with the amount of leached metals of the sanitary landfill and the slag compartment.

In residual landfills the metal which undergoes the greatest (but still very limited) leaching is copper, with about the 4 % of total concentration. Zinc follows with 1.23% , while cadmium and lead leaching are under the 1%. Globally these values are extremely low. This is an expected result, since the residual material landfill has a large carbonate buffer, which is not depleted within 60'000 years, and also it bears waste with lower carbon content than sanitary or slag landfills and hence there is less potential for biogenic activity (Doka (d), 2007).

Conclusions on the quoted models

All examples were taken from European studies about long term metal leaching. As climate (and especially annual rainfall) determines the amount of rain water that flows (or could flow) through the soil, the specific location of the site of study has to be taken into account.

In the Dutch Ronde Venen practical case study, an important leaching of cadmium and zinc was found. The difficulty to define an age for the site makes locating these emissions in the time scale difficult.

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[2] However, since 2000, it is denied to landfill burnable waste in Switzerland.

In the model elaborated by Birgisdóttir in Denmark, after 100 years the leaching of cadmium from the road material was significantly higher compared to the other metals, while for landfill copper was the dominant compound in leachates. Generally the emissions of metals were very low, under 0.2% of the total content for each metal.

Obersteiner et al. worked on data of sanitary landfills from Germany, Austria and Switzerland. Their results showed cadmium as the most leached metal compared to the others, but also its variability. Lead seemed to be the metal that proportionally remains more in the soil in long term periods, but the difference between average and the maximum value is considerable. The data are presented as absolute amounts, which do not allow a direct comparison with the Doka model.

The Ecoinvent model (based on Swiss data) presented by Doka also confirms cadmium leachability for short term periods in slag compartments, which proportionally is much higher than the fractions found for the other elements. The residual landfill is the only one that retains a large amount of metals, even after 60 000 years (the period after which sanitary and slag compartments have released the entire amounts of metals, except for zinc). If enough time is given, all phases could be weathered and turned into available phases. It would thus means that there is no stop to leaching.

No obvious trend has been found in the review of these studies, although the magnitudes found can help to assess the possible evolution of emissions from a specific site, and consequently to estimate its impact on the environment. Qualitatively it can be said that cadmium has the tendency to be proportionally leached more than the other three considered metals. Lead is apparently well retained by the soil matrix for short term periods in all the models and analyses, while zinc and copper do not show a clear pattern of behaviour.

Although the situations presented are very different in context and soil composition, all authors put an accent on the importance of the acidity neutralisation capacity, which stabilises the pH, and indirectly reduces the leaching of most substances.

These factors are consequently going to be the basic parameters that are going to be used to estimate the leaching behaviour of metals for long term periods in the next section.

4 Modelling Emission Evolution

In the following paragraphs, the approach to qualitatively model the leaching of metals is explained. A graphical summary of the model of Doka ((b), 2004) is shown in figure 6. In this figure the different evolution of transfer coefficients for different elements is represented. Two main points are underlined: 1) the importance of the drop of pH on the emission of elements, especially for metals, and 2) the low value of the transfer coefficients for short-time periods compared to the long term.

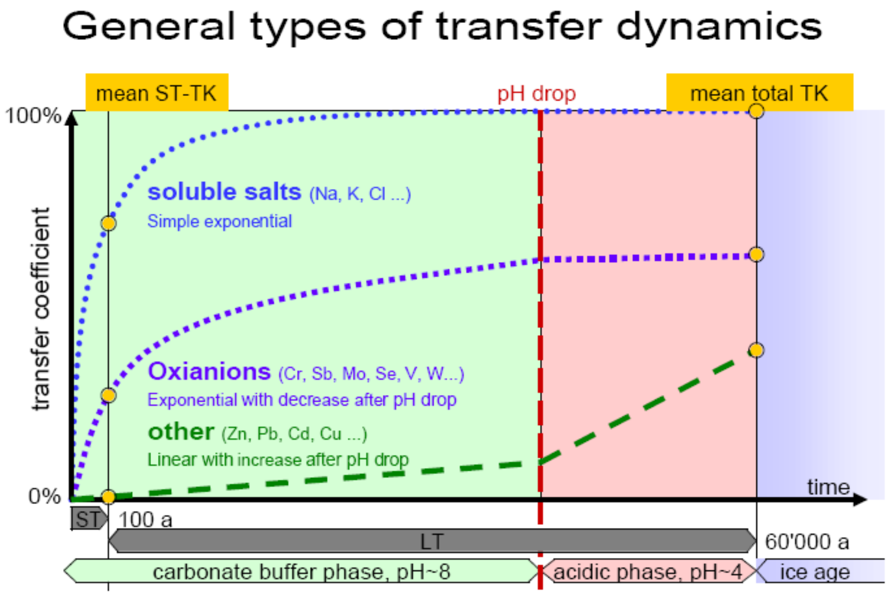

Figure 13 : Evolution of transfer coefficients (TK) used in SimaPRo and proposed by Doka for different pollutant species. Transfer dynamics, Doka Life Cycle Assessment, Zurich, 2004

If the releasing of the element in the environment means that it has a greater “pollutant potential” (for instance it is more bioavailable, more mobile, etc) this model lets be assumed that most impacts generated by these species will appear in long time periods. In fact, only about 20% to 40% of the metals reach the groundwater directly through flow in continuous macro pores in the first decades (3-25 years). The remaining 60%-80% are retained more strongly by the soil matrix and their reach the water table in 400 to 3000 years for Cd^{2+}, Ni^{2+} , Zn^{2+} , Cu^{2+} and Cr^{3+} , while 5000 to 33 000 years are necessary for Pb^{2+} and Hg^{2+} . (Doka, 2007).

Basic ideas

To evaluate the environmental impact of the four metals it is not sufficient to determine their total content: their behaviour in a given soil fraction and their mobilisation capacity are both important (Alvarenga et al., 2007). Metal leaching and, more generally, emissions from soil are determined by the nature of the linkage between the metal ion and the soil form, the transformation that it can undergo and the evolution of the soil itself. In addition to physico-chemical factors, biology also plays a role in the soil metal dynamics. As stated by Gobat et al. (2003) microorganisms are key actors in:

- transformation of vegetable and animal debris
- oxidation, reduction , precipitation and dissolution of mineral ions
- fixation of molecular nitrogen
- control of bioelement cycles, in particular carbon, oxygen, iron nitrogen and sulphur cycles
- transformation of parent material

Abiotic and biotic processes in the soil are strictly linked. Microorganisms and vegetation influence the soil conditions, for example soil pH. The composition of litter depends on the type of vegetation that is present on the site (that depends on the climate, soil type, etc.). Soils are at the interface between atmosphere, biosphere, hydrosphere and lithosphere, which makes them difficult systems to describe. In the soil quasi-instantaneous processes, such as the capture of an iron atom by an organic molecule, coexist with much slower movements like progressive clay migration towards the deeper soil horizon and the consequent accumulation in horizon B (Gobat et al., 2003).

In the following description, focus will be on the lithosphere and its evolution, as well as on its interaction with the biosphere. The atmospheric and the hydrological aspects will be considered later, as “external” parameters in soil evolution.

Describing the soil which interacts with metals

First of all, a general description of the main constituents of the soil is presented, followed by the method used to analyse the metal bonds in the soils. As the metals can bind to several different soil components, the evolution of each will have consequences on the fate of the others. This fact will be at the base of the creation of a model to predict metal emission in time.

4.1.1 Mineral constituents

The soil minerals are primary, inherited directly from the parent rock or secondary, created by the chemical transformation and found in the weathering complex. This complex is composed by salts (e.g. calcium or magnesium carbonate) or silicates (e.g. micas and clays). Clays behave as colloids, as iron or aluminium hydroxides and other secondary minerals. If the weathering is "total", it releases isolated ions but also micromolecules (Gobat et al., 2003). Physical disintegration and biogeochemical weathering produce a mix of constituents of very different size and mineralogy quality.

Climatic agents, such as wind, frost and water, break the rock down in smaller and smaller pieces. They physically degrade the soil but maintain the same chemical composition.

Biogeochemical weathering of rock involves water with oxygen, carbon dioxide or organic acids, and acts following five paths: hydratation, dissolution, oxidation, reduction and hydrolysis.

Hydratation first weathers the ferruginous rock, entering water molecules modifying the mineralogy, as for instance, hematite Fe_2O_3 hydrates to form goethite FeOOH.

Dissolution is present in alkaline environments, chelation and acid dissolution. An example of acid dissolution is the case of calcite, dissolved by carbonic acid H_2CO_3 .

Oxidation allows the liberation of Fe^{3+} from the Fe^{2+} present in the crystal lattice of some silicates.

Reduction dissolves Fe^{2+} from the oxides and hydroxides of Fe^{3+} found for example in cements of some sandstone. It takes place in poorly aerated environments.

Hydrolysis engenders important changes in crystal lattice. This process depends on the climatic conditions, and it increases with high temperature and humidity. It concerns practically all silicates and is often accelerated by microbial activity (Gobat et al., 2003).

4.1.2 Soil organic matter and organisms

Soil organic matter can be divided in two main groups: the first is inherited directly from the organic residues (litter) and the second are synthesised in the soil under humidification process. The first group is mainly composed of cellulose, lignin, proteins and lipids. In the second there are aromatic compounds with different levels of polycondensation, such as the highly stable macromolecules forming a part of humins. The limit between the two is difficult to locate, in particular for humins, which can be inherited from vegetable material or newly formed.

For these processes the time needed is extremely different. If on the one hand the exchangeable and the carbonate fraction can be degraded in relatively short time frames (less than 100 years), the complete (bio)degradation of organic matter is an extremely long process, which is influenced by "internal" (microbial activity) and "external" (atmospheric deposition) factors.

In the literature (Owen, 2001) half lives for fulvic acid and humic acids in an organic rich soil are estimate at 10-15 years and at more than 100 years respectively. Following Legros (2007) half lives of fulvic and humic acids are between 200 and 1000 years, while for formed humins it is more than 2000 years (if no changes in soil conditions and climate occur).

The organic matter in soils evolutes with acidification, changes in hydrology and changing in land use (Salomons et al., 1995). These changes are reflected in higher dissolved organic matter content and increased leaching of dissolved organic matter to surface waters, which are positively correlated to the leachable amount of metal (Sauvé et al., 2000).

Although organic matter is able to retain chemicals in soils and sediments, its breakdown products are able to mobilise heavy metals and organic micro pollutants in particular (Salomons et al., 1995). This is due to the solubility of humic, but especially fulvic acids, which can chelate metals.

Metals can be bound or complexed with organic matter. It can thus immobilise metals in the solid phase or increase it, due to the presence of dissolved organic matter. In fact, dissolved organic matter has been shown to reduce metal sorption onto soil surfaces by competing for free metals and forming soluble organo-complexes or being preferentially sorbed onto surfaces instead of the metals it is competing with (Gray and McLaren, 2006).

Bacteria are an important organic component in structuring the soil. In fact, they synthesise some molecules, especially polysaccharides particularly resistant to enzymatic degradation, which contribute to the pool of organic matter in soils. The microbial humins (so these products are called) participate to the formation of microaggregates (Gobat et al., 2003).

The effects due to the soil microflora are mainly due to bacterial functions. They include mineralization of organic matter, oxidation of inorganic reduced molecules, anaerobic reduction of inorganic oxidised molecules, dissolving or precipitation of minerals, without forgetting the transformation for certain organic compounds to humins.

Soil fauna also plays an important role. For instance, macroarthropodes of temperate litter, termites in tropical soils, microarthropodes in peat lands, earthworm in mosts soils. Every one of these actors is involved with the transfer of matter and elements in the soil (Gobat et al., 2003).

The presence of vegetation cover and earthworms has a big impact on the macro-porosity of the soil upper layer (Hellweg et al., 2005). In addition, they play a key role in the bioturbation phenomenon, which allows the transfer of organic matter in the verticality of the soil.

4.1.3 Redox potential

Redox potential changes the valence of the metals. Under oxidising and slightly reducing conditions, the solubility of heavy metal cations decreases due to the precipitation of oxides, hydroxides or carbonate and only for neutral to mildly alkaline pH values, while lowering the redox potential has the effect to increase metal cation solubility (Legros, 2007). If reducing conditions are present (no oxygen available, long submersion time), leaching of metals is thus more important. In aquatic environments metal cations tend to desorb from solids as pH decreases. As pH increases, they adsorb or precipitate, but only up to a certain limit, after which they are again solubilised due to the negative charges present in the solution (Salomons, 1995).

Oxygen availability in soils determines the mobility of contaminants but also the presence or absence of iron and manganese(hydro)oxides. Redox potential variations change the valence of metals (Fe, Mn, As, etc.); under reducing conditions anions are more soluble, increased soluble concentrations of these metals have been observed in reducing environments such as ground waters and porewater of sediments. Indirectly, under reducing conditions sulphate reduction will take place and metal sulphides become a stable phase. Metal sulphides have an extremely low solubility and concentrations of metals in porewater of sediments (where the reducing conditions are normally found) are generally low. On the other hand, an increase in the redox potential induces metal sulphide to become unstable and thus dissolved concentration of the metal will increase. This abrupt change happens for instance when polluted (anoxic) sediments are exposed to oxidising conditions. The speciation of iron can be an important parameter. In fact, with the presence of sulphide (estuarine and marine environments), the iron hydroxides become unstable and are converted to iron oxides, which have a lower adsorbing capacity and the net result is a loss of adsorbing capacity from the system (Salomons et al., 1995).

Inundation and acidification also causes hydroxides to become unstable, when these phenomena occur the metals associated with iron and manganese hydroxides are mobilised (Salomons et al., 1995).

4.1.4 pH

Parallel to redox conditions, pH has to be considered. The retention strength of the ionic fixation depends on the nature of the element: atomic radius, valence (monovalent atoms are less retained than bivalent ones), hydratation degree and pH are the determining factors (Gobat et al., 2003). The metal species that can exist in an environment are basically determined by these two factors, and shown in Eh-pH diagrams (figure 7) (Salomons et al., 1995). When pH drops (i.e. at the end of the carbonate phase), the metal cation concentration in leachate increases, while the metal oxyanions decrease as they are less soluble at low pH.

If pH drops (acidification) the release of heavy metals will occur more quickly. In actual cases, pH drops when the acidity neutralisation capacity (due especially to carbonates and organic matter) of the soil is consumed, this means that organic matter has been practically totally degraded (Legros, 2007). Oxyanions behave oppositely: they tend to adsorb and/or precipitate more as pH decreases (Salomons, 1995).

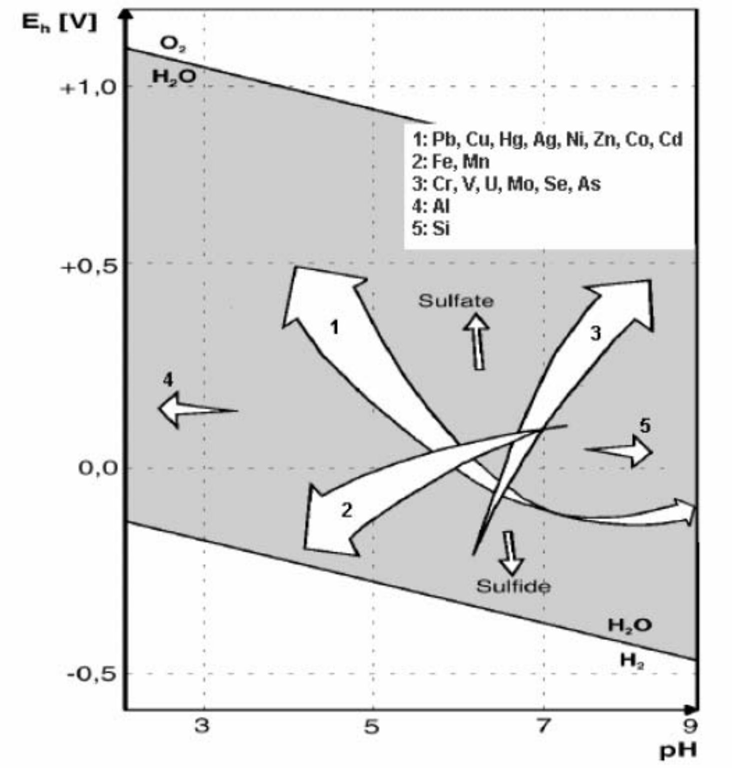

Figure 14 : Diagram of pH versus redox potential Eh and relative metal mobility. Arrows indicate direction of increasing relative mobility for the corresponding metals. (Doka, 2003, adapted from Förstner et al. 1989)

The retention strength of the ionic fixation depends on the nature of the element: atomic radius, valence (monovalent atoms are less retained than bivalent ones), hydratation degree and pH are the determining factors (Gobat et al., 2003). For a given pH, small radius and hydratation degree and high valence are the conditions to assure a stronger bond.

4.1.5 Climate

Climate (wind, rainfall, solar irradiation) plays a fundamental role in organic matter degradation, especially through the oxidation of organic matter. Higher temperatures and longer dry periods dramatically increase this phenomenon. Metals bound to iron and manganese oxides and organic matter, present stronger bonds than organic matter chelating metals. The degradation of organic matter destroys their bonds with the metals, increasing their release. Rainfalls have to be considered from two points of view: the hydrological one, which means the amount, length and frequency of rainfalls during the year, and the chemical one, which considers the composition of the water (for example, acid rains).

Temperature influences the weathering of the soil. With an increase of 10 celsius degrees, the rate of chemical reaction is expected to increase with a factor of two (Legros, 2007).

Seasonal contrasts in climatological conditions act on biological activity: organic matter is more easily mineralised. For example, there is less peat in the Alps than in the Massif Central due to the more contrasted climate in the Alps (Legros, 2007). Aridity helps speed organic matter degradation via oxidation, which brings to an increased sensitivity to erosion.

Modelling evolution emission of metals from soils

For long term predictions it is therefore not only necessary to consider variables like pH, competing ions, dissolved organic matter etc. that determine the initial adsorption, but even more so to consider the stability of the adsorbing phases which compose the soil.

Various chemical extraction procedures provide information about the reactivity of different metal pools in the soil (Pickering, 1981). Each pool thus has a different behaviour depending on chemical and physical parameters linked to the condition of the soil at a specific moment. Sequential extraction analyses give the partitioning of a specific metal among the soil; five fractions are defines by Tessier et al. (1979): exchangeable, bound to carbonates, bound to Mn/Fe oxides, bound to organic matter and residual.

Exchangeable and carbonate bound fractions are the most sensitive to natural degradation, and the metals bound to them are the most mobile. Once these fractions are not present in the soil anymore, metals that were bound to organic matter will be more and more present in the leachate, because of the degradation of the organic fraction of soil. This phenomenon is followed by the alteration of mineral oxides that consequently can liberate the metals bound to them. Iron and manganese oxides are important sorbents of trace metals (Sauvé et al., 2000). Both organic matter and oxide degradation strictly depend on external factors, such as temperature, annual rainfall amount and intensity, length of dry periods, etcetera. The residual fraction is also supposed to be released or degraded, for extremely long time periods. In this case, it is supposed to occur only after 60 000 years for cold regions subjected to the glacier expansion in the next ice age.

In general, with exception of selenium and molybdenum, metals are more soluble in soils at low pH due to the dissolution of carbonates, phosphates and other solid phases (Adriano, 2001). In the dynamics of metal leaching, pH is the most influencing factor (Sauvé, 2001). At low pH the sorption bonds of metals to adsorption sites on mineral surfaces is weakened and cation exchange capacity of organic matter is lower (Adriano, 2001).

The acid neutralisation capacity (ANC) of a soil is important to assess the sensitivity of the material to external influences, and crucial to address long term impact and stability of the components of a soil (Van der Sloot, 2001). With time, progressive acidification happens to most soils in humid and draining situations (Legros, 2007). This is due to the protons produced by organic matter and to the dissociation in the water of CO_2 of the soil. Minerals are destroyed and their bases are leached and drained. Van Breemen et al. (1983) defined acidification as following:

$$\begin{aligned}\text{ANC} = {} & 6\,[Al_2O_3] + 2\,[CaO] + 2\,[MgO] + 2\,[K_2O] + 2\,[Na_2O] \\ & + 2\,[MnO] + 2\,[FeO] - 2\,[SO_3] - 2\,[P_2O_5] - [HCl]\end{aligned}$$

[] represent concentration in $mol.m^{-3}$.

Gobat et al. (2003) describe the buffering capacity of the soils, and the responsible species, depending on the pH. The summary is presented in table 5.

Table 8 : Soil buffering capacity (Gobat et al., 2003)

Soil mineral buffering specie	pH range of buffering capacity of the specie
Carbonates	8.6 – 6.2
Silicates	6.2 – 5.0
Clays	5 – 4.2
Aluminium hydroxides	4.2 – 2.8
Iron hydroxides	< 3.2

Under humid conditions soils have a natural tendency to lose their cations and thus acidify. For municipal solid waste incineration slag the dominant loss of acid neutralising capacity is not due to protons of acid rain or acid producing internal reaction, but from the wash-out of buffering carbonate minerals. Thermodynamic calculations mentioned in the article by Doka ((d) 2007) indicate that for a normal soil under a CO2 partial pressure of 10^-2 atm (10 000 ppm) the carbonate washout and the carbonate buffer phase ends after 17’000 years. A study conducted on Hawaii Islands by Chadwick and Chorover (2001) show the decreasing curve of neutralisation capacity on the same basalt rock (average temperature 16°, annual rainfall 2500 mm). It reaches zero after about 10 000 years, but already after 1000 years it is under 100 mol/m^3.

The model elaborated in this study is based on four time frames of different lengths:

- The first period, during which the easily mobile metals are leached out of the soil basically due to convection movement, and the carbonates are dissolved in the water passing through the system. This leaching is very important especially for “young” pollution, which has not had the time to fix to the soil and thus is more mobile. This period ends with a pH value around 6 (see table 5). Heavy metals like Cd, Zn, Pb Cu are washed out 3 to 5 times faster after the carbonate buffer phase, due to the lowering of the pH.
- The second phase concerns organic matter, which is degraded by the decreasing pH which attacks the stable bonds. Vegetation cover and “external” factors, such as rains and climate, play an important role in the destabilisation of organic matter.
- The third phase sees (hydro)oxides as main actors. The pH has lowered, microbial activity is reduced and organic matter is no longer present. Physical disintegration and biogeochemical weathering have a stronger impact, as the soil is “naked”.
- The fourth and last period concerns the fraction of the soil where the metals are more strongly bound. The residual fraction contains the metals that are more strongly bound to the soil matrix, which normally correspond to clays, stabilised materials and rocks containing trace metals. The metal residual fraction is extremely stable in the soil. It corresponds to metals absorbed in clays or in minerals structure and it is very slightly sensitive to weathering (Legros, 2007). For the Swiss case, the metals are supposed to be emitted in the next ice age as hypothesised by Doka ((a) 2003) and confirmed by the pedologic observation of Legros (2007). For the zones of Europe which are not supposed to be on glacier pathways, further studies have to be done, to evaluate the kind of soil that could be present following local data.

The length of each period varies importantly, depending on the leading factors, which characterise the leaching, and therefore stem from the location and situation of the site.

For the total degradation of organic matter and iron and manganese oxides practically no data from the literature are available. Most soils rich in organic matter have low pH and are prone to reduction when moisture increases. Present oxides are easily depleted by reductive dissolution enhanced by the presence of the high dissolved organic carbon concentration (Salomons, 1995).

As first assumption the length of this period is thus (roughly) approximate: after 10 000 years these two species are supposed to have completely degraded. In nature reality, these phenomena co-exist and take place at the same time, but with different rates.

After 60 000 years the whole soil is supposed to be degraded: 100 % of the metals have been liberated at that time.

Linear trends are hypothesised within the different phases. Figure 8 illustrates the trend that would have a specific metal emission from a soil during long term periods.

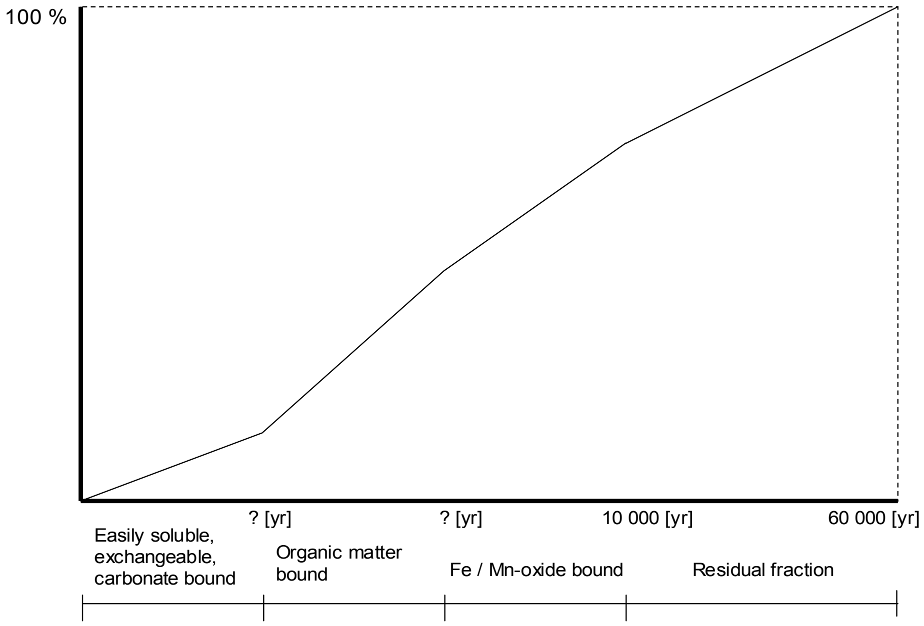

Figure 15 : General trend of metal emission from a soil during long term period

The shape of the curve varies depending on the soil conditions, as explained in the previous paragraphs and on local conditions. Depending on the nature of the metal and on its relative concentration, heavy metal cation solubility can vary from being entirely controlled by its direct interaction with an inorganic surface (cation-like) to being completely subjected to the sorption and solubility pattern of the organic matter (anion-like) (Salomons, 1995). The slopes of the segments would vary depending on the specific case.

Variation of soil pH and redox potential under natural conditions is due to several processes that can vary during time. pH is defined by the proton balance of the aquatic or terrestrial ecosystem. Atmospheric deposition and oxidation reactions (such as nitrification, sulphide oxidation, ferrous iron oxidation) supply protons, while weathering of carbonate and silicates minerals, the ion exchange and the reduction reactions (e.g. denitrification, sulphate reduction, iron oxide reduction) consume them. Redox conditions are influenced by the hydrology (flooding, rising water table), soil compaction (deterioration of the soil structure), excess of organic matter, increased biological activity due to the increased nutrient supply or temperature. Under natural conditions, most European soils present pH between about 4.5 and 8 (Legros, 2007).

To estimate the evolution of the four metals in time, it is fundamental to have an approximation of their distribution among the soil fractions.

In addition, it has to be considered that there is a fraction of the metal that will be leached very easily by rainfall in the “young” period of the pollution. This amount depends on the type of pollution and its application on the site, in addition to the rain characteristics (intensity, length, frequency).

5 Conclusions

As a conclusion, it is interesting to notice that current models in LCA are mainly based on rough assumptions such as a behaviour of metals in soils at 100 years or 60 000 years, and therefore it is not easily adaptable for assessing the transfer for soil to water. AT the same time, models comparing the mobility of Cd, Cu, Pb and Zn are coherent, highlighting the high mobility of Cadmium and Zinc afterward, while Cu and Pb appears to be more sensitive to the media conditions. Four key parameters are identified such as fraction of metals in soil, fraction of organic matter, Redox-potential and pH. The question of the most adapted way to consider and quantify the impact of emitted heavy metals on a long term period in LCA has to be addressed considering these parameters. Nevertheless, at this time we can only consider a general tendency and a relative impact of the different metals for cations. It is not easy to calculate which fraction of each metal will be released at 100 years and 1000 years. In spite of these limitations, it is possible to have an idea of the faction transferred of the cations that are commonly tested in ecotoxicology, such as copper, cadmium, etc. Models (such as the Free Ion Activity Models) can be used and eventually associated with a Biotic Ligand Model. This issue is even more complicated with anions since it is not feasible at this time to model the mobility and the effect of these metals and metalloids. It was not so problematic because considering the Ronde Venen case study; it was possible to focus on the 4 cations cited above. Nevertheless, anions are among the most impactant substances in LCA and it will be necessary to consider cases studies involving anions in further researches.

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Deliverables D2.2 and D3.2: Presentation of the key ecological parameters for multiple scale assessment, selection of the most

relevant indicator for the assessment of soil quality, and presentation of the minimum required dataset for contaminated soil characterization

Date of preparation: April 10, 2008

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Author of deliverables: Hélène Beauchamp

Institution in charge of the deliverable: VU – Department of Animal Ecology

Date: April 10, 2008

Summary

Integrating Multiple Scale Assessment on Ecosystems for Contaminated Land Management (MuSA) is one project within the ERA-NET project SNOWMAN. SNOWMAN is a research program focused on transnational issues related to soil and groundwater contamination. The goal of MuSA is to develop a decision-support tool integrating Ecological Risk Assessment (ERA) and Life Cycle Analysis (LCA). The utility of this development is that an integration of these existing methods will permit the quantification of ecological impacts of soil contamination at temporal and spatial scales that go beyond the site-specific assessment methods currently employed.

This report focuses on soil contamination by trace elements. Realistic characterization of the behavior and impact of trace metals on soil ecosystems is needed. Moreover, inorganic substances provide an ideal opportunity to expand risk assessment of contaminated land to long time-scales, because they are characterized by long residence times in soils.

The primary goal of this report is the identification of the key ecological parameters describing the relationship between soil contaminants and soil ecosystem elements (animals, plants, microbes, soil matrix). The scope of these parameters is broad enough to include large-scale processes such as seasonal events and land-use modifications, and small enough to describe the chemical behavior of contaminant substances in both the soil matrix and soil organisms. In this way, predictions can be made regarding the negative impacts of soil contamination on soil organisms, the ecological implications of these negative impacts, and the comparison between the ecological impacts of different management strategies aimed at reducing the contamination. Fourteen parameters are identified: season, soil age/history, hydrology, land use, connectivity, mineralogy, vegetation, specific composition, bioturbation, chemical composition, substance availability, microbial activity, soil organic matter content and composition, and toxicity.

The role of indicators in this method is explored. Suggestions are made regarding indicators of the level of ecosystem stress, the extent of ecosystem stability, and the potential for ecosystem persistence. Recommendations include the use of a system of bioindicators that include information at the levels of communities of soil invertebrates, plant communities, and soil ecosystem processes.

The use of these parameters in site characterization provides the framework for quantifying the ecological impact of soil contamination. In this vein, suggestions are given for onsite data collection that will facilitate comparison between contaminated sites once all data are combined into appropriate databases.

The next steps in this project include a description of the similarities, differences, and complentarity between ERA and LCA, placing the site characterization using key parameters into the context of sustainable land management, and illustrating the developed method using a case study based on a contaminated site in the Netherlands.

Preface

SNOWMAN research and MuSA

SNOWMAN is a transnational research program in the field of soil-water-groundwater systems with respect to contamination. MuSA is one project within the SNOWMAN research network. The focus of the MuSA project is the development of a decision-support tool combining the complementary analyses of Ecological Risk Assessment (ERA) and Life Cycle Analysis (LCA). MuSA especially responds to a need of soil resource management authorities for scientifically sound methods permitting the ecological risk assessment of contaminated soil at spatial and temporal scales beyond those employed currently. The current study recognizes the need for decision-support tools that can quantitatively evaluate the ecological impacts of site management strategies. Emphasis is placed specifically on contaminated sites; however, the theoretical framework could be applied to site management of non-contaminated sites as well, for authorities evaluating development plans at a regional or national level. This report contains Deliverables D2.2 and D2.3 of the MuSA project.

Table of Contents

1. Introduction

Risks of soil contamination

Soil contamination with chemical substances is one of the most pervasive forms of terrestrial ecosystem pollution. Chemical substances often have long residence times in the soil matrix, and soil constituents display chemical behavior that modifies their ability to contain or retain chemical molecules. This inherent and unique capacity means that soil can behave as a sink or a source of pollution. Pollution is associated with certain risks, namely risks to humans and risks to ecosystems.

The main risks of soil contamination are 1) the potential to spread to other environmental compartments (a and b in Figure 1) 2) negative impacts of contamination on elements of the terrestrial ecosystem (c in Figure 1) and 3) negative impacts on human health (d in Figure 1), via a) direct contact with soil (ingestion, inhalation, cutaneous contact) b) contamination of water or air supplies by contaminants originally emitted to soil and c) trophic accumulation.

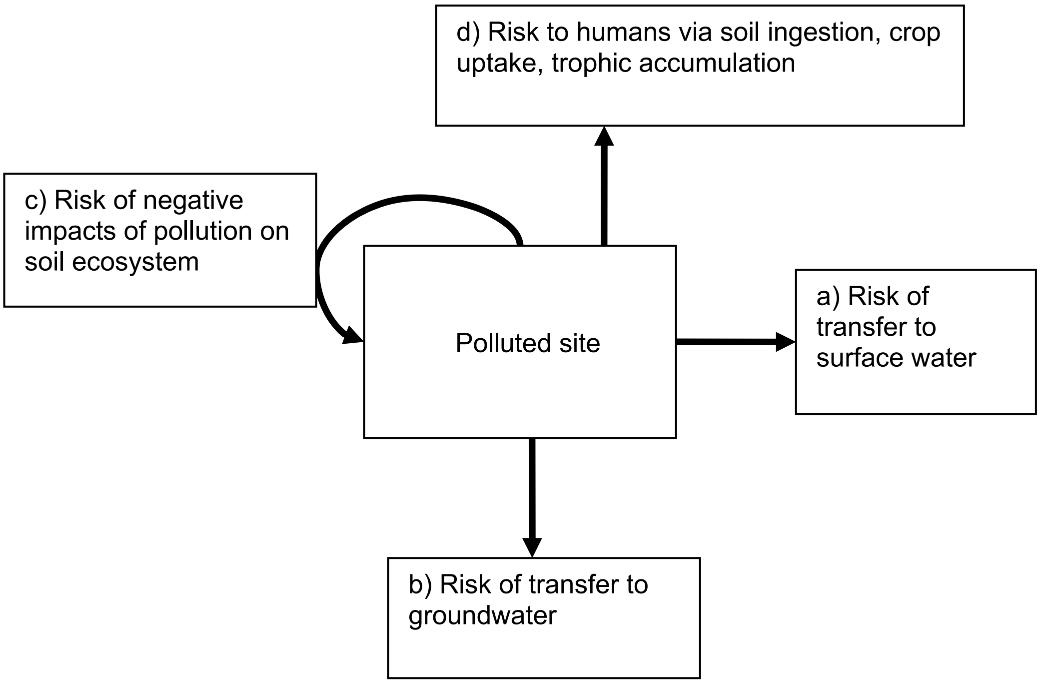

Figure 1. Different categories of risk assessment undertaken for sites with soil pollution.

The risk posed by soil contamination to the organisms inhabiting the soil column and the soil surface can be evaluated with respect to the effects of the contamination on 1) the ability of the populations present to sustain themselves through time, and 2) on the ability of the ecosystem to persist with time. While the two objects are linked, they are not identical. It is an emergent property of ecosystems that the energetic organization of biological communities forms the basis of ecosystem stability (Moore and de Ruiter 1997). Both objects 1) and 2) explicitly involve the notions of time, space, and biological organization; therefore, an evaluation of the risk posed to these objects necessarily calls for a consideration of these multiple levels.

The negative impacts of soil pollution on living elements of the soil ecosystem have the potential to lead to species extinction. Species extinction can occur at a very small scale such as a contaminated site, or at a larger scale, for a region, nation or continent; the ultimate scale is of course complete extinction at the global level. The difference between a local extinction event and a global extinction is the possibility for members of the same species to exist in different locations (Jepson 1997). Of the main risks presented by contaminated soils, species extinction is the only irreversible risk with no directly obvious impact on human health. However, species survival has impacts on human health through ecosystem functions. Species extinction is also a risk in the case of contamination spreading to other environmental compartments.

Risk-based management of contaminated sites

When associated with an unacceptable level of risk, contaminated soil must undergo treatment to reduce the risk it poses. Evaluating the level of risk posed by soil contamination is a complex task and proceeds along varied pathways depending on the geographical and political context of the contamination. If soil remediation is required due to pollution, risk assessment has typically concluded that the concentrations of chemical substances in the soil exceeded an established standard. These standards are set by the political authorities responsible for the oversight of environmental quality compliance. The establishment of these standards is typically based on a combination of political and ecotoxicological criteria.

Ecological risk assessment seeks to evaluate the risk of contamination to the ecosystem, and can proceed by evaluating the risk to biological populations actually or hypothetically inhabiting the polluted site, or by evaluating the risk to the functions performed actually or potentially by the contaminated site, or by evaluating both.

Ideally the kind of risk assessment used for contaminated sites evaluates how the risk posed by site contamination changes under different management strategies. In order to provide a realistic starting point, one of the management strategies considered for comparison should always include “change nothing”.

Any impact due to management or use of soil ecosystems will have effects on a long time scale. Soil formation is a process regulated at a geologic time scale; this process yields unique results depending on five major factors: climate, parent material, organisms, topography, and time (Fitzpatrick 1991). When humans are the organisms acting on soils, the impacts often take place very rapidly when compared to the effects of non-human organisms or the other soil-forming factors. This discrepancy is very apparent when compared to the speed at which soils recover from physical or chemical degradation: for soil microbial communities exposed to chemical pollution this is on the order of months to years (Griffiths et al 2001a), for soil microaggregates this is on the order of 15-20 years (Imeson 1995), for entire soil ecosystems this is on the order of hundreds of years (Fitzpatrick 1991). This considerable inertia also means that impacts on the biogeochemical cycles of soil systems may not be apparent immediately following chemical contamination.

The long time scales required for the development of soil ecosystems is also one of the main reasons to quantify the impact of the “change nothing” option. Remediation activities will have drastic effects on the structure of the soil profile, the physico-chemical properties of the soil, and the suitability of the soil as a habitat for biological communities. From an ecological point of view, the impact of the remediation activities may exceed the impact of soil contamination. This is counter-productive if the goal of soil remediation is to create or maintain the potential of the contaminated site to provide habitat for non-human populations. It is of great interest to managers of soil ecosystems to be able to compare the impacts of soil contamination with the impacts of remediation or other management strategies using a common unit.

Expanding the possibilities of site management

The goals of this project are the development, operationalization, and dissemination of an approach to contaminated land management that integrates the existing decision support tools of Ecological Risk Assessment (ERA) and Life Cycle Analysis (LCA). These two tools provide complementary information to authorities wishing to manage contaminated land in a sustainable way.

In this report a theoretical framework is developed that will allow decision-makers to evaluate and compare the ecological implications of site-management scenarios. It is one of the explicit development goals of this framework that managing authorities be able, and encouraged, to easily scale up from a site-specific management strategy to its ecological implications at the level of the site, region, nation, or continent. Conversely, managing authorities may wish to quantify the ecological impact of a regional (or national) management decision on one or a number of specific sites.

The starting point for this evaluation and comparison is a site characterization that includes all of the parameters necessary for the estimation of impacts at small (short-term, site-specific) and large (long-term, regional or continental) scales. In this report a description is given of the key parameters that interact to govern the behavior and describe the impact of contaminants in the soil ecosystem. Emphasis is placed on trace elements; however, the theoretical framework can be applied to any substance occurring at a contaminated site. The identification of these parameters leads to recommendations of suitable indicators and a minimum dataset for site characterization. Finally, future steps in the development of the integrated method are described.

2. Focus on metals

Metal pollution in soils

Metal ions are ubiquitous in soils, both as endogenous and anthropogenic pools. Metals can be classified based on their biological function as essential or non-essential. Essential metals are necessary for the proper functioning of biological processes, mostly as enzymatic co-factors. The most common essential metals are Fe, Cu, Zn, and Mn (Alloway 1990a).

Soil pollution is characterized by long residence times of pollutants due to the slow mass transfer within the soil column and the capacity of the soil solid phase to retain contaminant particles and molecules. This is true for metals, since metals are not subject to biodegradation, and are only exceptionally (as in the case of methyl-mercury) volatilized from the soil, leaving leaching, erosion, and biomass removal as the primary removal mechanisms (Smal and Salomons 1995).

Much is known about metal emissions to terrestrial and aquatic ecosystems. Some is known about their behavior in these compartments. Less is known about the behavior of metals with respect to organisms. It is necessary to incorporate the most realistic parameters into the assessment of the impact of metals on environmental quality.

Current Dutch policy regarding risk assessment of metals

Soil quality criteria for metals in the Netherlands account for naturally occurring background concentrations of trace elements, as well as anthropogenic emissions (Crommentuijn et al 2000, Vegter 1995). This procedure is unique to trace elements, which occur naturally in soil in varying concentrations. This variation is a natural phenomenon resulting from weathering of parent material or other soil-formation processes. As such, it can promote biodiversity by creating unique ecological niches. Logically speaking, there is no sense in setting environmental quality standards for naturally occurring substances that are lower than their natural concentrations.

The Dutch approach takes into consideration the following parameters: background metal concentrations, soil clay and organic matter content, toxicological risks for humans and ecosystems, land use, and political considerations (Rutgers 2008). Legally limiting values are determined, against which concentrations occurring in the field are compared. These legal values exist at three main levels: reference values (target values), intervention values, and maximum permissible values. Reference values are defined as the naturally occurring concentrations, independent of human action. Target values are defined as the theoretical values at or below which no adverse effects are expected for humans or ecosystems. For trace elements, target values equal reference values. Intervention values are defined as the concentrations at or above which adverse effects are expected, and thus remediation is legally required. The maximum allowable concentrations vary depending on land use category (Agriculture/Nature, Human habitation, Industry) and the surface area of the site considered (Rutgers 2008). Maximum allowable concentrations are lower than intervention values for sites whose land use is agriculture/nature or human habitation; for industrial sites the maximum allowable concentration equals the intervention value.

Table 1. Legal concentration values in the Netherlands for Cd, Cu, Pb, and Zn in soil

Element	Cb~ (mg/kg)	MPC_{Nature}~ (mg/kg)	$MPC_{Human\ habitation}$~ (mg/kg)	IV ~ (mg/kg)	Csludge (mg/kg)
Cd	0,6	0,6	3,7	12	23 #
Cu	40	40	54	190	1230 *
Pb	50	50	50	530	700 !
Zn	140	140	198	720	2780 *

Cb – background concentration, MPC – maximum permissible concentration, IV – Intervention value (see text for subscript explanation), Csludge – average value in sewage sludge (not only Netherlands); **Sources:** ~ Swartjes 1999, * Baker 1990 (median value), # Alloway 1990b (Ch. 6 - average value), ! Davies 1990 (median value)

In the Netherlands, the total concentration of trace elements in soils is compared to soil standards after correction for soil type. This correction is based on surveys of Dutch soils whereby regressions were established between total metal concentrations and the soil content of organic matter and clay particles in undisturbed, non-anthropogenically affected soils. In this way, it is assumed that soil concentrations of trace elements are more comparable between different soil types after the correction procedure.

Metals in soils: Partitioning and speciation

Some general statements can be made regarding the behavior of metal atoms in soil. The localization of metal atoms in the solid or liquid phase of the soil is referred to as partitioning. If the total soil concentration of the metal in question is known, the quantity of metal in the liquid phase (soil solution) can be estimated using the partitioning coefficient, K_p. The value of this coefficient will vary according to multiple factors including the element in question, total metal load, solution pH, and soil organic matter content in the solid and liquid phases (Sauvé et al 2000).

If a metal atom is not in the soil solution, it can be associated with more or less affinity to the solid phase. Sorption is a general term describing the association between a metal atom and the solid phase. Absorption refers to the penetration or permeation of one phase by another, and adsorption refers to an attachment to a surface (Hillel 1998). Desorption is the opposite of adsorption, and describes the metal atom leaving the solid surface. Metal atom partitioning between the solid and liquid phases is a dynamic process; therefore, any punctual measurements give an estimate or approximation of the order of magnitude of the concentrations in the different phases.

Metal speciation refers to the chemical formula of the metal atom. This is important for the behavior of metals in soil because different metal species have different solubility, electrostatic affinity, and perhaps even partitioning characteristics. Organisms display selective uptake affinities for different metal species as well (e.g McLaughlin 2002, Parker et al 2001). Common metal species in soils include the free metal ion (M^{2+}), hydrolysis products i.e. ($M(OH)^-$), inorganic complexes (MSO_4, MCl, MPO_4), and organo-metal complexes formed between organic molecules of varying molecular weights and metal ions (Lindsay 1979). Speciation is most commonly assessed for metals in solution; however, speciation in the solid state does affect soil chemical equilibria (Lindsay 1979).

Metals in soils: Different pools

Metal atoms in soil matrices can be separated into different pools. The movement of metal ions between these pools occurs as a function of shifting chemical equilibria in the soil (Bermond and Benzineb 1991). This has implications for the assessment of bioavailability of metals, because unavailable metals may become available as soil conditions change. This may also explain why in many cases the strongest correlations are found between metal concentrations in organisms and total soil metal concentrations, rather than with water soluble, pore water, or "exchangeable" metal concentrations (Boivin 2005, Spurgeon et al 2006, van Gestel in print, Veltman et al 2007a, Veltman et al 2007b).

The different pools are described below (*sensu* Kiekens 1990):

1) Soluble free ion: uncomplexed fraction present in soil solution
2) Complexed pool: metals bound to ligands
3) Exchangeable pool: metals bound by electrical charges to the solid phase
4) Secondary minerals: clays and amorphous minerals
5) Primary minerals: metal atoms occurring in geologic formations

The concentration of the soluble free ion will depend on the chemical equilibria in the soil (Lindsay 1979). This concentration can be measured with more or less accuracy using for example ion selective electrodes (e.g. Lund 1990), it can be estimated using empirical regression equations based on total concentration and soil properties (e.g. Sauvé et al 2000), or it can be estimated by speciation programs taking as inputs chemical equilibrium constants and concentrations of various chemical species in the soil solution (i.e. MINTEQ, WHAM).

The complexed metal pool includes organic and inorganic complexes. Complexes can form on the solid phase or in solution. As the chemical and hydrological conditions of the soil solution evolve, many inorganic complexes can precipitate or dissolve, resulting in changes in the soluble metal concentration (Lindsay 1979). The formation of complexes between chelating agents or dissolved organic matter and metal ions will increase the mobility of metals, and may increase or decrease availability for organism uptake based on the stability of the complex and the chemistry of the soil solution (Kiekens 1990). In particular, fulvic acids show selectivity towards metal ions, the result of which is increased mobility and solubility of metals in soils with high concentrations of fulvic acids (Kiekens 1990).

The exchangeable metal pool represents metals bound to some part of the solid phase due to electrostatic charges. The magnitudes of these charges depend to a certain degree on the pH (Janssen et al 1997a, McBride et al 1997). Clay minerals have (more or less permanent) negatively charged surfaces in the normal soil pH range; these are due to the isomorphous substitution of ions in the crystal lattices of the clay minerals (Sparks 1995). The charge on soil organic matter is more variable with pH due to the effect of protonation of its functional groups; of specific interest for metal binding are carboxylated phenolic groups and hydroxyl groups (Schulin et al 1995). Metal ions can also be attracted by the negatively charged functional groups on living or non-living bacterial surfaces; however, this binding is in general less reversible than metal binding to the inorganic solid phase (Beveridge et al 1995).

The pool of secondary minerals includes metal ions contributing to the structure of clay mineral crystal lattices, amorphous mineral coatings on other solid phase particles, and precipitated minerals resulting from shifts in soil chemical equilibria (Lindsay 1979, Alloway 1990a, Janssen et al 1997a). These metals are typically considered to be unavailable for uptake; however, they may become available due to shifts in chemical conditions resulting in the dissolution of precipitated mineral phases, or physical destruction of clay minerals.

Metals present in primary minerals are not directly available for uptake. However, the presence of soil parent material rich in metal(s) may indicate relevant background soil concentrations of trace elements. Localization of metal ions, and concentration gradients in the soil column can indicate their origin; metals primarily concentrated in the upper soil layers are most likely of anthropogenic origin (Marinussen 1997) whereas those concentrated in the lower soil layers are most likely the products of weathering (Baker 1990). Over the long term the elements contained in parent materials will be incorporated into the soil column via weathering and other pedogenetic processes (Fitzpatrick 1991).

There is much evidence to suggest that total metal load strongly influences metal behavior in both the solid and liquid phase (Lindsay 1979, Sauvé et al 2000). This is due to many factors, including modifications of chemical solubility, competition between metal ions of different elements for non-selective adsorption sites, competition between metal ions of the same element for selective adsorption sites, complexation reactions, modifications of soil pH, changes in degradation rate of SOM, and changes in biogeochemical cycles.

Metals in soils: Cadmium, Copper, Lead, and Zinc

Each trace element exhibits unique chemical behavior, and these individual characteristics will be described. For each metal a brief overview is given including the primary anthropogenic inputs to soil and metal-specific factors influencing partitioning and speciation. This report includes information on four trace elements: cadmium, copper, lead, and zinc. These four metals are well-characterized in the scientific literature, they often occur simultaneously in soils contaminated with trace elements, and they represent a large proportion of trace element emissions worldwide. Many other trace elements have the potential to cause ecological damage, including Ag, As, Be, Bi, Co, Cr, F, Hg, Ni, Mn, Mo, Sb, Sc, Sn, Tl, and V (Adriano 2001). It is however outside of the scope of this report and this project to treat them all. The trace elements As, Cr, and Ni will be addressed in a later report.

Cadmium

Cadmium (Cd) is generally considered to be non-essential; however, for some organisms it may be essential (Rainbow 2002, Wallace 1989). It is a relatively rare metal whose natural occurrence in soil is very low (Alloway 1990b). Anthropogenic inputs of Cd to soil include atmospheric deposition from mining, smelting, and other metallurgical activities, incineration of plastics and batteries, sewage sludge disposal, phosphatic fertilizer use, and various aerosol particles resulting from industrial/urban air pollution (Alloway 1990b). Cd is of specific concern due to the fact that it can accumulate in crop plants at levels that are toxic to plant consumers (humans, herbivores) but not phytotoxic (McLaughlin 2002).

The selectivity of soil adsorbents is lower for Cd than for Cu or Pb; thus, Cd is more mobile in soil columns than either Cu or Pb (Alloway 1990b, Marinussen 1997, Morel 1997, Weng et al 2002). There is some evidence to suggest that hydrous oxides of Fe and Mn have higher selectivity for Cd than does SOM (Alloway 1990b); this effect is strongly pH dependent. High concentrations of Cd or other metals also contribute to Cd mobility, as it is easily displaced from exchangeable sites by other cations (Alloway 1990b, Bongers 2007, Jonker et al 2004). However, the relationship between this mobility and uptake e.g. by plants is not clear, because excesses of some elements (Cu, Ni, Se, Mn, P) reduce Cd uptake, whereas excesses of others (Zn, Pb) may result in increased Cd uptake (Alloway 1990b). The most important soil property controlling Cd sorption is pH: for pH levels between 4 and 7.7, soil Cd sorption capacity increased 2-3 fold for every unit increase in pH (Christensen 1984a).

In polluted soils, the Cd^{2+} ion predominates, with neutral species (e.g. $CdSO_4$ or $CdCl_2$) present in increasing proportions where pH is greater than 6.5 and activities of these anions in soils are high enough (Alloway 1990b, Lindsay 1979). At higher pH levels (pH>7.5) solubility of Cd is limited by the formation of $CdCO_3$ (Lindsay 1979).

The only oxidation state of Cd that is stable in the redox range of soils is Cd(II). However, changes in soil redox conditions can affect Cd toxicity if reducing conditions result in the formation of insoluble cadmium-sulphides (Alloway 1990b).

Copper

Copper (Cu) is an essential element for plants, animals, and some microbial processes; it is also potentially toxic, and this margin between essentiality and toxicity is narrow in plants (Adriano 2001, Baker 1990). Soil Cu has both natural and anthropogenic origins (Baker 1990, Lindsay 1979). Non-anthropogenic soil copper occurs due to weathering of Cu-containing parent material and subsequent retention of Cu by soil constituents (Baker 1990). Anthropogenic sources include fertilization, pest control, application of livestock manure, sewage sludge, or dredged sediments, effluents from mine tailings, and atmospheric deposition (Baker 1990, Yaron et al 1996). Cu has a strong affinity for the soil matrix, and is one of the least mobile trace elements (Baker 1990, Schulin et al 1995, Yaron et al 1996). Cu preferentially sorbs to soil organic matter, this sorption is responsible for much of the strong soil retention of Cu (Baker 1990, Buekers et al 2007, McBride et al 1997, Sauvé et al 1997, Sukreeyapongse et al 2002). On the other hand, if the Cu in the soil profile is complexed to dissolved organic matter, this complexation may result in higher Cu mobility (Weng et al 2002). The soil pH influences soil retention of Cu somewhat; however, the effect of pH on Cu mobility is lesser than for other trace elements (Liang and McCarthy 1995, McBride et al 1997, Sukreeyapongse et al 2002, van Gestel et al 1995, Weng et al 2002). Cu occurs on the inorganic solid phase adsorbed to hydrous oxides, adsorbed to clay colloids, incorporated into soluble minerals (e.g. carbonates), and as isomorphous substitutions in clay mineral crystal lattices (Baker 1990).

The partitioning of Cu between the solid and aqueous phases of the soil matrix will depend mostly on the total Cu content of the soil and the quantity and nature of soil organic matter (McBride et al 1997, Sauvé et al 1997, Weng et al 2002). The concentration of dissolved Cu increases as the concentration of dissolved organic molecules increases (Weng et al 2002). The speciation of Cu in the soil solution depends on the quantity and nature of soil organic matter, the ionic strength of the soil solution, the nature and quantities of possible ligands, and the pH (Buekers et al 2007, McBride et al 1997, Paoletti and Sommagio 1996, Sauvé et al 1997). At typical soil concentrations, the most important inorganic ligands for Cu are sulphate (SO_4^-), hydroxyl ion (OH^-), and carbonate (CO_3^-) (Lindsay 1979). Under very reducing conditions ($pe + pH < 2.38$) Cu(I) becomes an important free ion species; above this level Cu(II) dominates as the free ion (Lindsay 1979); however, most of the Cu in the soil solution will occur as species other than the free ion. Characteristics of the Cu atom promote its binding to living and non-living organic matter: its tendency to form S-bonds contributes to its retention on SOM, its uptake by plants and soil organisms, its mobility in the organism, and its potential toxicity (Eijsackers 1996, van Kolck et al 2008). The same characteristics can lead to deficiency when Cu occurs as a mixture with other, more mobile trace elements such as Zn (Baker 1990).

There are complex relationships between soil organic matter in the solid phase, dissolved organic matter content, and soil pH (Bongers 2007, Ponizovsky et al 2006). As dissolved organic matter content increases, dissolved Cu increases; analysis of 68 field soils showed that in most of the soil solutions 98% of Cu was bound to organic ligands (Sauvé et al 1997). Addition of Cu will cause a reduction in dissolved organic matter content due to direct and indirect coagulation effects: Cu causes coagulation directly, and it displaces aluminum into solution, which also promotes coagulation (Ponizovsky et al 2006). The Cu-promoted displacement of ions is also responsible for decreasing soil pH following Cu addition (Bongers 2007, Ponizovsky et al 2006), and changes in sorption characteristics of trace element mixtures in soil (Bongers 2007, Jonker et al 2004).

Decreases in pH increase free Cu ion activity according to three main processes: dissolution of mineral phases containing Cu ions, reduction of organic complexation, and reduction of solid-phase Cu adsorption (Baker 1990, McBride et al 1997). Decreases in soil pH can lead to increases or decreases in dissolved organic matter content, depending on the composition of the organic matter and its corresponding solubility (Kumada 1987).

Lead

Lead (Pb) is not known to be essential for any living organisms (Adriano 2001). Lead occurs naturally in soils due to weathering of soil parent materials (Davies 1990). Primary anthropogenic inputs include sewage sludge spreading and atmospheric deposition due to mining, smelting activities, and vehicle exhaust (Davies 1990).

Pb in soils is readily immobilized, largely due to its strong affinity for the soil solid phase in general, and the organic fraction in particular (Davies 1990, Marinussen 1997, McLaughlin 2002). It also displays a tendency to form S-bonds (Eijsackers 1996, van Kolck et al 2008). Under strongly acid conditions ($pH<2.5-3$) leaching of Pb from soils becomes a quantitatively important removal mechanism (Davies 1990). Otherwise, Pb is prone to displace other ions with weaker affinity for the solid phase, such as H^+, Cd^{2+}, and Zn^{2+} (Bongers 2007, Morel 1997). Concentrations of Pb in solution will also increase in soils with high total lead concentrations (Sauvé et al 1997, Weng 2004). Typically, concentrations of soluble Pb represent less than 1% of total soil Pb; furthermore, 80% of the Pb atoms in solution can be bound to dissolved ligands, which implies that free Pb^{2+} ion activity will be low (Davies 1990).

Below pH 8 only Pb^{2+} and $PbOH^+$ contribute significantly to Pb in solution (Lindsay 1979). When $6.5<pH<8$, an increase in pH promotes the formation of Pb-organo complexes in solution, thereby increasing Pb mobility (Sauvé et al 1998). On the other hand, increased pH will result in greater formation of $Pb(OH)^+$ and $Pb(OH)_2$, which are preferentially adsorbed over the free Pb^{2+} ion (Weng 2004).

Zinc

Zinc (Zn) is an essential element for animals, humans, plants, and some microbial processes, which can become toxic at high concentrations (Kiekens 1990). It can occur naturally in soils due to weathering of parent material, or it can occur due to anthropogenic activities such as use of agrochemicals (fertilizers, pesticides), spreading of sewage sludge, compost, or dredged sediments, and metal smelting resulting in atmospheric deposition (Kiekens 1990).

Soil pH is the most important factor influencing Zn behavior in soil, both for solid-to-liquid partitioning and for speciation in solution (Lock and Janssen 2001, McBride et al 1997, Sauvé et al 2000, Weng et al 2002). The critical value for Zn desorption is a pH of about 5 (Kiekens 1990). As pH increases, Zn sorption increases; this pH-dependent sorption behavior is more marked in sandy soils than in soils with high concentrations of colloid-sized materials (Kiekens 1990).

Under normal soil conditions, Zn minerals are soluble, which implies that the solubility control of Zn in soils is more related to ion sorption than mineral dissolution (Lindsay 1979). Zn speciation in solution is highly dependent on pH: at $pH<7.7$ the free Zn ion, Zn^{2+}, is the predominant species, while $Zn(OH)^-$ is more prevalent above this pH; and at $pH>9.11$ the neutral species $Zn(OH)_2$ is predominant (Lindsay 1979). At $pH> 6$ Zn toxicity is expected to be low (McBride et al 1997). Under normal soil conditions Zn can form stable inorganic compounds with sulphate, phosphate, and hydroxyl ion, depending on the activities of these ions in solution (Lindsay 1979). Zn also forms complexes with organic molecules such as humic substances, although the affinity of Zn for these molecules is lower than that of Cu (Marinussen 1997, Weng et al 2002). The stability of Zn-fulvic acid complexes increases as pH increases, which can result in increased Zn mobility in the soil, unless the concentrations of flocculating agents such as Mg^{2+}, Ca^{2+}, Fe^{3+}, or Al^{3+} are high enough to remove the metal binding colloids from suspension (Kiekens 1990).

Ions of different elements can affect each other’s partitioning, speciation, and availability in soil. Zn is more mobile in soil than Cu or Pb, and high concentrations of either or both of these ions can displace Zn ions from sorption sites into soil solution, independently of any pH effect (Bongers 2007, Christensen 1984b, Jonker et al 2004, van der Wurff et al 2007). This displacement can result in higher bioavailability and thus toxicity, or accelerated leaching and thus lower toxicity, depending on the soil texture, hydrology, and characteristics of species in contact with the soil. In soils with high concentrations of multiple trace elements, Zn toxicity may appear before toxic effects due to other elements because soil Zn concentrations are often high relative to other metals, and Zn is more mobile in soil than Cu or Pb (Davies 1990, Hopkin and Hames 1994)

The essential nature of both Zn and Cu complicate the prediction of body concentrations based on soil characteristics such as total metal content, pH, OM content, or clay content (Bleeker and van Gestel 2007, Hobbelen et al 2006a). This is due to the fact that complex and effective regulatory mechanisms exist that govern uptake and storage of these metals to a certain extent (Hunter and Johnson 1982, Rainbow 2002, Vijver 2005). This implies that concentrations in animals will follow a biologically determined accumulation pattern, and that excretion of excess metals will take place (Rainbow 2002). This is found to be true to a certain extent; however, metal concentrations in animals from metal-rich soil are typically higher than those of animals in less enriched soils, which suggests that these limits are not absolute (Hobbelen et al 2006a, Hobbelen et al 2006b, Hopkin et al 1989).

3. Key parameters affecting ecological impact of metals in soils

The realistic characterization of the behavior of metals in soil ecosystems must account for the parameters controlling that behavior. Identification of these parameters allows the reliable prediction or diagnosis of the effects of a given concentration of metal on the soil ecosystem and its component populations. Fourteen parameters have been identified; these are described below.

Interactions between parameters: explanation of tables

The parameter descriptions below include for each parameter a table briefly explaining the interactions between each parameter and the others. These tables are meant to provide a succinct description of the nature of the influence of the parameter being described on the other 13 parameters. The idea behind the presentation of these tables is to give an overview of how the interactions between these fourteen parameters describe and create a dynamic bio-physico-chemical system. Estimations of impact and predictions of the ecological implications of site management strategies can be made based on this characterization of the system and the relationships between the identified parameters. Use of the greater-than symbol, ">", means that the parameter being described is within the domain of influence of a higher-level parameter. Use of the phrase "limiting conditions" means that the parameter being described has the potential to limit the possible values of the parameters it is interacting with.

Season:

This parameter reflects the fact that in all ecosystems there are cyclic phenomena that are predictable based on seasonal changes in abiotic factors such as length of day, average temperature, and precipitation. Many of these phenomena are reflected in the life cycles of organisms in the form of biological events such as egg laying, hatching, molting, breeding, hibernating, estivating, seed/spore production, flowering, senescence, stockage, emergence, and activity level. These biological cycles can interact with contaminants to reduce or amplify toxic effects (Boivin 2005, Davies 1990, Marinussen 1997, Maelfait 1996, Migula 1996, Morel 1997, Schaefer and Schauerman 1990). Species diversity can also be determined in part by seasonal variations in abiotic conditions (Maelfait 1996). Inundation is an example of a seasonal event having a strong impact on soil ecosystems (Klok et al 2006, Wijnhoven 2007). Seasonal variations in environmental conditions also affect contaminant behavior via modification of chemical reactions and physical processes (Förstner 1995, Wenzel and Blum 1995, Yaron et al 1996). Seasonality varies from one geographical location to another, but varies little within the same geographical area, and remains constant from one site to the next. This parameter is easily assessable via consultation of a map or meteorological data. The nature of seasonal variation should be included in any sampling or monitoring plan.

Effect of this parameter → on this parameter ↓ is:	Season
	🟩
Microbial activity	cycles of activity/dormancy
Chemical composition	via biogeochemical cycles
Mineralogy	historical development, erosion
Hydrology	precipitation, insolation
Land use	limiting conditions
Substance availability	via biogeochemical cycles
Soil age/history	historical development
Vegetation	limiting conditions
Specific composition	limiting conditions
Bioturbation	limiting conditions
Toxicity	limiting conditions
Connectivity	development of vegetation
SOM content and composition	limiting conditions via organisms
Season	🟩

Soil age/history:

This parameter is related to the soil taxonomy, the depth of the soil column, the stage of the development of the soil, and the topography of the site. This parameter is somewhat related to land use, in the sense that historical and present land use are partly determinant for the present physical, chemical, and biological state of the soil. However, soil age/history and land use should be maintained as separate parameters because they address different aspects of the soil system, and because one can be changed through policy (land use) and the other can only be described or assessed (soil age/history). This parameter is related to the history of disturbances of the site, anthropogenic in nature or otherwise (Smal and Salomons 1995, Yaron et al 1996). In the case of contaminated soil, the nature and extent of prior contamination events will influence the behavior of subsequent contamination events, as well as the organism and ecosystem response and recovery (Alloway and Morgan 1986, Van der Wurff et al 2007). The history of the system may influence soil retention or contaminants; in the case of slow diffusion of contaminants into minerals a longer time between contamination and remediation may lead to decrease in removal efficiency (Yaron et al 1996). This parameter is easily assessable via consultation of soil maps, GIS databases, expert consultation, review of historical documents, etc.

Effect of this parameter →	Soil age
on this parameter ↓ is:	🟩
Microbial activity	fungi:bacteria ratio
Chemical composition	limiting conditions
Mineralogy	limiting conditions, CEC
Hydrology	>
Land use	historical development
Substance availability	physical occlusion, aging
Soil age/history	🟩
Vegetation	specific suitability, succession
Specific composition	suitability via vegetation
Bioturbation	specific suitability
Toxicity	delayed long-term effects
Connectivity	historical development
SOM content and composition	limiting conditions
Season	>

Hydrology:

The hydrology of a site describes the relationship between the site and the surface, atmospheric, and ground-water with which it communicates. This parameter is influenced by precipitation, infiltration, the hydrological regimes of all bodies of water connected to the site, and human action thereupon. It is also influenced by the soil structure (Hillel 1998, Sparks 1995, Yaron et al 1996). The overall hydrology of a site varies at a large scale; however, the importance of small-scale variation in hydrology is also important in soil ecosystems (Förstner 1995). Variation in hydrological conditions will influence the chemical and biological activity taking place in the soil via modification of redox conditions (Sparks 1995), solute transport (Bengtsson 1997), or animal behavior, such as earthworm burrowing (Marinussen 1997, Wijnhoven 2007). Modifications to the regional or site hydrology in the form of climate change, hydrological management, or natural events have the potential to significantly modify contaminant behavior (Eijsackers 1995). This parameter is easily assessable via consultation of maps or GIS databases, meteorological data, measurement of soil moisture content, observation during a site visit, and consultation with regional or local experts.

Effect of this parameter →	Hydrology
on this parameter ↓ is:	🟩
Microbial activity	cycles of activity/dormancy, transport
Chemical composition	redox cycles, solute transport, reaction rates
Mineralogy	historical development, erosion
Hydrology	🟩
Land use	limiting conditions
Substance availability	solid-liquid partitioning, chemical speciation
Soil age/history	historical development
Vegetation	specific suitability
Specific composition	specific suitability
Bioturbation	limiting conditions
Toxicity	specific suitability
Connectivity	limiting conditions
SOM content and composition	historical development
Season	>

Land use:

This parameter describes the relationship between humans and the soil ecosystem, and the nature of human management. It is typically assessed using descriptive categories such as forest, agriculture, urban, etc. This parameter partly determines the expected specific composition of a site and the comparability between sites (Griffiths et al 2001a). Some general trends with respect to soil ecosystems can be described according to characterization into land use categories (Malkomes 1997, Yaron et al 1996). Decisions concerning the remediation and development of contaminated sites are typically related to past as well as potential future land use choices. Furthermore, options for future land use will depend on the toxicological profile of the soil, remediation strategies available, and management choices (Kuhnt 1995, Stigliani 1995). Some aspects of land use decisions involve legal frameworks, such as the protection of certain species or habitats (Posthuma et al 2005). Other aspects of land use decisions involve the impact of practices related to specific land-use categories, such as irrigation or liming for agriculture that modify the behavior of contaminants and/or trace elements in soil (Eijsackers 1995, Salomons 1995, Solis et al 2005). This parameter is easily assessable and is typically included as part of a site description, but can also be assessed via aerial photography, GIS databases, or directly via a site visit. Historical land use is assessable via consultation of archived documents.

Effect of this parameter →	Land use
on this parameter ↓ is:	🟩
Microbial activity	limiting conditions
Chemical composition	limiting conditions
Mineralogy	erosion, vertical transport, development
Hydrology	infiltration rates
Land use	🟩
Substance availability	inputs, aggregate structure
Soil age/history	historical development/disturbance
Vegetation	limiting conditions
Specific composition	limiting conditions
Bioturbation	limiting conditions
Toxicity	legal requirements
Connectivity	limiting conditions
SOM content and composition	limiting conditions
Season	>

Connectivity:

The connectivity of a site describes its relationships to other sites and/or ecosystems surrounding it. This parameter is determinant for the flow of matter and energy between sites, animal and plant population dynamics, biodiversity, and land use decisions. These relationships are physical as well as biological, including mass transfer of water and soil as well as the biological transfer of genes, and the physical relocation of organisms. The connectivity of a site can change according to factors that are anthropogenic (policy/land use changes), stochastic (earthquake, landslide), meteorological (flooding), and ecological (vegetational succession). Therefore, the value of this parameter will depend on the temporal scale being considered. In sites with patchy or spatially heterogeneous contaminant concentrations the degree of connectivity between patches of differing concentrations will also be important, especially for small soil-dwelling organisms with limited spatial ranges (Marinussen 1997, Wijnhoven 2007). Connectivity is thus determinant for ecosystem vulnerability and recovery. This parameter is easily assessable at a large scale via consultation of maps or GIS databases, and site visits. At the scale of patches of contaminant hotspots within a given site, this parameter is assessable via sampling, measurement, and interpolation.

Effect of this parameter → on this parameter ↓ is:	Connectivity
	🟩
Microbial activity	promotion of gene transfer
Chemical composition	contaminant transport
Mineralogy	particle transport
Hydrology	>
Land use	>
Substance availability	no interaction
Soil age/history	>
Vegetation	dispersal
Specific composition	(e)(im)migration, refugia
Bioturbation	via specific composition
Toxicity	decrease apparent toxicity
Connectivity	🟩
SOM content and composition	degradation rates via movement of organisms
Season	>

Mineralogy:

Soil mineralogy describes the chemical and physical nature of mineral particles in the solid phase. Soil mineralogy reflects all of the processes of soil formation, including human influence and biological processes. The nature of soil minerals is a deciding factor in the retention of contaminants, nutrients, and water in the soil (Bengtsson 1997, Hillel 1998, Weng et al 2001, Yaron et al 1996). The nature and abundance of clay minerals are important factors for soil structure and soil aggregate size (Fitzpatrick 1991, Hillel 1998), which is an important factor for bacterial aggregation (Fournier et al 1997) as well as erosion resistance (Imeson 1995). They also determine mineral catalysis of bound residues (Yaron et al 1996). Mineralogical differences between soils used in laboratory assays and field soils are also a source of variation between lab and field results (van Gestel 1997). For a given region, the mineralogy of natural soils is a result of the composition of the bedrock or parent material, the weathering processes, and the human land use (Smal and Salomons 1995). The mineralogy of a soil is partly determinant for the soil’s ion exchange capacity (Hillel 1998, Yaron et al 1996). Mineralogy is assessable via consultation of soil maps, GIS databases. An abbreviated description of a site's mineralogy is usually given in the description of the soil texture (loamy sand etc.). Particle size analysis is usually carried out in the course of a site characterization, although the exact nature of the mineral particles is not usually determined.

Effect of this parameter →	Mineralogy
on this parameter ↓ is:	🟩
Microbial activity	aggregation, microhabitats
Chemical composition	reaction rates, retention
Mineralogy	🟩
Hydrology	solute transport, leaching rates, water retention, porosity
Land use	infrastructure support, habitat potential
Substance availability	retention, adsorption, CEC
Soil age/history	>
Vegetation	specific suitability via chemistry + hydrology
Specific composition	specific suitability via chemistry + particle size distribution
Bioturbation	extent via specifc suitability
Toxicity	substance retention, release
Connectivity	continuity for vegetation suitability
SOM content and composition	aggregation, influence via vegetation suitability
Season	>

Vegetation:

Ecosystems are classified by the type of dominant vegetation, which serves as the primary source of food and energy for all of the other biological components (Hendricks 1997, Migula 1997). The type of vegetation, and specifically the dominant plant species, will be determinant for the biogeodynamics of a site (Schaefer and Schauerman 1990), and will have important implications for contaminant behavior (Morel 1997, ter Meulen-Smidt 1995). Many sites are small enough in area that they can be characterized by one type of vegetation, although this may not always be the case. If multiple vegetation types are present in the same site, this could modify potential habitats, dispersal characteristics of plants and animals, transfer between environmental compartments, temporal evolution of the site, and site management choices. This parameter is easily assessable at a large scale and according to non-precise categorization, along the lines of the physiognomy of the landscape. At this level, vegetation can be assessed via consultation of aerial photographs, GIS databases, or site visits. Vegetation can be assessed in more detail via standardized surveys of plant species (see below).

Effect of this parameter →	Vegetation
on this parameter ↓ is:	🟩
Microbial activity	promotion via rhizosphere effect
Chemical composition	plant-soil exchange
Mineralogy	>
Hydrology	evapotranspiration, infiltration
Land use	>
Substance availability	nutrient uptake, litter quality
Soil age/history	historical development
Vegetation	🟩
Specific composition	specific suitability via habitat
Bioturbation	specific suitability via habitat
Toxicity	food-chain transfer
Connectivity	limiting conditions
SOM content and composition	litter quality, decomposition via microbes
Season	>

Specific composition:

The specific composition of a site is made up of all of the species' inhabiting the site, including animals, plants, and microbiological populations. The specific composition of a site is not independent of its vegetation, since plant species make up the vegetation of a site. However, it may be helpful to consider separately the general physiognomy of a site (vegetation) from the specific composition, which also includes rare and/or non-dominant species (animal, plant, or other) and may be a deciding criterion for site management, in the case of species with legally protected status, for example (Posthuma et al 2005). Microbial composition may also be considered here, although techniques for assessing microbial diversity are, to date, far behind those used for other taxonomical groups. Furthermore, the active microbial species pool varies according to seasonal patterns; whereas, the specific composition of multi-cellular organisms should vary less (although the biomass will fluctuate), with the exception of migratory animals. See below for a description of the microbial activity parameter. The specific composition of a site can yield information regarding the state of disturbance, stress, or health of the site (Boivin 2005, Traas and Aldenberg 2007). The specific compositions of sites can be compared to evaluate exclusion of species from a site due to contamination, or to assess the degree of tolerance or adaptation to pollution (Boivin 2005, Griffiths et al 2001b, Posthuma 1997, Posthuma and de Zwart 2006). This parameter can be assessed via standardized surveys of the site according to appropriate methods for each biological group considered.

Effect of this parameter →	Specific composition
on this parameter ↓ is:	🟩
Microbial activity	predation, cyclical phenomena
Chemical composition	complex interactions
Mineralogy	transport via bioturbation
Hydrology	>
Land use	>
Substance availability	complex interactions
Soil age/history	> (historical development)
Vegetation	>
Specific composition	🟩
Bioturbation	limiting conditions
Toxicity	limiting conditions
Connectivity	>
SOM content and composition	consumption, degradation
Season	>

Bioturbation:

Bioturbation is the movement of soil materials as a result of “the expenditure of biological (photosynthetic) energy by roots and soil animals, largely invertebrates but sometimes mediated by the burrowing and other soil-disturbing effects of vertebrates,” (Lavelle and Spain 2003, p. 173). The degree to which bioturbation will take place depends on the specific composition of the soil and its physico-chemical properties (Bouché 1996, Imeson 1995, Wijnhoven 2007). Transport of contaminants, nutrients, soil particles, and water will be modified by bioturbation activities (Forbes and Kure 1997).

Animals such as earthworms, ants, moles, voles contribute to bioturbation of soil ecosystems. This contribution is non-negligible, and in the case of one Dutch floodplain soil, bioturbation by earthworms was demonstrated to be responsible for up to twice as much particle relocation as flooding events (Wijnhoven 2007).

Bioturbation events will take place at a spatial scale of the same magnitude as the organism responsible, but in general can be assessed at a spatial scale of the entire site, since the specific composition of the site is expected to vary little. This parameter can be assessed visually in the case of large burrows of small mammals, or deductively based on the occurence of species known to exhibit this kind of behavior (Wijnhoven 2007).

Effect of this parameter → on this parameter ↓ is:	Bioturbation
	🟩
Microbial activity	promotion, transport
Chemical composition	redox conditions, solute transport
Mineralogy	particle transport, changes in aggregate structure
Hydrology	preferential water transport
Land use	>
Substance availability	changes via chemistry, transport
Soil age/history	soil macro (horizon) structure
Vegetation	nutrient transport
Specific composition	>
Bioturbation	🟩
Toxicity	mobile animals have more contact w/toxicants
Connectivity	>
SOM content and composition	degradation, microbial transport, redox conditions
Season	>

Chemical composition:

The chemical composition of a soil describes the nature and concentration and/or activity of all chemical substances (organic and inorganic) in all three phases of the soil. This composition is determined by many factors and is determinant for many factors. The master chemical variables of most ecosystems, both aquatic and terrestrial, are the pH (proton activity) and the redox potential or pe (electron activity) (Sparks 1995). These are both results and drivers of the physical, chemical, and biological processes taking place, and that have taken place (Bongers 2007, Jonker et al 2004, Ponizovsky et al 2006, Sparks 1995). The speciation of many substances depends on these variables; for many substances this speciation will have an effect on toxicity (Alloway 1990a, Bang and Hesterberg 2004, Spurgeon et al 2006, Yaron et al 1996) or degradability (Doelman 1995). The presence and concentration of non-metal ions (such as Na$^-$, Cl$^-$, Ca^{2+}), and other molecules (such as EDTA) can also affect metal speciation and toxicity (Förstner 1995, Schulin et al 1995, Yaron et al 1996) (see below, Substance availability) and can have direct effects on the community structure of organisms (Maelfait 1996, Posthuma and de Zwart 2006).

The pH status of the medium will have both direct and indirect effects on most organisms (Tobor-Kaplon 2006, Spurgeon et al 2006, van Gestel et al 1995, van Gestel and Koolhaas 2004). The redox potential of the soil solution is determinant for the behavior of chemical substances, microbial activity, exchanges with other environmental compartments, and biological mechanisms such as nutrient uptake and toxicity (Alloway 1990a, Bartlett and James 1993, Bourg and Loch 1995, Contin et al 2007, Grundl 1995). The redox potential of the soil column will vary greatly over small to medium spatial scales, from microconditions within soil aggregates to changes from aerobic to anaerobic conditions with soil depth (Bartlett and James 1993, Förstner 1998, Rossel et al 1997, Yaron et al 1996). The redox conditions are especially important for speciation of metals in the soil (eg Alloway 1990a, Bartlett and James 1993, Yaron et al 1996), and the influence of redox potential on the degradation of organic substances is non-negligeable (Doelman 1995, Förstner 1995, Yaron et al 1996). Natural and anthropogenic variations in chemical composition will influence the suitability of the medium or site for habitat purposes in the following ways: toxic effects of substances, nutrient availability, changes in pH, and changes in soil structure (Förstner 1995, Ponizovsky et al 2006, van Gestel et al 1995, Schulin et al 1995, Stigliani 1995).

This parameter is assessable via chemical analysis of field samples. Redox measurements are difficult to carry out, and their accuracy is questionable; however, chemical analysis can give an impression of the redox potential via the presence/absence of characteristic chemical species. Measurement of pH can proceed using different solutions (H_2O, KCl, 0.01 M $CaCl_2$); if regional monitoring or comparison between sites is the goal, care should be taken that the same solution is used to measure pH in all sites.

Effect of this parameter → on this parameter ↓ is:	Chemical composition
	🟩
Microbial activity	limiting conditions
Chemical composition	🟩
Mineralogy	mineral formation, dissolution, CEC
Hydrology	>
Land use	legal requirements
Substance availability	limiting conditions
Soil age/history	>
Vegetation	limiting conditions
Specific composition	limiting conditions

Bioturbation	limiting conditions
Toxicity	limiting conditions
Connectivity	limiting conditions
SOM content and composition	persistence
Season	>

Substance availability:

Substance availability describes the relationship between the chemical substances in the soil matrix and the organisms living therein. Availability is a relative term, and in order to be meaningful the relationship must be specified. Substances are available with respect to an organism or biological process; the relative nature of substance availability is the primary reason it will exhibit such great variation at spatial and temporal scales.

Substance availability is governed by properties of the substance, the medium, and the organism or biological process. Substance availability is especially important to take into account when considering the simultaneous roles of soil as repository for waste effluents and as habitat for terrestrial ecosystems. The substances exhibiting variable availability include nutrients, trace elements, and organic molecules. Unavailable substances are present, but are prevented from entering into contact with organisms due to the composition and structure of the soil matrix.

The consideration of substance availability comprises three different levels (Vijver 2005): chemical availability, biological availability, and toxicological availability. Chemical availability is related to soil factors controlling the mobility and speciation of substances in the soil. These factors are the particle size distribution and composition of the mineral phase (mineralogy), the content and composition of organic matter, and the chemical composition of the soil solution (Jonker et al 2004, Lofts et al 2004, Marinussen 1997, Morel 1997, Peijenberg 2002, Sauvé et al 2000, Spurgeon et al 2006, Sukreeyapongse et al 2002, van Gestel and Koolhaas 2004). Other factors that can modify availability include modifications of hydrology, land use, and vegetation (Christensen 1984b, Sauvé et al 1997, van Gestel et al 1995, Yaron et al 1996).

It is important to distinguish the two separate but related concepts of mobility and availability. Mobility applies to both dissolved substances and particles, as particle-bound transport is an important mechanism of inter-compartmental exchange. If substances are mobilized in the liquid phase, they can experience increased uptake by organisms via the soil solution as well as increased translocation in the soil column via leaching and erosion. If a mobile substance leaves the soil area of biological activity, it is no longer available for uptake from the soil compartment. On the other hand, immobile substances may be more available than mobile ones for organisms whose primary exposure route is via the solid phase, provided that conditions exist during the substance's residence time in the organism for the substance to leave the soil matrix and enter the organism.

Biological availability is related to the factors previously mentioned in addition to the spatial distribution of the contaminants in the soil, the home range of the organism, the mobility of the organism, the organism's ability to modify its environment, and the organism's diet (Hunter and Johnson 1982, Krivolutsky 1996, Marinussen 1997, Morel 1997, van Wensem 1997, Veltman et al 2007b). The primary rate-limiting step between chemical and biological availability is uptake by the organism (Vijver 2005). Thus, for a given soil matrix, organisms with different uptake modes will experience differing bioavailable concentrations (van Straalen et al 2005).

Toxicological availability is related to compartmentalization of substances within an organism. See below in the description of Toxicity for more information.

Substance availability is a dynamic parameter of soil ecosystems. The proportion of a substance present in the soil matrix that is available will vary over time as the environmental conditions change. There is empirical evidence to suggest an aging effect, whereby contaminants present for long periods become unavailable and therefore no longer contribute to toxicity of soils. However, there is also evidence to suggest that this aging effect is reversible if parameters controlling substance availability are modified (Buekers et al 2007, Christensen 1984b). This parameter is therefore difficult to assess. Much disagreement exists about the validity of predictions of availability based on chemical extractions using different solvents.

Effect of this parameter →	Substance availability
on this parameter ↓ is:	🟩
Microbial activity	cyclical relationship
Chemical composition	>
Mineralogy	>
Hydrology	>
Land use	legal requirements?
Substance availability	🟩
Soil age/history	>
Vegetation	>
Specific composition	>
Bioturbation	>
Toxicity	limiting conditions
Connectivity	limiting conditions via toxicity
SOM content and composition	degradation rate
Season	>

Microbial activity:

Microbial activity is the link between nutrient/element cycling and environmental conditions (Doelman 1995). The microbial groups include bacteria, archaea, and fungi. Many of these organisms are capable of existing in resistant forms that are viable but inactive. Therefore, while microbial composition of a given site may vary little, microbial activity of the site varies over different scales of space and time. Much of this variation will be seasonal, and thus cyclical. However, some variation will be due to differences in soil physico-chemical properties, which will vary even at small spatial scales (Tobor-Kaplon 2006, Yaron et al 1996); these variances in are due in part to the microbial activity itself.

The diversity of micro-environments available to microbes will have a strong impact on the potential detoxifying capabilities of the soil (Boivin 2005, Griffiths et al 2001b, Kuhnt 1995). Microbial activity is intimately related to microbial degradation of biogenic and xenobiotic organic compounds. This degradation depends on the availability of the degradable substances and the genetic potential of the microbial communities to degrade them (Miller and Herman 1997, Toffoletto et al 2005). In addition to carrying out the bulk of the degradation and nutrient cycling processes (Kuperman 1996), microbial organisms catalyze chemical reactions involving inorganic molecules (Hendricks 1997, Schultze-Lam et al 1995), and are capable of displaying stress-induced responses assessable at the community level (Boivin 2005, Griffiths et al 2001a, Hendricks 1997, van Beelen et al 2004). This parameter can be assessed via laboratory assays on field samples. Due to the large natural variability in bacterial responses, the evaluation of this parameter requires careful interpretation. Special attention should be paid to the effect of season when designing sampling campaigns.

Effect of this parameter →	Microbial activity
on this parameter ↓ is:	🟩
Microbial activity	🟩
Chemical composition	element cycles
Mineralogy	particle aggregation, mineral nucleation
Hydrology	>
Land use	>
Substance availability	immobilization
Soil age/history	>
Vegetation	mycorhizzae
Specific composition	predator-prey
Bioturbation	>
Toxicity	food-chain transfer, mobility, adsorption
Connectivity	>
SOM content and composition	degradation, composition
Season	>

Soil Organic Matter (SOM) content and composition:

The term soil organic matter is used to refer to “the non-living components [of the soil organic fraction] which are a heterogeneous mixture composed largely of products resulting from microbial and chemical transformations of organic debris,” (Hayes and Swift in Kumada 1987, p. 1). SOM content is typically expressed as a percentage, which can vary greatly between soil types, sites, and patches of soil within a site (Yaron et al 1996). SOM is of major importance for determining the structural and chemical properties of the soil, in both the solid and the liquid phases (Bourg and Loch 1995, Lacerda et al 1995, Schulin et al 1995, Sparks 1995, Yaron et al 1996). The chemical reactivity, persistence, and behavior of the SOM will depend on its composition (Baldock and Skjemstad 2000, Schulin et al 1995, Kumada 1987) as well as on the processes of degradation or enrichment of the soil in OM (Osté et al 2001, Schaefer and Schauerman 1990). These processes are dependent on the physical, chemical, and biological properties of the soil (Kumada 1987), as well as on human influence in the forms of land use and emissions (Salomons 1995).

Soil organic matter is determinant for the ion exchange capacity of the soil (Weng et al 2001, Weng et al 2002, Schulin et al 1995, Yaron et al 1996), as well as for the mobility of trace elements (Buekers et al 2007, Ponizovsky et al 2006, Sauvé 2002, Schulin et al 1995, Schultze-Lam et al 1995, van Gestel et al 1995, Weng et al 2001, Weng et al 2002).

The content of organic matter in a soil is assessable via laboratory measurements of field soil samples; analysis of the composition is difficult to carry out and is rarely done.

Effect of this parameter → on this parameter ↓ is:	SOM content and composition
	🟩
Microbial activity	energy and nutrient source
Chemical composition	degradation conditions, products
Mineralogy	aggregate structure, size, CEC
Hydrology	infiltration via porosity, tortuosity
Land use	>
Substance availability	adsorption, binding, absorption, uptake
Soil age/history	historical development
Vegetation	limiting conditions possible
Specific composition	degradation rates
Bioturbation	aggregate structure/size, food quality
Toxicity	via subst. availability
Connectivity	>
SOM content and composition	🟩
Season	>

Toxicity:

Toxicity is related to substance availability in that only available substances can create toxic effects. However, availability does not encompass the effects of a substance on an organism, since inherent biological attributes will also determine the response of an organism or population to a toxicant (Bouché 1996, Rainbow 2002, van Gestel et al 1995, van Straalen et al 2005, Vijver 2005). Toxicological availability describes the relationship between the concentration of a substance in an organism and the access of that substance to the sensitive targets within the organism. If an organism possesses sufficient mechanisms to detoxify or eliminate the substances it has taken up, no toxic effects are to be expected (Rainbow 2002).

Toxicity is determinant for the persistence of populations with time, which depends on the combination of survival and reproduction. Reproductive cycles are determined and influenced by many factors, only one of which is toxicity. If reproduction is possible but toxicity reduces the survival of juveniles, the population may not be sustainable (Crommentuijn et al 1997, Hendriks et al 2005). A similar result is obtained if toxicity does not induce mortality, but prolongs the reproductive cycle past the conditions that the juveniles can survive (Klok et al 2006). Toxicity can manifest at the level of the individual through direct acute effects, at the level of the population through reductions of reproductive fitness, and at the level of an ecosystem through elimination of the organisms responsible for the cycles of matter and energy that characterize them (Bengtsson 1998, Hendriks et al 2005, Laskowski et al 1996, Migula 1996, Pokarzhevskii 1996, Posthuma et al 1998, van Straalen et al 1989).

This parameter is difficult to assess in the field. Models that account for the spatial heterogeneity of contaminants, the spatial range and activity of organisms, the variations of hydraulic conditions, and the thermodynamic nature of chemical equilibria in soils will have the most success linking a measurable soil concentration to its predicted toxic effect.

Effect of this parameter →	Toxicity
on this parameter ↓ is:	🟩
Microbial activity	limiting conditions
Chemical composition	>
Mineralogy	erosion via vegetation
Hydrology	>
Land use	suitability
Substance availability	>
Soil age/history	>
Vegetation	limiting conditions
Specific composition	limiting conditions
Bioturbation	limiting conditions
Toxicity	🟩
Connectivity	>
SOM content and composition	degradation rate
Season	>

Key parameters: Synthesis

All of the parameters described above can be modified by site use and/or management by humans, with the exception of season. The sensitivity of each parameter to human modification is not equal. The functions governed by the above parameters are therefore differentially sensitive to human influence. While this list may seem very long and nearly exhaustive, maintaining categories of this nature and using descriptive variables of field surveys or sampling campaign makes data more comparable between studies and researchers, and facilitates data inclusion into large databases (Bouché 1996).

It is the interactions between the parameters described above that generate and explain contaminant behavior and the ecological impacts of soil contamination. These interactions are rarely simple and uni-directional; rather they often involve cyclical or iterative phenomena creating complex patterns of interaction. They can be explained to a certain degree in a hierarchical arrangement because some parameters that occur at a large scale are not influenced (or only very tangentially or indirectly) by parameters occurring at a small scale. For example, the season of a site will influence all the other parameters; however, no other parameter will influence the season. Interactions between parameters may influence responses to seasonal stimuli, thereby modifying seasonal influences; but the forces responsible for creating seasonal phenomena are outside the domain of influence of site-specific parameters.

The interactions between parameters are also responsible for assessable soil attributes such as ion exchange capacity, soil fertility, soil texture, and soil horizon structure. One of the attributes most often considered when addressing the issue of metals in soils is the cation exchange capacity (CEC). This describes the total number of exchangeable cation charges expressed in terms of chemical equivalents per unit mass of soil particles (Hillel 1998). This attribute can be measured or estimated according to different procedures. It can be measured on a field soil sample, or estimated based on the proportion of different components (mineral particles, SOM) contributing to its total value (if the CEC of the individual components is known). In field situations the CEC will deviate from an estimate made based on summing the soil constituents based on three main reasons: 1) soil constituents interact with each other electrostatically, whereby each occupies some of the other's charged sites, 2) living and non-living microbial surfaces influence the soil CEC, but this is rarely accounted for in bulk soil samples analyzed in a lab, 3) SOM and Fe- or Al-oxides have charges that vary with soil pH (Beveridge et al 1995, Janssen et al 1997a, McBride et al 1997, Schultze-Lam et al 1995). Soil CEC is an example of a soil property strongly contributing to the behavior of metals in soil whose value is determined by the interactions of many of the key parameters described above (mineralogy, SOM, microbial activity, chemical composition).

4. The most relevant indicator for the assessment of soil quality

Biological indicators

Indicators for the assessment of soil quality provide information about the physical, chemical, and biological status of the soil ecosystem. Bioindication is an attractive diagnostic and monitoring tool because living organisms integrate the effects of biotic and abiotic conditions over time and space. Their presence, absence, abundance, dominance, and populational status give assessors powerful clues regarding the soil quality, and may provide a quantitative idea of the impact of the contaminated soil on the soil organisms. Inhibition of growth caused by the unique mix of pollutants and soil/site characteristics may indicate to what extent pollution conditions exclude species from the site, or eliminate present species over a specifiable time period.

The choice of organisms to use as indicators will depend on the subject matter requiring indication; this is especially true as regards scales of space and time. There is a relationship between the size of an organism and the length of time of effects of pollution: small organisms will be informative as regards acute pollution at small spatial scales, whereas larger organisms will be informative as regards chronic pollution at larger spatial scales (Pokarzhevskii 1996).

Indicator criteria

Recommendations regarding bioindicators have been developed within the context of polluted soil ecosystems (van Straalen and Krivolutsky 1996). These recommendations include the use of indicator systems that reflect the heterogeneous nature of soil ecosystems as well as the need for predictions over long time scales. With these factors in mind, the following recommendation was formulated during a NATO Advanced Research Workshop on bioindicator systems for soil pollution as instruments for risk assessement: “An ecologically relevant bioindicator system should be based on information obtained from each of three categories: 1) invertebrate animals (macrofauna, mesofauna, nematodes), 2) microorganisms, 3) ecosystem processes (litter decomposition, rates of change in nutrient status, dynamics of soil food-webs)” (van Straalen and Krivolutsky 1996, p. 250).

In addition to these recommendations, a certain number of criteria have been retained for community-level indication and monitoring (Pearson in Spurgeon et al 1996):

1) taxonomy that is well known and stable

2) natural history that is well known

3) readily surveyable

4) higher taxa have broad geographical distribution over a breadth of habitat types

5) lower taxa must be specialized and sensitive to habitat changes

6) taxa must have potential economic importance

7) patterns of biodiversity must be reflected in other related and unrelated taxa

Criteria have also been retained for ecosystem processes (Edwards et al 1996):

1) must be critical to soil ecosystem functioning

2) must have well-established methods for assessing changes in rates or functions

3) must produce data readily interpretable in terms of overall impact

An indicator system

The indicator system described above, and corresponding to the above criteria, would give a quantitative answer to questions regarding the impact of contamination on populations living at the site. However, a comprehensive risk assessment must also account for plant species, which are responsible to a large degree for the biogeochemical signature of the soil ecosystem, and which define the potential for the ecosystem to persist over long time periods. Knowledge about which species are present, and their abundances, is necessary for site characterization and prediction of the site's fate over time. Although the numerical relationship between biodiversity and ecosystem function is not understood, it is understandable that ecosystem functions depend on the presence of its species (Bengtsson 1998, Dighton 1997, Griffiths et al 2001b, Hendricks 1997, Rossel et al 1997, Rost et al 2001).

Studies into soil ecology have uncovered several biological communities and functions useful for indication of soil quality. Information about these communities can be combined with spatial information on contaminant location to assess ecological risk (Colgan et al 2003, Jepson 1997). These include nematodes (Korthals et al 1996, van der Wurff et al 2007), earthworms (Bouché 1996, Colgan et al 2003), macro-invertebrates (Spurgeon et al 1996), substrate-use profiles of microbial communities (Boivin 2005, Griffiths et al 2001a), soil enzyme assays (Edwards et al 1996, Rossel et al 1997), litter decomposition (Edwards et al 1996, Hendricks 1997, Kools 2005, McGrath 2002), and recovery time of microbial function after stress (Boivin 2005, Griffiths et al 2001a, Kools 2005, Rossel et al 1997). Biomass and soil respiration are two insensitive characteristics with little indicative ability when evaluated separately (Kools 2005, Tobor-Kaplon 2006, Traas and Aldenberg 2007). When related to each other, for example in a metabolic quotient, they yield more information (Hendricks 1997, Tobor-Kaplon 2006); microbial activity can also be indicative when in the context of a microbial toxicity assay (Kahru et al 2005).

5. Field data/ minimum dataset

Need for standardized methods

Soil is a complex and dynamic environmental compartment with unique characteristics such as significant temporal inertia, spatial heterogeneity, and multifunctionality with respect to human use (Nortcliff 2002). A soil ecosystem is the result of an integration over time and space of the physical, chemical, and biological processes which have contributed to its formation and current state. From this perspective, each soil ecosystem is a unique entity. While comparison between such unique entities may seem impossible, it is necessary from a management point of view. The best way to manage this diversity seems to be to adopt standardized methods, to ensure that data are as comparable as possible between sites. The importance of this grows as the spatial scale of risk assessment of contaminated sites grows. If the goal is to assess the risk associated with many sites, and to rank the sites according to urgency of remediation, having data that are comparable between different sites is of utmost importance.

The sampling density in the field can influence the representativity of samples. The spatial heterogeneity of soil systems means that large numbers of samples are needed in order to ensure that the desired limits of accuracy are reached (Wenzel and Blum 1995). There is much evidence to suggest that sampling season, methods of sample storage, and analysis methods can influence the results (de Rooij and Smits 1997, Wenzel and Blum 1995). Three primary areas of concern with respect to the data standardization for site characterization and site-specific risk assessment: 1) sampling times and frequency, 2) number and spatial resolution of samples, and 3) which observations are made/ which parameters are measured.

Sampling times and frequency

Seasonal variations in abiotic factors such as temperature and precipitation will induce variations in biological activity and physico-chemical parameters, in particular pH and pe (Bleeker and van Gestel 2007, Peplow and Edmonds 2005, Wenzel and Blum 1995). Site visits, ecological observations, and soil sampling should reflect these seasonal variations. This will most likely entail the need to conduct multiple sampling campaigns during different seasons.

The need for frequent sampling may seem problematic if there are many sites to assess. This is a key reason to design data storage and analysis according to well-organized databases. As the number of sites samples grows, data analysis will probably reveal patterns that permit optimalization of sampling dates and frequency. Thus, the benefits of approaching site-specific risk assessment from a coherent, regional perspective become apparent.

Sampling density

The detection and description of contamination will depend on the size of the sampling grid due to the spatial heterogeneity of soil constituents (Marinussen and van der Zee 1994). If the sampling grid is too large (> 0.5 km) it is probable that contamination will not be detected (Hopkin et al 1989). The smallest soil sample should be large enough (> 10 g) to be representative of the point it is intended to be measuring (Sheppard 1997).

The depth of measurement should be relevant to the behavior and habitat of the organisms being considered (Veltman et al 2007b, Wijnhoven 2007). Observation of the soil profile may indicate whether there is clear horizon structure. Soil profiles that are well-homogenized in the zone of biological activity may require fewer samples from different depths. It is recommended that the variation with depth should be investigated at each site in order to determine the sampling depth necessary for representative characterization.

Which parameters to assess?

Site visits should combine ecological observations and sampling campaigns. There is some benefit to measuring pH in the field (as well as in the lab) because storage and drying of samples can alter pH (Wenzel and Blum 1995).

The importance of standardized sample analysis is especially crucial in the laboratory analysis phase because different protocols exist; however, results are only directly comparable if the same protocol is used in all analyses (de Rooij and Smits 1997). Scientific debate exists as to which methods of extraction are most appropriate for determining metal concentrations and other physico-chemical parameters (Bermond and Benzineb 1991, Houba et al 1996, McBride et al 1997, Nortcliff 2002, Wenzel and Blum 1995). There is probably not one definitively correct answer; what seems more important is comparability. Therefore, regardless of which method is chosen, the same one should be used for all sites and all samples.

Table 2 gives an example of the information making up a minimum dataset for multiple scale site-specific risk assessment. Some of the information required can be obtained during a field visit, some would be able to be completed after laboratory analysis, and some would require expansion, such as the data regarding the site's specific composition.

Table 2. Example field visit data sheet including key parameters

Site identification	Site name, sample point	RV-10-23
Season	sampling date	January 10, 2008
Hydrology	water level (cm from surface)	0
Soil age/history	soil type	histosol
	soil profile depth (cm)	100
Land use	land use category	grassland, pasture
Connectivity	closed/open ecosystem	open
Mineralogy	soil texture	peaty sandy clay
	percent clay	11
Vegetation	vegetation type	grassland
Bioturbation	tunnels, burrows observed?	yes
	estimated quantity (g/sq.m)	20
SOM content and composition	OM content (%)	30
	soil color	black
Chemical composition	pH	5,5
	redox state (Eh)	-3
	total conc. Cd	
	total conc. Cu	

	total conc. Pb	
	total conc. Zn	
	total conc. SO4	
	total conc. PO4	
Substance availability	available Cd	
	available Cu	
	available Pb	
	available Zn	
Microbial activity	bioassays	
Toxicity	bioassays	
	toxicity Cd	
	toxicity Cu	
	toxicity Pb	
	toxicity Zn	
Specific composition	plant species	
	earthworm species	
	nematode species	
	collembola species	

6. Questions & Answers

Is the method being developed in this report supposed to replace existing risk assessment methods such as the Triad approach?

No. This integrated ERA-LCA method is meant to provide a supplementary tool for decision-makers wishing to evaluate the impacts of their site management strategies at multiple scales of space and time. Existing site-specific assessment approaches such as ERA provide a punctual indication of the risk associated with a contaminated site, but do not provide decision-makers with a framework to evaluate the ecological implications of their management decisions at a scale larger than that of the site under consideration. The method being developed here is meant to expand on the existing risk assessment methods.

How does the indicator system described here relate to current methods?

Ecological observations are part of the Triad approach, forming one of the three Lines of Evidence used to compile the risk indicator (Jensen and Mesman 2006). Bioindicator systems are a specific kind of ecological observation. The bioindication system recommended here gives suggestions as to which ecological observations have the greatest potential for indication of ecological impact at temporal and spatial scales larger than the site under investigation.

Where in the decision-making process does the method described here come into play?

The method being considered here can be applied at multiple moments of a site-specific risk assessment or during the landscape management decision-making process. The method being developed will be useful in the planning stages of a site-specific risk assessment, to help managing authorities plan data collection and site evaluation. It will also be useful to prioritize risk reduction efforts in the face of a large number of contaminated sites. Finally, it will be very useful for the evaluation of remediation options applicable to sites for which remediation is the chosen management option.

Does the method being described apply to the decision-making process for sites with forms of ecological degradation besides contamination (salinization, acidification, etc.)?

Not yet. The current stage of development will be applicable for the contamination of soil ecosystems by inorganic trace elements. However, the long-term goal in the development of this method is to be able to quantify the ecological impact of different categories of soil degradation.

7. Conclusions and next steps

The goals of this report were to qualitatively describe the key parameters for multiple scale risk assessment (Ch.3), select the most relevant indicator for the assessment of soil quality (Ch. 4), and to present the minimum required dataset for site characterization (Ch.5).

The parameters described thus far provide the theoretical basis to describe and predict contaminant behavior with respect to the abiotic and biotic components of a soil ecosystem. The next steps in the development of this integrated decision-making tool are to make explicit the interactions between the soil properties, organism exposure to contaminants, and toxic effects at the ecosystem level.

The next steps in the current project involve:

- quantitative indication of site-specific partitioning of trace elements over different soil constituents under different environmental conditions (pH, soil moisture content)
- description and quantification of the soil behavior of As, Cd, Cr, Cu, Ni, Pb, Zn
- quantification of the interactions between the soil matrix, soil organisms, and toxic effects
- application of the method within the context of sustainable development illustrated using a case study of the Ronde Venen, the Netherlands

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D3.1 Description of the key parameters for biological and chemical availability of cations and their relation with time,

D4.2 Application of the sustainable land management approach to the Ronde Venen case study,

summary of MuSA Workshop (September 22-23 2008)

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Summary

This report describes the parameters of the soil system that are responsible for the solubility controls of metals, and ultimately their biological and chemical availability. Chemical biological, and ecological factors with a strong influence on the solubility and mobility of Cadmium, Copper, Lead, and Zinc are summarized from the relevant literature. The complexity associated with the soil matrix and the terrestrial ecosystem it supports are illustrated using examples of how the influences of multiple scales of time, space, and biological organization will modify metal behaviour in soil over both short and long time periods. In the context of Sustainable Land Management, and in the special situation of Risk Based Land Management, the concepts related to metal solubility and mobility are illustratedusing a case study. The historically metal-contaminated Demmerik polder site located in the Dutch municipality of de Ronde Venen is used to illustrate an approach to the assessment of contaminated site management options based on management scenario evaluations. Three main management scenarios are evaluated: change nothing in the current management, excavate the contaminated soil, and use vegetation to remove metals from the soil over medium to long time scales (phytoremediation). The effects of phytoremediation are further explored by considering different plant species. Management scenarios are evaluated by two environmental assessment methods: Life Cycle Assessment (LCA) and Ecological Risk Assessment (EcoRA). The results of both assessments are combined to provide recommendations for site management strategies that incorporate concerns for sustainability into the decision-making process. However, interpretation of the results is not strictly a scientific question, and the choice of the most appropriate management strategy ultimately depends on the priorities of the decision making group. Due to the interdisciplinary nature of this multiple-scale combined assessment, efforts have been made to include involvement of researchers, regulators, and industry members in the development of an innovative decision support framework. To this end, an interactive workshop provided the opportunity for interested parties to come together and collaborate. Special focus was placed on efforts to examine existing knowledge regarding four principle themes, which are the spatial differentiation of environmental impact assessment, biodiversity, ecotoxicology, and the bioavailability and mobility of metals in soils. The examination of each of these themes from a perspective of both LCA and EcoRA resulted in the identification of areas where these tools overlap, areas where they complement each other, and areas where they each need improvement. Overall, the MuSA project has been able to provide some insight into how the simultaneous consideration of local and global impacts of contaminated site management can inform decision-makers and stakeholders as to the short and long term effects of management strategies. Further research efforts are indicated, with strong recommendations to continue the fruitful exchanges that result from an interdisciplinary approach.

Preface

SNOWMAN research and MuSA

SNOWMAN is a transnational research program in the field of soil-water-groundwater systems with respect to contamination. MuSA is one project within the SNOWMAN research network. The focus of the MuSA project is the development of a decisionsupport tool combining the complementary analyses of Ecological Risk Assessment (EcoRA) and Life Cycle Analysis (LCA). MuSA especially responds to a need of soil resource management authorities for scientifically sound methods permitting the ecological risk assessment of contaminated soil at spatial and temporal scales beyond those employed currently. The current study recognizes the need for decision-support tools that can quantitatively evaluate the ecological impacts of site management strategies. Emphasis is placed specifically on contaminated sites; however, the theoretical framework could be applied to site management of non-contaminated sites as well, for authorities evaluating development plans at a regional or national level. This report contains Deliverables D3.1 and D4.2 of the MuSA project, as well as a section summarizing the output of a workshop organized by participants in the MuSA project. The workshop took place on September 22 and 23, 2008 on the campus of Vrije Universiteit Amsterdam.

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1. Introduction

The goal of the project MuSA was to explore the link between two existing methods for evaluating the ecological impacts of soil contamination by metals. The two methods under consideration are Ecological Risk Assessment (EcoRA) and Life Cycle Assessement (LCA). Metal-contaminated soils pose management challenges, especially when concerns about the sustainability of management strategies enter into the decision-making process. Owners and managers of parcels of land whose soil is characterized by high levels of substances suspected (or known) to be present in concentrations posing toxic risks to organisms, natural resources (crops, water supplies), and/or human populations are required to evaluate (or have evaluated) and investigations are carried out in an attempt to determine if the source-pathway-receptor chain is complete, and thus, if there is an actual risk (NICOLE 2002). In cases where these risks are revealed to be non-negligible, choosing between management strategies designed to manage and/or reduce risks is rarely straightforward. Each possible management strategy has its benefits and drawbacks. Activities carried out within the framework of contaminated site management will have impacts on the local and global environment. The systematic comparison of these impacts from different points of view may provide contaminated site managers with necessary support for decision-making process. LCA and EcoRA are two tools that can provide such support. Users of LCA and EcoRA include business and governmental entities motivated by the desire and/or legal obligation to consider the environmental consequences of their decisions regarding contaminated land management. LCA provides an attractive framework for comparing the impacts of various alternative methods for accomplishing the same goal; this goal is expressed in LCA terminology as the functional unit. In the case of contaminated soil, an example of a functional unit is a given volume of clean soil, and the alternatives being evaluated are the management strategies that could be undertaken to yield the desired result (see further for a more detailed explanation). The attractiveness of the LCA approach is related to the big-picture view it gives of the various impacts on human and ecosystem health, as well as to the way it is able to clearly identify which of a host of input parameters are responsible for the resulting impacts in the category(ies) of interest to the decision-makers. However, there are some questions with particular relevance for land management that current LCA methodology is unable to address satisfactorily. These include:

1) the quantitative nature of the toxic impacts of inorganic elements in porous media (soils and sediments)

2) changes (positive or negative) in ecosystem quality as a result either of multiple stressors (toxic substances, eutrophication, climate change), or of ecological restoration efforts (i.e. removal of toxic substances, adoption of conservation methods, habitat creation/alteration)

3) the quantitative relationship between local biodiversity loss and species extinction The first two of these three issues are often addressed at the level of a contaminated site within the framework of an ecological risk assessment (EcoRA). Many regulatory agencies have established guidelines for evaluating the sensitivity of sites based on their a) intended or current use, b) geographical context, and c) physical, chemical, and biological make-up (Cappuyns and Swennen 2007, Suter et al. 2000, Swartjes 1999). When the assessment of the situation is that substances present in the soil of a site constitute an *unacceptable* risk (a determination that is social and political in nature rather than scientific) many regulatory agencies require site owners to undertake remediation/restoration efforts to reduce the risk to acceptable levels. The nature of these efforts will be highly specific to the parameters of the system under consideration, including the nature and quantity of the substances present, the nature and quantity of the contaminated media, the nature of the site, the intended site use, and the site's socio-political context. The attractiveness of the EcoRA approach is that information is tailored to the site under consideration, and can include such aspects as the probable effects of toxic substances on animal populations inhabiting the site (permanently, or as part of a migratory schedule), the possible (or probable) behavior of contaminants over time, and the effects of various management strategies or scenarios on the quality of the site ecosystem. The downsides of this approach include the large inputs of time, expertise, and data necessary for such an evaluation, the limited application of the results to sites other than the study area, and the lack of a systematic description of how the site and activities carried out within it relate to the bigger picture, in an ecological/environmental and/or socio-political way. The third issue in the list above (relationship between local and global biodiversity) is not currently addressed by either an LCA or an EcoRA approach, because its resolution requires simultaneous consideration of small and large scales of space, time, and biological organization. Evidence to support this statement can be found in population biology (i.e. meta-population theory) (Nisbet et al. 1993), landscape ecology (i.e. patch dynamics) (Caswell and Etter 1993), agricultural pest management, and ecotoxicology

(Van Gestel 2008). The large number of sites (and large volume of contaminated media) to be addressed at the national level of every industrialized county indicates that a systematic approach is worthwhile. While comprehensive knowledge could be gained by undertaking EcoRA's of each suspected contaminated site, such an approach would be very costly in terms of time, money, and resources. In the event that environmental quality standards are not met, extensive remediation efforts would be required under the existing legislation in many countries. Decisions about remediation and environmental quality measures are often bounded by budgetary concerns. One opinion is that this is short-sighted and unjustified. In industrialized countries, it is not a question of there being enough money; it is a question of how the money is allocated. These are political, not scientific decisions. Transparency in the political process may go a long way towards improved environmental quality, if access to information results in public pressure in support of increased budgets for conservation/nature preservation. However, as long as the scientific evidence is inconclusive regarding the role of biodiversity in the provision of ecosystem services this question remains one of opinion, personal belief, and moral code (and therefore subject to hot debate).

However, other reasons not to pursue intensive remediation activities exist that have greater ecological justification. In the conceivable situation where remediation activities will result in the destruction or long-term alteration of the site ecosystem (e.g. due to excavation and thermal treatment of contaminated soil solids, or phytoremediation that replaces existing vegetation with different plant species), the question of whether or not remediation is an improvement is justified. This is illustrated very clearly in the case of accumulation of inorganic pollutants (metals and metalloids) in porous media (soils and sediments). There is a body of evidence to suggest that many inorganic substances present in porous media are either inert with respect to the biological cycles taking place there, or that their participation in said cycles does not disrupt (to an unacceptable degree) the ability of the ecosystem to provide the services required by human populations. At the same time, there is a growing awareness at the regulatory level that applying short-term thinking to the management of natural resources has resulted in a diminished environmental capacity to provide ecosystem services. Simultaneously, public concern about ecosystem quality, responsible environmental stewardship, and the negative side-effects of modern industrial processes is steadily growing. This boils down to a sort of standstill situation: if the inorganic substances are indeed inert, and permanently fixed within the porous media, they may not pose an unacceptable risk to populations (human, plant, animal, or microbial) and can therefore be tolerated. However, in cases where the migration of substances cannot be excluded (for example, migration from soil to groundwater), and/or if the substances are demonstrably involved in biological cycles with negative repercussions on ecosystem quality, action should be taken. The remaining task is to differentiate between the different cases. Inorganic substances occupy a unique place in the decision-making process regarding environmental quality for a number of reasons. Because of their elemental nature they are completely non-degradable and therefore persistent. Many inorganic elements are necessary for biological activity and are described as essential for life; however these elements can also be toxic. Some of the same elements, as well as many not considered essential for biological activity are essential for the industrialized activity on which the global economy depends. It is inconceivable that modern societies would eliminate the use of metals, metalloids, and their various compounds. This complicated situation demands thoughtful consideration if a balance is to be struck between economic activity, social justice, and environmental quality as the mandate of sustainable development requires. The outputs of this project that are included in this report are the review of the current LCA and EcoRA approaches to metal-contaminated soils with an emphasis on considerations of metal bioavailability and mobility, a description of possibilities for a multiple-scale modeling approach, a synthesis of the applications of concepts employed throughout the project to a metal-contaminated terrestrial site in the Netherlands, a description and summary of the international workshop that took place at Vrije Universiteit Amsterdam to address some key topics related to a possible integrative approach, and conclusions and recommendations for future research efforts.

2. Two methods: LCA and EcoRA

The focus of this project has been the integrative use of EcoRA and LCA in the decisionmaking process regarding contaminated land management. The subject of the similarities and differences between these two approaches deserves proper treatment, and is the topic of a separate document. However, due to the fact that these two approaches are discussed at length in the current document, it seems appropriate to give a brief introduction to each.

EcoRA methodology: approach to space and time, and information output

EcoRA is "A process of collecting, organizing, and analyzing environmental data to estimate the risk of contamination for ecosystems" (Jensen and Mesman 2006). In the case of pollution that is already present, it is more precise to speak of the assessment/estimation of damage rather than risk, since risk implies the consideration of the probability that an adverse event will occur (NICOLE 2002). The basis for damage quantification is a comparative assessment between the contaminated site and a reference site, which by definition is considered to represent an undamaged condition. One of the main principles applied in EcoRA is the designation of the source-pathway-receptor paradigm (NICOLE 2002). In the case of contaminated soils the soil is the source, the exposure route is the pathway, and the sensitive target is the receptor. If any of these elements is missing from the situation under evaluation, risk (damage) does not exist. Furthermore, risk management strategies may include interrupting any of the links between these elements by excavating the contaminated soil, manipulating soil chemistry conditions to reduce uptake, or by eliminating the habitat function of the soil (by paving, for example). The designation of one (or more) sensitive target(s) is thus explicit in an EcoRA (Suter et al. 2000), and the effectiveness of the risk management strategy will be evaluated with respect to the sensitive target of interest. These targets may include general soil-dwelling flora and fauna, ecosystem processes (such as nutrient cycling), specific organisms of interest (such as an endangered species), or other environmental compartments (for example, groundwater supplies) (Suter et al. 2000).

Another main principle is the delimitation of the site under consideration, which gives a spatial aspect to a site-specific EcoRA. This delimitation may correspond to a legal rather than an ecological boundary (for example, the owner of a parcel of land is required to demonstrate that the soil of said land is not contaminated to an unacceptable degree). Further spatial aspects related to EcoRA include the determination of the spatial extent of contamination compared to the sensitivity of site based on current or future land use. For example, in the Dutch system, there is a direct link between the land use category (natural, residential, industrial) and the perceived ecological sensitivity, such that larger spatial extents of contaminant concentrations in exceedance of environmental quality standards are acceptable in areas of lower ecological sensitivity (Rutgers 2008). Temporal considerations are incorporated into an EcoRA to a limited degree; in some cases (for example, risks of contaminant migration) the notion of time is explicitly considered in determining whether contaminants pose an unacceptable risk to ecosystems or other sensitive targets. For example, in the Dutch risk assessment framework, soil contamination is considered unacceptable if it is likely to spread in a time period of ten years or less (Stefan Kools, personal communication). However, long time scales are not yet explicitly incorporated into current EcoRA methodology. The information generated by an EcoRA (when applied to contaminated soil) is an answer to the question: do existing contaminant concentrations pose an unacceptable risk to the designated sensitive targets? If the answer is yes, the next question to be evaluated is: which management strategy reduces the identified risk to an acceptable level?

LCA methodology: approach to space and time, information output

LCA is a process of analyzing the emissions, resources, and energy consumption related to the provision of goods and services throughout the entire life cycle of the good or service. One step in an LCA is the LCIA, or Life Cycle Impact Assessment, which estimates the relative impacts on various endpoints (human health, climate change, ecosystem quality, and resource depletion) of the emissions related to the production or use of any good or service. The good or service can be anything imaginable; all that is required to perform an LCA is the definition of the functional unit. The functional unit provides a means to compare in a consistent fashion different ways of achieving the desired good or service. As an illustrative example, it would not be appropriate to ask the question "Which has a higher impact on the environment, a can of soup or a hermetically sealed box of soup?" because the two soup containers may contain different volumes of soup, and results that differ may do so due to this volume difference. To correct for this, a functional unit is defined as, for example, one liter of soup. Following this designation, the amount of packaging of each sort (can, box) that is necessary to contain (or deliver) the functional unit can be compared in a consistent way, since the good they are delivering (one liter of soup) is identical, and any differences in environmental impact due to the inputs necessary to produce the containers are related to the containers themselves, rather than to the amount of soup supplied. When the question at hand is the evaluation of different management options related to contaminated soil, an example of a functional unit is one cubic meter of soil with contaminant concentrations equal or inferior to the legal concentration limit (for more details, see the Master's thesis of Francesca Gambazzi (Gambazzi 2008). The LCA approach provides a quantitative, comparative estimate of the relative contribution of each part of the process of providing a good or service (i.e. emission of a chemical substance, energy consumption, consumption of non-renewable resources) to the overall environmental impact expressed as indices for the four endpoints listed above. The LCIA approach to spatial and temporal aspects of impact assessment has evolved over time. In the original methodology, impacts were calculated based on aspatial multimedia (air, water, soil, sediments, plants) models that were strictly theoretical and bore almost no relationship to the physical environment. Concentrations in each environmental compartment (air, water, soil, sediments, plants) of substances emitted in the process of producing or using a good or service were calculated based on steady-state assumptions using mass-balance equations taking average transfer functions (atmospheric advection, infiltration, runoff) as inputs. Implicit in the steady-state assumption is the notion of an equilibrium being reached as time approaches infinity; this results in impacts being integrated over an infinite time scale. Recent developments include linking the multimedia models used in LCIA to spatial data taken from the physical environment; this allows users to incorporate data regarding location-specific parameters, such as precipitation rates and human population density into impact calculations. The result of these developments is that emissions into more or less sensitive areas will modify impact calculations (although all impacts remain hypothetical). In these spatially differentiated models impacts are still expressed as time-integrated values; however, recent developments in the interpretation of LCA results exist that separate impacts likely to occur in the short term (less than 100 years) from those likely to occur in the long term (greater than 100 years) (for an illustration see Master's thesis of Francesca Gambazzi). The information generated by LCA (when applied to management options for contaminated soil) are answers to the questions: which management strategy will generate the highest level of impact on the four endpoint categories (reported as separate categories) (human health, climate change, ecosystem quality, and resource depletion) when all the inputs necessary to carry out the management strategy are considered? LCA therefore enters the decision-making process after a site-specific EcoRA has determined that existing contaminant concentrations are too high to be acceptable.

Four themes that influence the integration of EcoRA and LCA: spatial differentiation of impacts, biodiversity, ecotoxicology, and metal mobility and bioavailability

There are four themes that may represent significant overlap in an integrative approach that involves EcoRA and LCA in the management of metal-contaminated soil. These are the spatial differentiation of impacts, biodiversity, ecotoxicology, and metal mobility and bioavailability. Because these topics are dealt with differently in EcoRA and LCA, the two approaches may yield different answers to the question of which management strategy to adopt with the goal of reducing the impacts (or risks) associated with the management of metal-contaminated soil. Due to their high degree of relevance regarding the question of the feasibility of an EcoRA-LCA integrated approach to contaminated land management, these topics formed the basis of an international workshop aimed at identifying the state of the art and current perspectives for such an integration. This workshop took place at the Vrije Universiteit Amsterdam on September 22 and 23, 2008. More detailed information is given in Section 6 of this report.

3. Metals in soils: specifically Cd, Cu, Pb, Zn

Metals have received abundant attention in the scientific literature especially concerning risk assessment of aquatic and terrestrial environments. This project has focused on four elements (Cd, Cu, Pb, Zn) because of their relatively elevated concentrations in the soil of the study site, the abundance of literature regarding their behavior in environmental compartments, and their high global levels of use and emission (i.e. Morgan and Stumm 1991).

Treatment in LCA and EcoRA

LCA and EcoRA have slightly different approaches regarding the treatment of metals as toxic substances in terrestrial and aquatic ecosystems. Please note that this discussion is concerning the risk assessment of metals known to be present in various compartments, rather than the risk assessment procedures associated with setting environmental quality standards. LCA uses an equilibrium partitioning approach to determine the concentrations of metals in various environmental compartments based on the amount of substance emitted and average partitioning coeffecients for each element (Haye et al. 2007). Uptake of metals by animals destined for human consumption is modeled using a dimensionless bioaccumulation factor (BAF), whereas uptake *per se* of metals by non-food organisms (such as soil fauna) does not enter into LCA calculations. However, exposure is considered in a simplified fashion. The concentrations of metals calculated in each environmental compartment are taken to be the concentrations to which organisms are exposed. Ecotoxicological risks due to this exposure are evaluated based on a single HC50 value for each compartment, which is the geometric mean of chronic EC50 values evaluated for at least four phyla for a given substance (element) (Jerome Payet, personal communication). Ideally, theses toxicological data are taken from organisms inhabiting the compartment in question; however, paucity of terrestrial toxicological data often means that data from aquatic organisms are used to calculate terrestrial HC50 values. Uptake of metals by plants is considered differently for plants destined for human or livestock consumption and plants that make up the terrestrial ecosystem. Metal concentrations in fodder and produce consumed by humans are modeled using BAF's, whereas metal concentrations in plants forming the terrestrial ecosystem are not modelled (*N.B.* some controversy exists as to whether these concentrations are calculated and not reported in LCIA multi-media models, or whether they are simply not modeled; for the end-user of LCA the result is the same regardless). EcoRA has more flexibility when it comes to dealing with metals than LCA does. When EcoRA is applied to contaminated sites suspected or known to have metal concentrations exceeding legal threshold values, site-specific parameters known to be important for influencing metal behavior can be quantified. This quantification can range from relatively simple analyses of soil texture, pH, and total metal concentrations to more indepth investigations that include measuring metal concentrations in different compartments (above and below-ground plant tissues, animal body/tissue concentrations, sequential extractions, soil pore water concentrations and speciation) as well as measuring and/or modeling metal behavior in response to hydrological conditions, biotic interactions, and management strategies. The development of chemical speciation, 2- and 3-D transport, and food-web models provide end-users of EcoRA with powerful tools to reduce uncertainty associated with the diagnosis and prediction of metal behavior in terrestrial and aquatic ecosystems. However, the output generated by these models depends heavily on the quality of the input data, as well as the ability of the model to accurately describe real-world phenomena; this underscores the need for reliable field data to calibrate and validate these models. Of the two frameworks, EcoRA currently gives the most reliable information concerning metal bioavailability and mobility; however, this information is far from complete. Of primary concern regarding the management of metal-contaminated soils are the risks of spreading to neighboring aquatic compartments (surface and groundwater), risks of trophic accumulation and subsequent toxicity, and risks to human health due to a) ingestion of toxic concentrations of metals in food products and b) ingestion of soil itself (mostly a concern for children and animals whose meat or milk is destined for human consumption). On the other hand, LCA gives valuable information regarding the global effects of treatment options related to contaminated soil management. While an EcoRA may yield as output a recommendation to excavate contaminated soil based on concerns for the soil organisms, human inhabitants, or the aquatic ecosystem, it is unable to quantify how the effects of this excavation and treatment will extend to a larger spatial area than the initial contamination, thereby (potentially) endangering a larger potential human and nonhuman population.

Bioavailability

Of primary concern in porous media (soils and sediments) is the issue of the "bioavailability" of existing metal concentrations. This is a complex notion with as many definitions as there are groups attempting its description and quantification. Many researchers prefer to describe three separate notions: chemical availability, biological availability, and toxicological availability (Vijver 2005). This division provides a useful framework for fundamental research at the level of the individual organism (or cell in the case of microbes and unicellular organisms). However, depending on the question, these separations may be somewhat artificial, as the quantity of metal that is "available" for entry into biological cycles depends on the scale of time, space, and biological organization being considered (Van Gestel, in press). For example, many researchers take the dissolved (or extractable with various solvents) concentrations of metals in soil pore water to be the available fraction (e.g. Houba et al. 1996) These same researchers neglect to quantify or describe both the nature of the relationship between the metal concentrations in the soil solid phase and those in the liquid phase, and the replenishment rate for the soil liquid phase. However, it is well known that changes in capacity controlling parameters of the soil system (pH, redox, soil texture) will alter the solubility relationship and the replenishment rate (e.g. Van Gestel et al. 1995). Therefore, over medium to long periods of time, the available fraction does not remain constant (influence of time scale). An example illustrating the effect of the spatial scale is that of varying body concentrations of metals (Pb or Zn, for example) in animals inhabiting soil whose contamination is patchy rather than homogeneous (in soils, patchiness is the rule rather than the exception (Van Es 1993)). As the animal moves throughout the patchily contaminated soil it will encounter varying concentrations of dissolved and total metals; however, all of the metals that are taken up were by definition available. This uptake would have taken place via a combination of routes including cutaneous absorption and ingestion (Hobbelen et al. 2006b, Vijver et al. 2003). It would be impossible to predict the resulting body or tissue concentration in the animal based on sampling the dissolved (or extractable) metal concentrations in any single patch; in effect, the animal will average these varying concentrations over the span of its lifetime (Hobbelen et al. 2006a, Oste et al. 2001, Marinussen 1997). An example demonstrating the influence of the scale of biological organization is that of the movement of lead through terrestrial trophic chains. Lead that is taken up by an woodlouse inhabiting contaminated soil will not be taken up by a predator consuming the woodlouse because the lead tends to concentrate in the uneaten parts of the animal (carapace); thus, the amount of lead that was available to the woodlouse is not the same as the amount that is available to the woodlouse's predator (Hunter and Johnson 1982). Therefore, the "bioavailable" fraction for one organism is not the same as the "bioavailable" fraction for another organism; and if the scale of biological organization under consideration is the ecosystem (including trophic relationships) the "bioavailable" fraction is not constant. Furthermore, considerable disagreement exists in the scientific literature as to the best method for determining the "bioavailable" fraction (see e.g. Peijnenburg and Jager 2003). Some researchers find the most significant correlations between body/tissue concentrations and "total" metal concentrations, whereas others find the most significant correlations between body/tissue concentrations and "dissolved" or "extractable" concentrations. These discrepancies are no doubt due to a combination of a) the complex interactions between the contaminant, the environment, and the organism and b) the wide variety of methods and approaches used to address the question. The notion of bioavailability is thus operationally-defined and is only meaningful if the following factors are explicitly described: substance under consideration, medium (soil, sediments, water), organism (or group of organisms), time scale, and effect under consideration (e.g. Van Gestel, in press). This last factor is related to empirical observations that accumulated metals do not necessarily contribute to toxic effects if effective detoxification or tolerance mechanisms exist (sequestration, immobilization, excretion) (e.g. Rainbow 2002, Van Straalen et al. 2005, Vijver 2005). Accumulation and transfer to other elements of the trophic chain will be related to a different sort of availability than intoxication. It is impossible to give a single definition of the "bioavailable" fraction that will apply to all organisms and effects in a given ecosystem. Despite there being no universal "bioavailability", when the necessary details are given, interesting results have been obtained by analyzing invertebrate uptake rates of metals from soils and/or food (Gimbert et al. 2006, Gimbert et al. 2008, Van Straalen et al. 2005, Vijver 2005). Uptake rates of various metals have been evaluated for snails, earthworms, and woodlice on a variety of soils. In many cases uptake rates (expressed for example in micrograms of metal per day) correspond well to CaCl2 metal extractions (Gimbert et al. 2006, Scheifler et al. 2008). Many questions remain concerning assimilation rates, elimination rates, the length of exposure necessary to reach a steady-state concentration, whether or not a steady-state concentration will be reached (depending on the metal, the organism, and the soil/food metal concentration), and the applicability of laboratory generated data to field situations where food availability and heterogeneous habitats are not controlled (Scheifler et al. 2008). However, measurements of uptake rates combined with analyses of soil and ecosystem properties contributing to soil pore water metal replenishment rates provide a link between the physico-chemical situation of contaminated soils and the exposure of many organisms to potentially damaging metal concentrations. Research in this area is promising and should be encouraged.

No discussion of bioavailability would be complete without mentioning two of the most widespread models in use in modern ecotoxicology, which are the Free Ion Activity Model (FIAM) and the Biotic Ligand Model (see e.g. Lofts et al. 2004, Koster et al. 2006, Parker et al. 2001). The basis of these models is that the free metal ions, and not the total solid or dissolved metal concentrations, are responsible for observable toxic effects (see e.g. Lofts et al. 2004). The biotic ligand model takes this reasoning one step further by incorporating the "protective" effects of non-metal ions (such as H+, Ca2+, Mg2+) into quantifications and predictions of toxicity. These models are based on observations that toxicity benchmarks such as the EC50, or LC50 vary widely when expressed as total or dissolved concentrations, but vary much less when expressed in terms of the activity of the free metal ion (Van Gestel and Koolhaas 2004). These models seem to perform well for aquatic media, and apply reasonably well to organisms for which exposure to metal ions is known to occur primarily through the skin or respiratory surface. However, these models perform significantly less well when applied to terrestrial organisms (see e.g. Koster et al. 2006, Parker et al. 2001, Spurgeon et al. 2006). Many exceptions to the FIAM have been noted in the literature (for an excellent overview see Campbell 1995) for terrestrial and aquatic organisms, including those known to inhabit the thin layers of water associated in permanence with soil particles (e.g. algae, bacteria). Moreover, there is strong evidence to indicate that the free ion is not the only chemical species capable of penetrating cells in unicellular and multicellular organisms (Campbell 1995, Collins and Stotzky 1989, Doyle 1989, Parker et al 2001). However, the question remains open as to whether metal complexes (such as MeCl2 complexes) indeed cross cell membranes, or whether the dissociation of the complex takes place exactly at the membrane surface. This question, while challenging and interesting on a fundamental level, is not resolved enough to form the basis of sound policy decisions (this researcher's opinion). In porous media, especially terrestrial systems, it has been demonstrated that cutaneous uptake does not suffice to describe body/tissue concentrations in organisms where this question has been investigated (Spurgeon et al 2006, Veltman et al 2007, Vijver et al. 2003), and that ingestion of soil, leaf litter, and other metal-containing food products are important uptake routes (Oste et al. 2001) inadequately described by these models. In order to avoid any confusion, and due to the ambiguous nature of the notion of "bioavailability", the following discussion related to the behavior of metals in terrestrial ecosystems will refer to the solubility relationship of metals between the soil liquid and solid phases, which is measurable according to accepted standard procedures, unlike "bioavailability" which currently has no widely agreed-upon standard definition. Furthermore, by focusing the discussion on this solubility relationship, the link between uptake by biota via soil pore water and mobility at the level of the ecosystem is more straightforward. Lastly, a discussion of the solubility relationship provides a logical framework to link the ecosystem factors responsible with the time scales of interest to contaminated site managers. The relationship between the total soil content and the dissolved concentration of any given element is a complex one influenced by many abiotic factors including: soil texture, soil particle size distribution, soil aggregate size/composition/distribution, soil mineralogy, site hydrology, site porosity (including macropores), and soil chemical composition (Appelo and Postma 2005, Hillel 1971, Lindsay 1979, Sposito 1981). Furthermore, biotic factors have a strong influence as well, and can be anthropogenic (such as land use (past and present), history of contamination, management strategies) and non-anthropogenic (such as effects of microbial activity, rhizosphere effects, bioturbation) (Ferris et al. 1989, Fest et al. 2007, Morel 1997, Olson and Panigrahi 1991, Romkens 1998). (For more details and specific references, see MuSA Deliverable D2.2/D3.2). Due to the heterogeneous nature of the soil medium, this relationship will vary over horizontal and vertical distances, leading to differences in dissolved concentrations throughout a site as small as one hectare (e.g. Kools 2005, Van Es 2003) and in different layers of the soil profile (Romkens 1998). Dissolved concentrations of metals will also vary over the course of seasons; this has been verified for some terrestrial sites where measurements have been taken often enough to demonstrate this effect (Van den Berg et al. 1998). It is impossible to describe a relationship between the solid and dissolved concentrations of metals that applies universally to all soil systems. Solubility controls of metals in soils vary according to the metal in question, the soil's total metal load (of a given metal, plus all other metals present), the soil's chemical composition (non-metal inorganic elements as well as organic substances), the soil's mineralogical composition (and the stability of mineral phases with fluctuations of soil moisture content, redox potential, and temperature) (see e.g. Elliot et al. 1986, Hesterberg 1998, Lofts et al. 2004, Gao et al. 1997, Mansfeldt 2003, McBride et al. 1997, Sauvé et al. 2000), and the biological activities taking place in the soil ecosystem. This emphasizes the need for site-specific information, as well as the difficulties associated with large-scale predictive modelling efforts (such as those used in LCA), because attempts to generalize parametrized models developed for a specific site to other, similar or dissimilar sites will result in only mediocre data fits (Römkens 1998). It also complicates models used for long-term prediction, as solubility controls of metals in soils can change over time from being controlled by first or second-order adsorption reactions at low to intermediate metal loadings, to being controlled by precipitation/dissolution reactions at higher metal loadings (Appelo and Postma 2005, Harmsen 1992, McBride et al. 1997, Sauvé et al. 2000). It is important to note that the notion of "dissolved" concentrations is not itself unambiguous. Metal atoms can exist in the soil solution in a variety of forms, associated to a variety of organic and inorganic ligands, or as free ions. The description of these forms is the goal of the great variety of chemical speciation models that exist, some of which are freely available for download and use. Furthermore, the line between the soil's solid and liquid phases is blurred by the somewhat continuous distribution of soil organic matter between solids, amorphous gels, and dissoved organic molecules (Gustaffson and Pechova 2003), as well as by the existence of multiple biological, organic, and inorganic colloidal phases present in the soil solution (e.g. Lofts and Tipping 2000). Efforts to explicitly include these colloidal phases in investigations into both aquatic and terrestrial systems are advancing rapidly, as scientific understanding and computational capacity increase (see e.g. Gustafsson and Pechova 2003, Lofts and Tipping 2000). This is very promising for evaluating the risks of vertical mobility of metals in soils (and possible migration to groundwater aquifers) and transport of metals to aquatic ecosystems via runoff/erosion, as well as organism exposure. This last point may be especially relevant in terrestrial ecosystems. Soil particles are surrounded by thin films of water, which are typically enriched in metal ions compared to the bulk soil solution (e.g. Appelo and Postma 2005, Sparks 1995). Uptake of metals by bacteria inhabiting this thin water film may represent a quantitatively important point of entry of metals into the terrestrial food chain, via e.g. bacterivorous nematodes feeding on these metal-rich bacteria (Geesey and Jang 1989, Olson and Panigrahi 1991).

Mobility

Mobility of metals in ecosystems can encompass both the movement of metal atoms between different compartments of a given ecosystem (soil solid phase, soil liquid phase, colloid phase, organism) and the movement of metals from a given site to neighboring sites (or compartments, or ecosystems). It is important to specify the spatial scale under consideration when discussing mobility. Furthermore, this notion is not considered in an identical way in LCA and EcoRA. In LCA the mobility of metals (or any other substance) is quantified based on transfer functions including atmospheric advection and deposition, erosion, infiltration, and uptake (modeled in an abstract way using bioaccumulation factors); however movement is uni-directional and non-cyclic due to the integration over long time periods and the equilibrium partitioning method employed. A site-specific EcoRA may consider multi-directional mobility, due for example to the soil hydrological regime, as well as cyclical patterns of mobility based on land-management interventions (e.g. dredging ditch sediments and depositing on land) or the biological cycles of the organisms making up the ecosystem (e.g. bioturbation). A concrete implication of these biological cycles is the variation in exposure and sensitivity of organisms depending on the season (Brekken and Steinnes 2004, Gimbert et al. 2008, Hunter et al. 1987a, Hunter et al. 1987b, Hunter et al. 1987c). At the spatial scale of a contaminated site, mobility of metals in the soil is related to fluxes of metals entering and leaving the site. These fluxes are primarily: atmospheric deposition, application of solid/liquified metal-containing products (e.g. sewage sludge, manure, dredged sediments), offtake via plant growth and removal, leaching to ground and/or surface water, and transport via erosion of soil particles. Uptake and removal by organisms is a possible flux that may be considered part of metal "mobility", this will depend heavily on the size of the site and the organisms inhabiting it. Uptake by earthworms, for example, is unlikely to represent a significant source of mobility outside of a site whose area is greater than the worm's typical home range (tens of meters, LaVelle and Spain 2003). Uptake by plants is also unlikely to represent a quantitatively important flux unless the plant biomass is removed from the site (e.g. phytoremediation, or removal by grazers who do not return the metals to the grazing site via feces). However, participation of soil metals in biological cycles via uptake by plants, animals, or microbes can affect mobility through effects on metal speciation and ecosystem properties. Concrete examples of the effects of organisms on field-scale metal mobility include macropores created by plant roots (Hillel 1998) and worm or mammal burrows (Wijnhoven 2007), as well as the possible contribution of ruminant fecal matter to increased dissolved organic carbon (DOC) levels in soil pore water (unverified), high levels of which have been demonstrated to increase vertical mobility of metals with a high affinity for soil organic matter (Pb, Cu) (Karathanasis et al. 2005, Linde et al. 2007, Römkens 1998, Steenhuis et al. 1999). On the other hand, some organisms may have the effect of stabilization of metals in the soil; for example, willows may heavily concentrate metals in their root tissues (Meers et al. 2007). This will retard metal mobility and may be an effective risk reduction strategy when protection of groundwater supplies is a top priority; however, these high concentrations in root tissues may also pose high risks to organisms that consume them. The effects of organisms on metal mobility is highly dependent on site-specific factors, and no meaningful generalizable conclusion can be made, unless it is that organisms will have a non-negligible effect on metal mobility in terrestrial ecosystems. One biological group that heavily influences the mobility of inorganic (and organic) substances in soils are soil microbes, specifically bacteria and archaea (for simplification, the following discussion will employ the term bacteria, however, this term is not meant to exclude archaea). Bacterial activity affects metal and metalloid (including cations and oxyanions) mobility in two primary ways: a) via creation and modification of redox zones in soil profiles and soil aggregates, and b) via the metal binding and accumulation by bacterial cells and cell walls leading to high local concentrations of metals and potential vertical transport as a bacterial colloid phase (Appelo and Postma 2005, Beveridge 1989, Beveridge et al. 1995, Hunter et al. 1998, Olson and Panigrahi 1991). Many bacterial cell walls have high binding affinity for metal ions (Beveridge et al. 1995, Doyle 1989. Ferris 1989, Geesey and Jang 1989). In many cases bacterial cell walls provide nucleation sites for metal-containing minerals, which may result in the precipitation of metastable mineral phases in soil (Ferris 1989, Ferris et al. 1989). This may increase or decrease the vertical mobility of metals in soils depending on soil physico-chemical conditions; it may also result in metal sequestration in chemical forms unlikely to enter trophic chains if metal concentrations are high enough to induce an avoidance response of bacterivorous organisms (unverified). If this is true, microbial accumulation of metals may represent a protective mechanism for soil ecosystems (also unverified). Although the current state of knowledge concerning the mechanisms and evolutive advantages of bacterial metal accumulation/mineral precipitation is in its infancy, there is a potential for using microbial scavenging of metal ions from soils, porous media, and solid/liquified waste products as an economically beneficial metalrecovery strategy. Developments in the understanding of soil microbial diversity (Van der Heijden et al. 2008), and identification of the microbial genera responsible for efficient metal removal may yield promising technological and ecological benefits.

Tools to quantitatively describe and predict metal bioavailability and mobility in soils

It follows from the above discussion that the description and prediction of the solidliquid- colloid partitioning, and speciation of metals in soil pore water will be useful in evaluating the availability of metals to terrestrial organisms as well as the mobility of metals within and between ecosystems (or environmental compartments). It should be noted that the separation of partitioning and speciation is somewhat arbitrary, because soil systems are dynamic and metastable, amorphous phases may be the rule rather than the exception (Linday 1979). Furthermore, analytical and modeling tools are advancing to the degree that the need to apply this separation is disappearing. However, in many cases this kind of simplification is useful. For example, it is well known that plant root uptake of inorganic substances is mediated by the soil pore water (it is not necessary at this stage to designate which chemical species are "available"). Therefore, if the question is to model plant metal uptake over the course of a growing season, the thorough description of the solubility relationship between the soil solid phase and the soil pore water is not strictly necessary. However, if the question is to predict how dissolved metal concentrations will vary over longer time periods (decades to centuries) with varying conditions such as pH or redox potential, this solubility relationship is of great importance. The most widespread tools to describe and predict partitioning and speciation include direct sampling and measurement of total and dissolved concentrations, sequential extraction methods, mineralogical observation (e.g. XANES), chemical speciation programs/calculations based on thermodynamic data, and linear regressions derived based on large ecotoxicology and soil property datasets. All methods have their strengths and weaknesses, and all are described in full detail in numerous sources; it is outside the scope of this project to give thorough descriptions. It is within the scope of this project to mention how these tools could be used in an approach to contaminated land management that integrates EcoRA and LCA. Direct sampling of both total (e.g. HNO3 extractable) and dissolved (e.g. soil pore water, or CaCl2 extractable) metal concentrations is a straightforward way to grossly characterize metal partitioning between the solid and liquid soil phases. The ratio of these two measurements can provide a site-specific partitioning coeffecient (Kd value) for each metal measured. While this approach is an oversimplification that does not account for many factors known to influence partitioning coefficients greatly, it nonetheless provides some information that is directly useable in simple multi-media models such as those employed in LCIA. Although the reliability of this approach remains questionable, modelers will run their models with or without reliable input data; this provides a motivation to give the least unreliable data when it is feasible to do so, and may reduce the uncertainty of predictions made by spatially resolved multi-media models at the regional or site level. Many sequential extraction schemes exist that employ different solvents, in varying concentrations, in attempts to quantify different pools of soil metals. The pools generally fall into the categories such as dissolved, organic-material bound, carbonate bound, and residual (although many variants exist). Criticisms of sequential extraction methods include the observations that metals can shift from one pool to another during the sampling (Wenzel and Blum 1995) and extraction processes (Bermond and Benzineb 1991); however, some studies have found useful relationships between the results of sequential extractions and metal uptake by plants (Wang et al. 2002) or the influence of management strategies (Van der Welle 2007). Due to the fact that the method employed will influence the results to the same degree as the site conditions, it is difficult to imagine the results of sequential extractions being applicable to a global LCA multimedia model. However, in the context of a site-specific EcoRA the results of a sequential extraction may be helpful in predicting the exposure of organisms to metals. Furthermore, if LCA is used to evaluate the impacts of different site management scenarios, the results of a site-specific sequential extraction may be extremely useful for predicting the rates of metal emission to neighboring bodies of water (for an illustration of this idea see the Master's thesis of Francesca Gambazzi). Sequential extraction methods may be most informative when combined with other methods of analysis (Néel et al 2007). Microscopic observation of soil minerals will provide useful information regarding the sequestration of metals in crystal lattices or the precipitation of metal-containing solid solutions (Néel et al 2007, Terzano et al 2007). If these precipitated phases can be identified, direct predictions are possible regarding the stability of these minerals in the face of changing soil chemistry conditions. Concrete examples are the precipitation of metals as either carbonate or sulphide minerals, the former of which will be soluble with decreasing soil pH, the latter of which will be soluble with oxidizing conditions (e.g. Peters and Shem 1995). This kind of observation is most applicable to EcoRA (as opposed to LCA) because of the highly site-specific nature of soil evolution. Many chemical speciation applications (computer programs, computer scripts) exist that predict the chemical form under which a given element can be found based on the input data and system description given by the user. These programs vary greatly in their interface appearance, their ease of use, the computer language employed, the forms of input and output data they can accomodate, and the degree of expert knowledge necessary to operate them effectively. However, they vary little in their fundamental approach to the calculations they carry out, which are universally based on thermodynamic data and mass-balance equations (for an excellent theoretical discussion see Sposito 1981). Of particular interest regarding questions of metal mobility in soils (and risks of metal migration to groundwater) are models that are able to combine partitioning, speciation, and transport calculations simultaneously (for example, PHAST); however, the complex nature of these models implies a certain level of expertise if their output must be reliable ("garbage in, garbage out"). It is feasible (while being demanding of data and time) to construct and run a model for a given contaminated site that is able to predict the effects of different management strategies on metal speciation and mobility to deeper soil layers or neighboring water (ground or surface) compartments. This can be done in the context of an EcoRA of a contaminated site. However, it is difficult to imagine a feasible modeling approach that could simultaneously describe movement between different soil layers (millimeters to meters) and movement between watersheds (kilometers) (the computational demand alone would be staggering). It is therefore difficult to imagine applying a highly resolved speciation and transport model to a large-scale LCIA, which by definition integrates emissions over large spatial scales. It is imaginable to define a "restricted" LCIA that could run a combined speciation and transport model calibrated for the (simplified, average) soil properties of a watershed, for example. The question remaining is whether or not such an evaluation is truly an LCA (or is it something entirely different, albeit highly valuable)?

Some authors have derived statistically significant regressions to predict free ion concentrations and/or partitioning coefficients for metals in soils based on large databases of soil properties (Lofts et al. 2004, Sauvé et al. 2000). These relationships are highly significant for some metals, and less so for others (typically, the behavior of Pb is difficult to predict). While this kind of statistical analysis is very valuable, it is of limited applicability directly in LCA, as in most cases the partitioning coeffecient and/or free metal ion predictions depend partly on the soil metal concentration. This implies an iterative approach to determine metal partitioning and concentrations; however, such iteration is not possible within current LCIA multi-media models, which employ an equilibrium partitioning approach to all emitted substances. These equations may be more useful in EcoRA, especially early in the investigative process for a suspect site. Field validation is necessary, especially if these equations are to be applied to soils with a) extremely high metal loads or b) conditions not encountered in the original datasets used in their derivation (e.g. extremely high levels of soil organic matter). It is appropriate to mention the well-known Hard and Soft Acids and Bases Principle (HSAB) when discussing metal speciation. The following summary is taken from Sposito 1981, which is highly recommended to readers wishing for a more thorough treatment. Furthermore, tables listing examples of hard and soft acids and bases can be found in many sources (e.g. Ross 1994, Morgan and Stumm 1991, Yaron et al. 1996). Briefly, this principle is derived from the classification of chemical species into groups exhibiting tendencies to form complexes of a similar nature based on a few inherent chemical properties: size, oxidation state, electronegativity, and polarizability. The main tenet of the HSAB is that hard acids tend to form complexes with hard bases, and that soft acids tend to form complexes with soft bases. Some elements exist that exhibit intermediate behavior; these are referred to as "borderline" (both acids and bases can be borderline). All of the bivalent metal cations are considered to be borderline acids, except for Cd, which is considered to be a soft acid (Sposito 1981). Soft and borderline bases (chemical species likely to form complexes with soft and borderline acids) likely to be important interrestrial systems include nitrite, sulphide, and sulphite (not an exhaustive list). Hard bases include oxygen, nitrate, sulphate, and phosphate.

While this principle is based on sound chemical observations and data, it is of limited utility in the case of the bivalent metal cations considered here, all of which (except Cd) fall into the borderline category and thus exhibit intermediate behavior that will be influenced by other factors such as the soil pH and redox potential. However, this principle may be useful in indicating which data are necessary to include in speciation models for a given soil; for example, Karlsson et al. (2007) found that including sulphur groups present in organic matter improved model fits of cadmium-organic matter binding data especially at high Cd loads. This represents a change from many speciation calculations with organic matter, which typically include only phenolic and carboxylic groups. The degree of sophistication of existing speciation models may somewhat eliminate the need to predict complexation using a broad principle such as that of HSAB; however, knowledge of this kind of chemical behavior is crucial for the quality control and interpretation of speciation model output (the models may give an answer that does not fit with common sense, the user must have enough knowledge of basic chemical behavior to be able to interpret this).

Conditions affecting the solubility and mobility of Cd, Cu, Pb, and Zn

Throughout this project special focus has been placed on the four elements Cd, Cu, Zn, and Pb. A brief summary will be given below of conditions known to increase or decrease the mobility of each of these elements. It is worth mentioning that a rich body of knowledge exists concerning each of these elements and their behavior in soil; what follows here is merely the briefest of summaries of information that may be useful in a broad way to site managers concerned with reducing risks associated with their occurrence. Interested readers are referred to the excellent works of Langmuir et al. 2003 and Ross 1994, both of which give impressive overviews, as well as the detailed chapters on each element found in Alloway 1990, Brookins 1988, Lindsay 1979, and Merian 1991. The information summarized below is taken from these sources, as well as from some other references (cited below).

CADMIUM

Factors that will increase Cd solubility in soils: high Cd content, high soil content of trace elements with a stronger affinity for the soil solid phase than Cd (e.g. Ca, Cu, Pb) (Bongers 2007), neutral to acidic pH (pH < 6) (Scokart and Meeus-Verdinne 1986), high salinity (Linde et al. 2007), oxidizing conditions, sandy soil Factors that will decrease Cd solubility in soils: extremely high concentrations of Cd leading to precipitation of Cd minerals (however, these conditions are unlikely to be reached in most soils, and the minerals may be subsequently resolubilized if soil conditions change), high pH (pH > 7.5 - 8), high carbonate contents (Buekers et al. 2007), moderate to strongly reducing conditions, low concentrations of trace elements with high affinity for soil solid phase

COPPER

Factors that will increase Cu solubility in soils: high concentrations of DOC, low or highly labile solid phase soil organic matter (SOM) content, soil pH < 5 (at very acidic pH values the organic-bound Cu may be released), or 6 < soil pH < 7.5 (in this pH range organic colloids are the most stable leading to increased vertical transport of the colloids and their bound metals), oxidizing conditions Factors that will decrease Cu solubility in soils: low concentrations of DOC, stable solid phase SOM content, 5 < soil pH < 6, or soil pH > 7, stable Fe- and Mn-oxides.

LEAD

Factors that will increase Pb solubility in soils: stable organic colloids (similar to pH conditions for Cu, see above), high concentrations of DOC, competition with Ca, high concentrations of Pb Factors that will decrease Pb solubility in soils: high pH leading to precipitation of Pb carbonate minerals, low DOC concentrations *Pb is the element whose behavior is the most difficult to predict, this conclusion has been found by many researchers investigating different elements at varying spatial and temporal scales (Bergkvist et al. 1989, Beyersman 1991, Linde et al. 2007, McBride et al. 1997, Sauve et al. 2000, Weng et al. 2001)

ZINC

Factors that will increase Zn solubility in soils: high salinity (Linde et al. 2007), acidic pH (pH < 5) (Scokart and Meeus-Verdinne 1986), oxidizing conditions, high soil content of trace elements with a stronger affinity for the soil solid phase than Cd (e.g. Ca, Cu, Pb) (Bongers 2007), sandy soil Factors that will decrease Zn solubility in soils: soil pH > 7 - 7.5, reducing conditions, high phosphate concentrations (may enhance Zn sorption) Investigations seeking to describe and/or quantify the behavior of multiple elements in the same system seem to inevitably find two groups (of the four elements whose factors are summarized above). Cadmium and zinc are more suceptible to increased solubility due to a pH drop, and organic complexation is less important (although non-negligible, especially in systems with high DOC concentrations); both can pose significant risks based on exponential release if pH drops below 4 - 4.5(5) (Bergkvist et al. 1989). The solubilities of copper and lead are more strongly related to the solid and colloidal phases of Fe/Mn-oxides and SOM, as well as to the soil concentration of DOC (Weng et al. 2002b). DOC concentrations are known to vary according to seasonal cycles, due for instance to higher levels of biological activity during warmer periods (Bonnett et al. 2006). Leaching of DOC (and associated metals) occurs when pH is higher and the organic colloids are more stable (Bonnett et al. 2006, Karathanasis et al. 2005). It follows that the management strategy most effective at reducing risks associated with high solubility and mobility will depend on which element is of most concern at the site in question, since no management strategy seems to be able to simultaneously result inopposite conditions necessary for controlling both groups of metals.

4. Multiple-scale modeling approach

The current state of the art in LCA and EcoRA has evolved over the past decade to include a finer spatial resolution in LCA (spatial scale of a watershed versus continent) (e.g. Humbert 2008) and to broaden EcoRA from a site-specific approach to a regional or national approach (e.g. De Vries 2008, Rutgers et al. 2008). The spatial scale of the watershed seems to be a coherent unit at which an integrative approach using both tools may be valuable. Furthermore, the watershed is a coherent hydrological, geological, pedological and ecological unit, compared to, for example, municipal or political boundaries. It may therefore be highly valuable to integrate existing knowledge and databases into a medium-scale modelling approach capable of interacting with a global assessment (such as LCA) as well as a site-specific assessment (such as EcoRA). Despite the aversion of many to modeling approaches based on criticisms of lack of realism, there are questions that cannot be answered using measurements alone due to limitations of resources. Furthermore, when the question is “Which management strategy to adopt in order to reduce risk associated with contamination?”, the different options are usually mutually exclusive and therefore cannot all be verified through measurement. Promising advances continue to be made regarding efforts to model large and small scale situations, such as the evolution of ecosystems over time, or changes in local biodiversity associated with modified environmental conditions (such as increased concentrations of atmospheric carbon dioxide). The number and complexity of parameters that must be incorporated to model terrestrial ecosystems (and especially the influences of terrestrial organisms on geochemistry) with any degree of realism may seem daunting, but the necessary knowledge and expertise do exist. These parameters have been elaborated in an earlier report, and will not be individually discussed here (see MuSA Deliverable D2.3/D3.2). Rather, two existing models that illustrate promising approaches will be discussed as an illustration of the existing state of the art, and of what is possible (given the appropriate time and resources).

Before going into detail regarding individual models, three general concepts will be briefly discussed that are (in this researcher's opinion) key points to incorporate into any multiple scale assessment model: a kinetic (as opposed to equilibrium) approach, a modular approach (incorporating different modules to make up an ecosystem model), and an approach that explicitly incorporates biotic influences on the ecosystem.

Kinetic, modular, organism based approach

Kinetic approaches are applied to many questions related to ecological and environmental risks posed by soil contamination. Metal uptake rates yield useful information for questions related to ecotoxicology (Gimbert et al. 2006, Gimbert et al. 2008, Van Straalen et al. 2005, Vijver 2005). Microbial activity rates yield useful information regarding biogeochemistry, migration rates of pollutant plumes, and degradation of organic compounds (Appelo and Postma 2005, Hunter et al. 1998). Mineral formation and dissolution rates (often linked to microbial activity) yield useful information regarding the stability of mineral compounds in soil (and porous media), colloid mediated contaminant mobility, and/or prospects for long-term geochemical engineering solutions to soil and/or groundwater contamination (Appelo and Postma 2005, Miotlinski 2008). A kinetic approach provides a more realistic way to model the complex interactions taking place in soils and porous media, especially where inorganic compounds are concerned (cases where equilibrium models do not apply) (Sparks 1995). Furthermore, knowledge and quality data concerning the kinetic aspects described above (among others) are increasingly available, which further justifies a kinetic approach.

In a modular approach to ecosystem modeling, different modules (e.g. hydrology, vegetation, invertebrate, different soil and litter layers, soil chemistry, land-use decision making) are related to each other, but are not required to incorporate the same parameters and/or degree of detail. There are two main advantages of such a modular approach: each module is able to incorporate as much (or as little) detail as is available based on reliable information, and interdisciplinary cooperation may be facilitated if teams of experts are able to work independently when necessary, but are also able to combine resources, knowledge, and results into a coherent model. The decision-support tool BIOCHEMORCHESTRA (Vink and Meeussen 2007) is an example of modular modeling approach (see below). As concerns for the effects of human activities on ecosystems grow, and as the legal need to incorporate sustainability into the decision making process become the status quo, the need to explicitly consider the effects of organisms on the ecosystems they inhabit (and in fact, create) becomes obvious. This is especially true regarding concerns about the effects of contamination, habitat destruction (or creation), and global climate change on large and small-scale biodiversity. Furthermore, the science of ecology is providing quantitative (although not always unequivocal) evidence as to how organisms influence their abiotic environment (through e.g. bioturbation (Wijnhoven 2007) or community interactions (Lavorel and Garnier 2002)).

BIORXNTRN

Based on the three aspects described above (kinetic, modular, organism-incorporating approach), two models will be briefly discussed that provide promising directions for future multiple scale modeling efforts. The first is a fully kinetic model, BIORXNTRN, that incorporates microbial metabolism (and the redox conditions resulting from it) into subsurface pollutant transport. This impressive model is described in Hunter et al. 1998, a reference that gives among other things, a very thorough discussion of modelling approaches to reactive transport in subsurface environments. The model described in this article deals with pollution of a pristine aquifer by organic leachate from a neighboring landfill, a situation which has little superficial resemblance to the question at hand (soil contaminated by metals). However, the approach incorporates many aspects that are key to the understanding of metal behavior in porous media, including the explicit incorporation of redox conditions (of prime importance for metals) and the influence of microbial activity. Hunter et al. demonstrate that when models fail to incorporate chemical reactions due to secondary redox products and the interactions between organic materials (whether pollutants, as in this case, or hypothetically speaking, soil organic matter) and microbial biomass, erroneous conclusions will be drawn regarding the fate of both organic and inorganic substances (1998).

BIOCHEM-ORCHESTRA

The second model, BIOCHEM-ORCHESTRA, is a decision-support tool designed to evaluate the effects of various river and floodplain management strategies on the uptake of potentially toxic inorganic substances by biota (flora and fauna) (Vink and Meeussen 2007). The model includes chemical, plant, and invertebrate modules (the latter separated into hard and soft-bodied organisms based on data from Vijver 2005). The chemical module explicitly includes parameters of importance for soil and porous media chemistry such as soil moisture content, water level, redox potential, and oxidation processes (organic matter degradation) (Vink and Meeussen 2007). The plant module is adjustable for different vegetation types based on parameters such as rooting depth and metal uptake rates. Overall, BIOCHEM-ORCHESTRA seems to be a very promising decision-support tool, although questions still exist regarding its applicability to non-floodplain soils. (Due to the proprietary nature of the model, it was not available for evaluation by this researcher). While neither of the models described above corresponds exactly to the question at hand, namely how to evaluate the effects of management strategies on terrestrial ecosystems characterized by high metal concentrations, both provide valuable insight and examples of how such a question may be approached, namely by integrating the effects of organisms on ecosystems in a kinetic, modular approach that is flexible enough to be scaled up to encompass a watershed, or scaled down to apply to a field-scale contaminated site (and its connected hydrological regime).

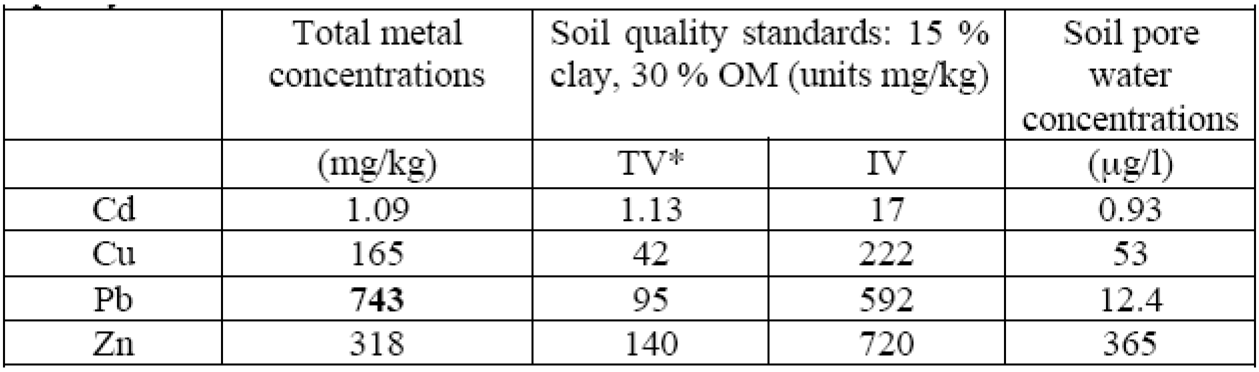
A certain number of ecological parameters have been identified in an earlier phase of this study as being key to the determination and prediction of the ecological impacts of metals (theoretically, any substance) in soil ecosystems. These are (in no particular order) season, microbial activity, soil chemical composition, vegetation, hydrology, soil age/history, land use, substance availability, mineralogy, toxicity, connectivity, soil organic matter content and composition, specific composition, and bioturbation. The study, quantification, and prediction of each of these factors is a scientific discipline in its own right. It is beyond the scope of this project to quantify how each of these factors contribute to the ecological impacts of metals in soils, in particular because the degree of quantitative importance of each of these factors will depend heavily on the physicochemical characteristics of the site as well as the current and projected land use. This is obviously a long (and probably exhaustive list), leading to criticisms that identifying all possible parameters does not help in simplifying the task of accounting for all of them simultaneously in a diagnostic or predictive model. However, if medium to long-term perspectives are to be used to evaluate the effectiveness of contaminated land management strategies, ignoring any of these factors will lead to erroneous conclusions. In some cases of contaminated sites it may not be necessary to take all of these factors into account; for example, if a contaminated site is to be used as a paved parking lot, and substance migration to groundwater can be excluded, it is unnecessary to evaluate the ecological effects of substances present in soils, because it is questionable whether or not a parking lot should even be considered as habitat for anything but automobiles (although it is part of the human-influenced terrestrial ecosystem). However, when the suitability of a contaminated site as habitat for humans, or as a nature reserve is the question at hand, the consideration of all of these factors gains importance (and predictive power). The development of a multi-compartment, kinetically based modular terrestrial ecosystem model is a worthwhile endeavor whose success will depend on interdisciplinary cooperation and the availability of quality data. In order to effectively model the exposure of soil-dwelling and terrestrial organisms to potentially toxic chemicals in soil, the possibility of including multiple uptake routes including cutaneous absorption, ingestion of soil and detritus, ingestion of plant tissues (above and belowground, as well as plant litter), and ingestion of other organisms (including microbes) should be available (although depending on the system in question, not all of these routes will necessarily apply). Furthermore, in order to provide the flexibility that would allow such a model to be useful in multiple assessment frameworks (such as LCIA and EcoRA), care should be taken to include various modalities of substance fluxes including atmospheric deposition, solid/liquified matter (i.e. sewage sludge, manure), leaching, erosion, and runoff. Lastly, in order to provide links to the ever-growing body of data concerning the relationships between population demographics and ecotoxicology (and therefore the most reliable predictions of the effects of substance emissions on large and small-scale biodiversity), care should be taken to include possibilities to model organism uptake at the individual level that is linked to the relevant spatial and temporal scales for predictions of population-level effects. As a final remark related to what a multiple-scale ecosystem model capable of evaluating the risks of metal-contaminated soils might look like, and how it might be implemented, it is worth mentioning that much of the information necessary for the construction of such a model already exists. For example, at the level of the European continent there are existing databases that provide information about soil properties such as pH, soil texture, land use, organic C content, and water level in spatially referenced format (see e.g. http://eusoils.jrc.it/ESDB_Archive/ESDB/Index.htm). [Unfortunately, these data are not provided in a terribly user-friendly format, such that a non-expert in GIS manipulation will have a hard time applying the information in a useful way. However, this situation would be easily resolved in an interdisciplinary approach that included a GIS expert as part of the team.] Such a spatially-referenced database provides an obvious link to existing spatially resolved impact assessment models, such as those currently available to users of LCIA, provided the spatial information used in an ecosystem model and an impact assessment model are compatible and available (which is not currently the case for Europe, but is theoretically possible). It is not difficult to imagine a spatially referenced database that included information on soil properties, trace element background concentrations, land uses, vegetation types, and even biogeographical species distributions.

6. de Ronde Venen: site-specific case study

Background information

The contaminated site used as a real-life application for the current study is located in the municipality of de Ronde Venen, in the province of Utrechet in the Netherlands. The site, known as Demmerik polder, exists currently as grassland used for extensive agriculture, in the form of pasture for low densities of cows and sheep. The history of the site involves centuries of human intervention. Drastic changes to the site began with cutting and removal of the original peat/peaty soil and conversion to agricultural fields surrounded by ditches (Stefan Kools, personal communication). Cutting the peat exposed the soil profile to oxidizing conditions resulting in rapid subsidence; attempts were made to slow or halt this process by covering the peat soil with layers of municipal waste, dune sand, and manure in a process known in Dutch as *toemakken*, resulting in a 15-50 cm layer of highly-organic soil referred to in Dutch as *toemakkdek* (Kools 2005). This practice was not unique to the site under consideration; indeed, the *toemakkdek* covers hundreds of hectares in the central part of the Netherlands, including the provinces of Noord-Holland, Zuid-Holland, and Utrecht (Kools 2005). At the Ronde Venen site, in addition to containing high levels of organic material, this anthropogenic soil layer also contains relatively high levels of metals, including levels of lead that exceed the environmental quality standards (EQS) developed based on site-specific parameters (content of organic matter, percent clay) according to Dutch national policy (Vegter 1995, Swartjes 1999, Crommentuijn et al. 2000). Several investigations have been carried out to determine the risks posed by these elevated levels of metals. These investigations have been conducted by national authorities and research institutes (Bosveld et al. 2000, Bosveld and De Poorte 1999). The site has also been investigated within the framework of a Dutch national research program designed to describe and quantify the ecological effects related to large-scale, diffuse pollution (SSEO program, see Posthuma and Vijver 2007 for more details). Three PhD studies involved in this framework included in-depth investigations into various aspects of the terrestrial and aquatic compartments of the site (Boivin 2005, Kools 2005, Van der Welle 2007). One study focused in particular on the ecotoxicological aspects of the soil ecosystem and its response to elevated metal concentrations; it was in the course of this investigation that the spatial heterogeneity of the contamination was revealed to be quite important (Kools 2005). Sampling of the soil using a grid of 0.5 to 5 meters revealed patches of elevated metal concentrations; it also revealed a positive spatial correlation between the different metals present (primarily Cd, Cu, Pb, and Zn) (Kools 2005). The table below shows the site-specific environmental quality standards based on the average site-specific soil contents of clay and organic matter (see Vegter 1995 for derivation of these site-specific values), as well as the average values of metal concentrations reported for the upper 10 centimeters of soil.

Table 1. Demmerik polder site-specific metal concentrations and environmental quality standards



	Total metal concentrations	Soil quality standards: 15 % clay, 30 % OM (units mg/kg)		Soil pore water concentrations
	(mg/kg)	TV*	IV	(µg/l)
Cd	1.09	1.13	17	0.93
Cu	165	42	222	53
Pb	**743**	95	592	12.4
Zn	318	140	720	365

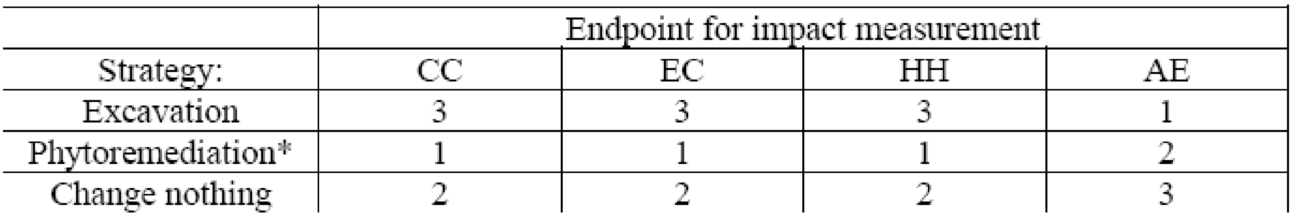
TV: Target value; values here are slightly different from those reported in F. Gambazzi's thesis, however this difference is slight and is assumed to be negligible, IV: Intervention value, Demmerik values from site measurements (Stefan Kools, personal communication), EQS derived according to Vegter 1995

It is worth mentioning that when soil properties influencing the site-specific EQS (% clay, % OM) vary significantly for the same site, the Target and Intervention values may also vary as well. In an effort to simplify, one (average) value of clay and OM content is taken for the entire site. As with all investigations into soil physico-chemical properties, considerable spatial heterogeneity is the rule rather than the exception. Comparison of many aspects of the soil ecosystem of the Ronde Venen site to that of several reference sites revealed no significant differences based on ecological parameters such as the nematode maturity index, plant species diversity, and earthworm diversity and/or biomass (Kools 2005); furthermore, no tolerance to pollution was detected in microbial communities from the site (lack of tolerance implies lack of undue stress) (Boivin 2005). However, laboratory experiments using terrestrial mesocosms consisting of soil cores taken from the site revealed an increased vulnerability to stress (heat stress, additional metal stress) for the patches of soil with high levels of metals (Kools 2005). The main conclusions of all investigations into this site are that although lead concentrations exceed intervention values in many spots, this contamination does not currently pose significant risks to the human or non-human populations inhabiting this site. However, there are four main lines of evidence to suggest that risks may increase over time: a) metal concentrations show some evidence of vertical (downward) migration (see below), b) concentrations of lead in liver and kidneys from animals grazing on the site are above legal threshold limits (Bosveld and De Poorte 1999), c) atmospheric deposition of lead may represent a non-negligible input pathway leading to increasing concentrations of lead over time (Van Drecht et al. 1996) (see below), d) metals are carried to the surrounding aquatic compartment (water and sediments of ditches) representing a possible ecological risk to the aquatic communities. Furthermore, the existence of lead concentrations in excess of legal standards limits the options of site managers regarding land use. For example, some discussion has taken place focusing on developing this area as a natural park. However, if it were to be developed as such and open to the public, there are non-negligible risks that children visiting the park may be exposed to dangerous levels of lead, which are known to cause neurological and developmental disorders (Van Wezel et al. 2008). No clear answers present themselves as to how to effectively manage the site in order to reduce the risks associated with the elevated metal concentrations.

LCA analysis of management strategies: master's project of Francesca Gambazzi

This project has sought to evaulate different management strategies using two tools, namely EcoRA and LCA, in a comparative assessment. Three main strategies have been retained for consideration: change nothing in the current management strategy (maintain the site as pasture for low densities of cows and sheep), excavate the contaminated soil, employ phytoremediation (use plant species to "pump" the metals from the soil followed by removal of the plant biomass (two variants: reeds and willows), and a modified version of the phytoremediation strategy involving removal of existing plant biomass (pasture grass) from the site. The first three of these scenarios have been evaluated from an LCA point of view, these results are given in the Master's thesis of Francesca Gambazzi and will only be briefly discussed here. The LCA of different management strategies was based on quantification of the impacts on the endpoints of climate change, energy consumption, human health, and aquatic ecotoxicity. Impacts on climate change were calculated based on the greenhouse gas emissions related to, for example, heavy diggers to excavate and lorries to transport the excavated material. Impacts on energy consumption were related to similar factors, except in the case of phytoremediation, where energy is created rather than consumed due to energy co-generation that is possible with the removed plant material. Impacts on human health were calculated based on the carcinogenic and non-carcinogenic health impacts of metals emitted into the aquatic environment due to the various activities associated with management activities. Impacts on aquatic ecotoxicity were calculated based on modeled emissions of heavy metals from the contaminated soil to the neighboring ditches. It is interesting to note that in this analytical framework the contaminated soil is considered a source of contamination for the sensitive targets that are the human and aquatic populations; however, because LCA considers only fluxes of substances, the metals that remain in the soil do not enter into the calculations. A simplified analysis of the comparison of the different management strategies are that each strategy has its benefits and drawbacks based on different scores for each endpoint category. The results are summarized in the following table, however readers are strongly encouraged to see the original report by F. Gambazzi for a more detailed explanation of how these results were obtained. Furthermore, the relative nature of the ranking needs to be emphasized. The impacts are not absolute, therefore the interpretation of the impacts of each management strategy are only meaningful when compared to the other strategies.

Table 2. Summary of LCA impact rankings of management strategies Endpoint for impact measurement



	Endpoint for impact measurement			
Strategy:	CC	EC	HH	AE
Excavation	3	3	3	1
Phytoremediation*	1	1	1	2
Change nothing	2	2	2	3

* reeds vs. willows not specified because choice of plant species does not affect ranking

CC= climate change, EC= energy consumption, HH= human health, AE= aquatic ecotoxicity

ranking 1: lowest impact level (best score), 2: middle impact level (intermediate score), 3: highest impact level (worst score)

The above results do not reveal one strategy as being unequivocally the best option; rather, they show that the "best" management strategy depends on the priorities of the site managers. However, one result does appear obvious, which is that the management strategy involving taking no action never receives the highest score, while both excavation and phytoremediation receive the highest score (lowest impact) for at least one impact category. LCA methodology does not accurately characterize the impacts associated with metals in soils; therefore, these impacts were not evaluated in Gambazzi's analysis. Aspects associated with the risk assessment of metals in the soil of this contaminated site will be discussed below, as will aspects related to the effects of the different management strategies on the soil ecosystem itself, which is not quantified in LCA. It is worth repeating at this stage that some modifications due to adoption of various management strategies are impossible to evaluate in LCA. These include modifications in habitat quality such as the total destruction associated with excavation or drastic changes in vegetation type (grassland to reeds or willow coppices) (Udo de Haes 2008). LCA is not the only framework incapable of evaluating this kind of impact; there is currently no quantitative framework available in EcoRA to evaluate the impacts of these changes on the habitat quality of the soil ecosystem, the probable effects on biodiversity, or the combined impacts of multiple stressors such as eutrophication, contamination, and invasion by non-native species. This is an area where both assessment frameworks would benefit from expansion.

Current status of metals in the soil: a possible internal cycle

Following a sampling campaign carried out in March 2001, metal concentrations at various soil depths were determined at 15 points in one of the grassland fields of the Demmerik polder. These profiles reveal two main characteristics of the metal contamination of this soil: a) the lead concentrations in the upper soil layers (10 cm) exceed legal values in many of the sampling spots investigated by less than one order of magnitude, and b) lead concentrations in lower soil layers (30-40 centimeters) are two to ten times higher than legal values. Of the other metals included in this report (Cd, Cu, and Zn), only copper exceeds the intervention values established for the site. The soil profiles also reveal typically higher copper concentrations in the lower soil layers (30 cm). Profiles of Cd and Zn reveal the highest concentrations to be in general, restricted to the upper soil layers (10 cm). In summary, Cu and Pb concentrations tend to increase with depth, whereas Cd and Zn concentrations tend to decrease with depth (although in some cases, Zn concentrations also increase with depth). Two explanations immediately present themselves: levels of Pb and Cu were higher in layers of *toemakkdek* spread earlier in history (lower layers = older layers), or vertical migration of Pb and Cu has enriched the lower soil layers, whereas Cd and Zn have remained more or less sequestered in the upper soil layers. No information has been found to verify either explanation. For example, spreading of material high in Cu and Pb and low in Cd and Zn content could have taken place sequentially in thin layers over the entire field's surface, followed by spreading of material relatively higher in Cd and Zn and lower in Cu and Pb in thin layers. In this researcher's opinion, this series of events is unlikely. What seems more likely is that organically (or other colloidal phases, such as iron oxide) bound forms of Cu and Pb have been transported into lower soil layers over time. The soil of the site is characterized by exceedingly high organic matter contents, as is typically the case in peatlands (e.g. Mathur and Farnham 1985). The solubility of Cu and Pb has been demonstrated to depend heavily on the soil organic matter content in many studies (McBride et al. 1997, Weng et al. 2001, Weng et al. 2002a, Weng et al. 2002b). Leaching studies have also shown that of all treatments investigated, high concentrations of DOC have been most effective at removing Cu and Pb from soil columns via vertical migration; wheras Cd and Zn are more susceptible to leaching by lowering soil pH (Linde et al. 2007). Measurements of soil pH and DOC concentrations at various depths have not been reported for this site, nor were pH or DOC concentrations measurements taken of the same samples used for the total metal analyses reported in the profiles described here. This lack of data means that any explanations are hypothetical.

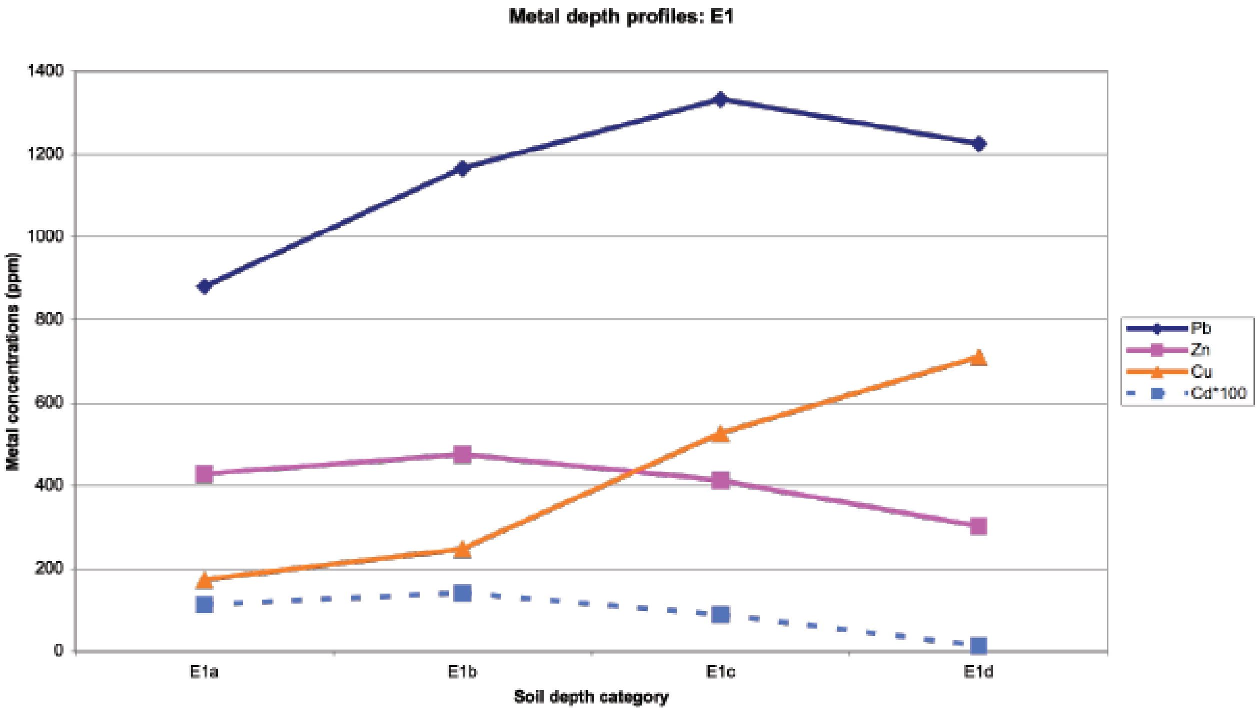
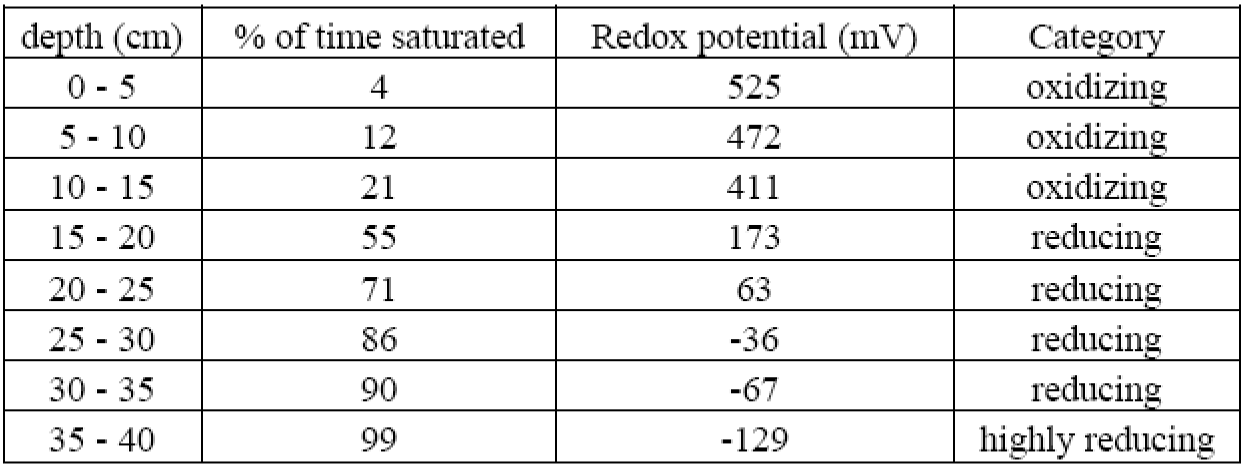


Figure 1. Example of a soil profile taken in the Demmerik polder. Soil depth categories: a = 10 cm, b = 20 cm, c = 30 cm, d = 40 cm (note for Cd, concentration = Cd *100)

It is well known that redox potential can have a strong effect on metal speciation. There is also a link between redox potential and pH: because many redox reactions consume protons, reducing conditions tend to promote higher pH levels, and oxidizing conditions tend to promote lower pH levels (Bartlett and James 1993). Measurements of soil redox potential are notoriously difficult to obtain, and are often unreliable (Mansfeldt 2003, Niedermeyer and Robinson 2007). Despite these difficulties, promising results have been obtained with some permanent redox measurement installations in the field. Specifically, good results have been obtained when soil water level and saturation status are monitored. The study of Mansfeldt (2003) provides a highly significant correlation between the percentage of time a certain soil layer is saturated and the redox potential expressed in mV: redox potential (mV) = -6.902 x (% of the year layer is saturated) + 555.4 (R = 0.971). This relationship provides site managers with a useful tool for predicting redox potential at a given soil layer if information is known regarding the soil water level. Although soil water levels for the fields in question have not been closely monitored (or if they have, these data have not been made available), soil water levels measured in a field with similar dimensions, similar characteristics, and similar regulation of the water levels in the surrounding ditches have been generously provided (Marlies van der Welle, personal communication). Based on these water level data and the equation of Mansfeldt (2003) the following calculations were carried out for soil layers in increments of 5 centimeters (see Table 3). Please note that the reported values of redox potential should only be taken as indications; indeed, most researchers use a classification system based on ranges of values, these are given in the fourth column of Table 3.

Table 3. Redox potential at various soil depths based on Mansfeldt 2003



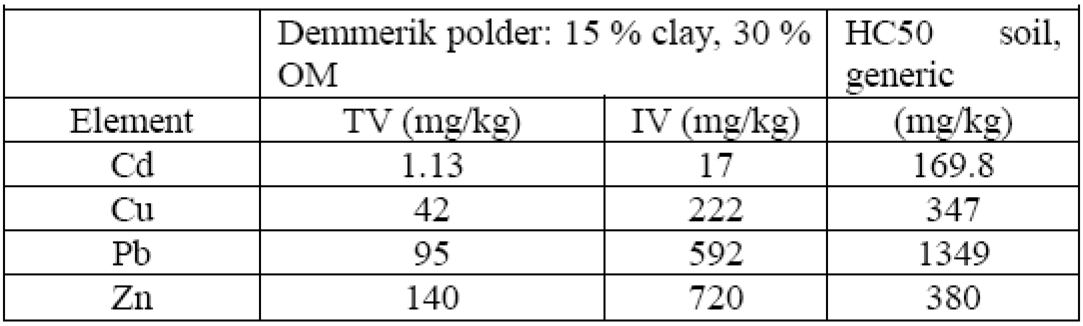
depth (cm)	% of time saturated	Redox potential (mV)	Category
0 - 5	4	525	oxidizing
5 - 10	12	472	oxidizing
10 - 15	21	411	oxidizing
15 - 20	55	173	reducing
20 - 25	71	63	reducing
25 - 30	86	-36	reducing
30 - 35	90	-67	reducing
35 - 40	99	-129	highly reducing

Based on these calculations, it is expected that reducing conditions dominate below 15-20 centimeters depth (although seasonal and/or annual fluctuations, and heterogeneity due to field-scale macroporosity no doubt occur). This is in keeping with observations in the field, which reveal that soil layers below 30-40 centimeters are saturated more or less permanently (Stefan Kools, personal communication), as well as observations of other researchers in peatlands where anaerobic conditions are expected below depths of 10 cm (Niedermeyer and Robinson 2007). A possible (albeit speculative) mechanism by which vertical migration of Cu and Pb takes place can thus be elaborated. Atoms of Cu and Pb are preferentially bound by soil organic matter in solid, colloidal, and dissolved forms. The oxidizing conditions (low pH) and high microbial activity in the upper soil layers result in the maintenance of high concentrations of DOC capable of binding and translocating Cu and Pb atoms. During the warmer months of the year, higher temperatures combine with decreased precipitation (relative to the colder months) result in a lowering of the soil water level, and progressive downwards vertical transport of the dissolved and colloidal organic matter (and bound metal atoms). These are then immobilized by two factors: they encounter a more or less static water layer at 30-40 centimeters depth (physical barrier) that is characterized by reducing conditions (higher pH) (elecrochemical barrier). This promotes a) coagulation/sedimentation of DOC/organic colloids and b) precipitation of sulphide mineral phases that are stable as long as reducing conditions dominate. There is thus some evidence to support the hypothesis that metal ions may be sequestered in mineral phases in the deeper soil levels (although unverified by direct microscopic mineralogical observation). However, there is also evidence to support the hypothesis that these mineral phases continue to be involved in the biogeochemical cycles of the grassland ecosystem. Visits to the site reveal abundant evidence of small mammal burrows (i.e. moles); furthermore, field surveys at the site indicate significant annelid communities in the form of earthworms and enchytraeids (Kools 2005). While it is not known to what degree the activities of enchytraieds contribute quantitatively to bioturbation, both earthworms and small mammals have been shown to contribute significantly to the translocation of soil from deeper soil layers to the soil surface in a Dutch floodplain soil where this question was investigated (Wijnhoven 2007). At the floodplain site where the investigations took place, earthworms were shown to be responsible for almost two times as much soil relocation as sedimentation caused by seasonal flooding events (Wijnhoven et al. 2006). It is therefore this researcher's opinion that rather than representing stable mineral phases, the high soil concentrations found in deeper soil layers of the Demmerik polder are involved in cyclical movements between the upper and lower layers mediated by the combined effects of the hydrological regime, colloidal/DOC-mediated transport, and the actions of soil-dwelling organisms. If this is true, there are implications for organisms inhabiting the soil of the site, grazers occupying the site, and long-term site management. For the organisms inhabiting the soil of the site, exposure to high (potentially toxic) levels of metals cannot be ruled out; this may be true for both organisms whose primary route of exposure is cutaneous transport via dissolved concentrations in soil pore water, and organisms for which ingestion of soil particles is an important route of exposure. Furthermore, accumulation by organisms low in the food chain may represent a non-negligible point of entry for metals into the terrestrial food chain. For the grazers occupying the site (cows and sheep), the high levels of metals maintained in the upper soil layers due to this (hypothetical) internal cycle may result in continuous exposure to metals, which may eventually result in their meat and/or milk being unfit for human consumption. The implications for site management are that although the ecosystem seems to be functioning normally based on ecological observations, the metals present in all soil layers may represent a constant source of toxic pressure. This pressure may result in an increased energy demand for detoxification, decreased energy availability for growth/reproduction, and an increased vulnerability to additional stress factors (demonstrated in incubation experiments using TME's from the site (Kools 2005)). [There is some evidence to support the contention that toxic stress is depressing reproduction at this site: some researchers have noted a tendency of organisms capable of vegetative/asexual reproduction to dominate in contaminated soil systems (Dechamps et al. 2007, Krivolutsky 1996, Schultz and Hutchinson 1991). The high densities of enchytraieds (often capable of asexual reproduction (Lavelle and Spain 2003)) may indicate moderate reproductive stress. This hypothesis is however unverified.] The concrete implication for site management is that at the absolute minimum of the intervention spectrum, monitoring based on a set of biological and physico-chemical parameters is strongly recommended (see below, section Recommendations for site management).

A special case: the element Zinc

Although the above discussion focused on the elements Cu and Pb, which appear to display vertical mobility in the soil column, the presence of elevated concentrations of Zn also warrant attention. This may seem unnecessary, because soil concentrations of Zn are below the site-specific intervention values. However, these values have not been determined on the basis of ecotoxicological data (Rutgers 2008). When intervention values for Zn are compared to risk values derived for soil ecosystems (HC50) the obvious conclusion is that while intervention values for Cd, Cu, and Pb are lower than ecosystem risk values (and are therefore assumed to be adequately protective), intervention values for Zn are approximately the double of risk values (see Table 4).

Table 4. Site-specific soil metal EQS, and generic soil HC50 for metals



	Demmerik polder: 15 % clay, 30 % OM		HC50 soil, generic
Element	TV (mg/kg)	IV (mg/kg)	(mg/kg)
Cd	1.13	17	169.8
Cu	42	222	347
Pb	95	592	1349
Zn	140	720	380

This observation, combined with documented cases of Zn being the element most likely (among a mix of metals) to result in observed ecological damage (Hopkin and Hames 1994), and evidence to support toxic effects of Zn appearing well below legal intervention values (Van Beelen et al. 2004) suggest to this researcher that the ecological risks of Zn in soil are far from being well understood, and that their management is far from being well mastered. The complex socio-economic reality contributing to the setting of environmental quality standards above ecological risk values is understandable as being part of the compromise that is inherent to sustainable development. However, the persistence of metals in all environmental compartments, the documented toxic risks posed by Zn, and the need to consider the long term impacts of environmental policy on a) biodiversity and b) the provision of necessary ecosystem services result in the recommendation that environmental monitoring agencies not be complacent concerning the toxic risks posed by elevated concentrations of Zn in soils.

Metal balance model of Demmerik polder site

While it would have been very interesting to employ some of the speciation and transport models mentioned in Section 3 of this report, lack of data made this impossible. The soil of the site is a unique combination of a rich peaty soil and a completely anthropogenic layer (not corresponding to any well-described soil type). The soil solid phase has not been investigated or described to any great degree, nor have mineralogical data been gathered that would permit reasonably reliable assumptions to be made. Initial (and highly uncertain) speciation calculations indicate that soil the physico-chemical situation is at the border of stability for several mineral phases (among them iron and manganese oxides) that may be very important in controlling metal solubility as well as colloidmediated metal mobility. Furthermore, the soil pore water has not been subject to any rigorous or complete physico-chemical description that would have permitted reliable speciation calculations. It would have been interesting to characterize ecological risks associated with metal mobility throughout the terrestrial trophic chain; however, no systematic measurements were taken that would permit this sort of analysis (for an example, see Hunter et al. 1987a, Hunter et al. 1987b, Hunter et al. 1987c). It would also have been interesting to link soil concentrations to uptake by individual soil-dwelling organisms, and then to link concentrations in individuals to effects at the population level (i.e. effects on the population growth rate following e.g. Van Straalen et al. 1989). This was impossible due to lack of sufficient time and data. Due to the limited nature of available data, conclusions regarding the effects of various management strategies are very general and highly speculative. However, one outcome of the use of this site as a case study has been the identification of which data would be necessary for a more informative analysis. This outcome will be discussed in more detail at the end of this section. A mass-balance approach has been used to evaluate the impacts of different management strategies on fluxes of metals leaving the site (plant offtake, leaching), as well as on the evolution of soil metal concentrations over medium to long time periods (t = 200 years). The model described below is a modified version of that described in Harmsen 1992, and bears some similarity to models advocated in Moolenar and Lexmond 1999, Moolenar 1999, and demonstrated in Moolenar et al. 1998. The model as described in Harmsen 1992 will be briefly summarized in order to acquaint readers with the general idea. This will be followed by a more in-depth discussion of the site-specific model used to evaluate management strategies for the metal-contaminated Demmerik polder. In his chapter dedicated to the long term behavior of metals in agricultural soils, Harmsen elegantly describes how the solubility relationship of metals in soils can be used to predict the evolution of a) metal fluxes leaving the soil and b) the evolution of soil metal concentrations over time (1992). The main fluxes considered in this simple model are input (atmospheric deposition, fertilizer application) fluxes, and output fluxes (leaching and plant uptake in this case, although no theoretical barrier limits including additional output fluxes such as erosion). The relationship between metals in the soil solid phase and dissolved metal concentrations is identified as belonging to a zero, first, or second order case. Specifically, in soils where precipitation/dissolution reactions control metal solubility, zeroth order rate laws apply to the dissolved metal concentration, which is assumed to remain constant over time. In soil where linear adsorption reactions control metal solubility, first order rate laws apply and dissolved metal concentrations increase in a proportional fashion to total metal concentrations. In soils where the increase in dissolved concentrations is more than proportional to total metal concentrations, second order rate laws apply; an example of such a case is a soil with extremely high (although not high enough to result in precipitation of metal minerals) metal concentrations such that the exchange/adsorption sites on the soil solid phase are close to saturation. This analytical model is very attractive due to its simplicity (this is a compliment to the author, rather than a criticism, simplicity being a desirable trait) as well as to its applicability to cases of soils with multiple metals, each of which may demonstrate a different mechanism of solubility control. With a minimum of input parameters, and a limited number of initial assumptions, evaluations of different management strategies on soil metal concentrations can be effectuated. This has been done for the case of the Demmerik polder site, as is described below. The assumption was made that linear adsorption describes the solubility relationship of the metals in this soil based on the following lines of reasoning:

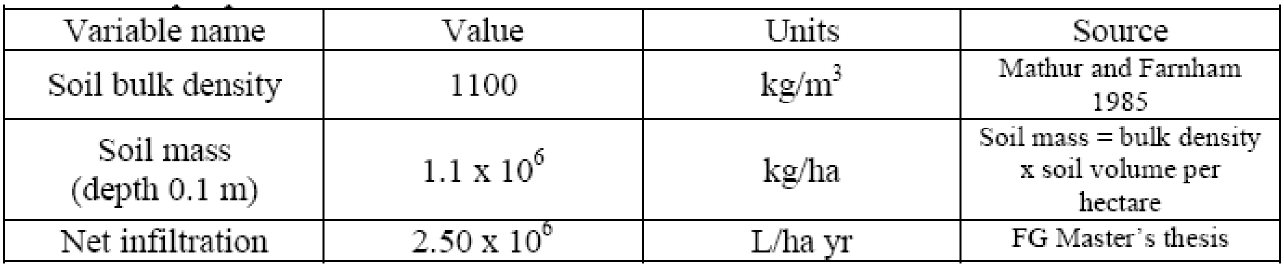
1) peat soils (and organic-rich soils in general) are known to have high cation exchange capacities (Mathur and Farnham 1985), which implies that it would take very high metal loads to saturate the exchange complex

2) the soil has a relatively high clay content, which also contributes to a high cation exchange capacity

3) no site-specific investigations have been carried out that would allow a more detailed appreciation of the solubility relationship of the metals in this soil (in the study described in Moolenar et al. 1998, information was available concerning an adsorption isotherm for Cu. This information was then incorporated into a site-specific mass balance model able to incorporate simplified speciation information. This kind of information regarding the specific solubility control for a given site will drastically improve long-term predictions of metal

mobility/evolution of soil metal concentrations over time). The following input parameters were used to develop a site-specific predictive model (see Tables 5 and 6).

Table 5. Input parameters common to all metals and all scenarios

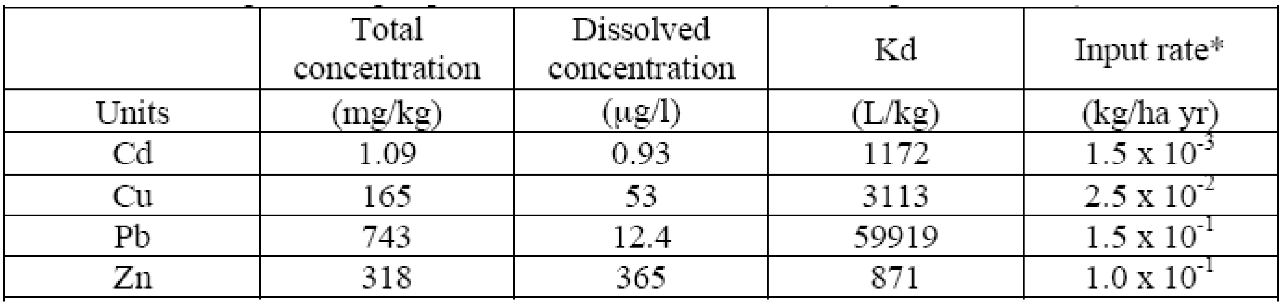


Variable name	Value	Units	Source
Soil bulk density	1100	kg/m^3	Mathur and Farnham 1985
Soil mass (depth 0.1 m)	1.1×10^6	kg/ha	Soil mass = bulk density x soil volume per hectare
Net infiltration	2.50×10^6	L/ha yr	FG Master's thesis

Soil mass = bulk density x soil volume per hectare

Net infiltration 2.50 x 106 L/ha yr FG Master's thesis

Table 6. Metal specific input parameters for all scenarios (except excavation)

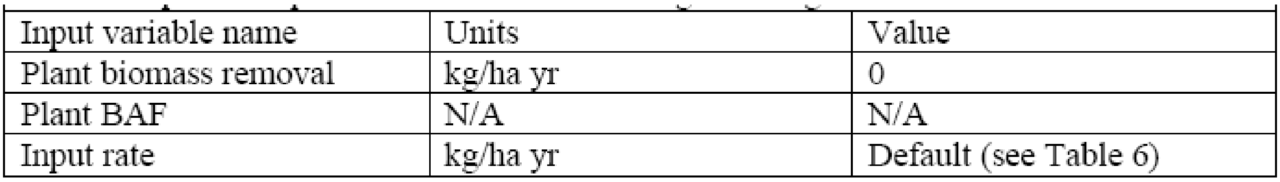


	Total concentration	Dissolved concentration	Kd	Input rate*
Units	(mg/kg)	(μg/l)	(L/kg)	(kg/ha yr)
Cd	1.09	0.93	1172	1.5×10^{-3}
Cu	165	53	3113	2.5×10^{-2}
Pb	743	12.4	59919	1.5×10^{-1}
Zn	318	365	871	1.0×10^{-1}

*includes atmospheric deposition only, values taken as mid-range estimate from Van Drecht et al. 1996

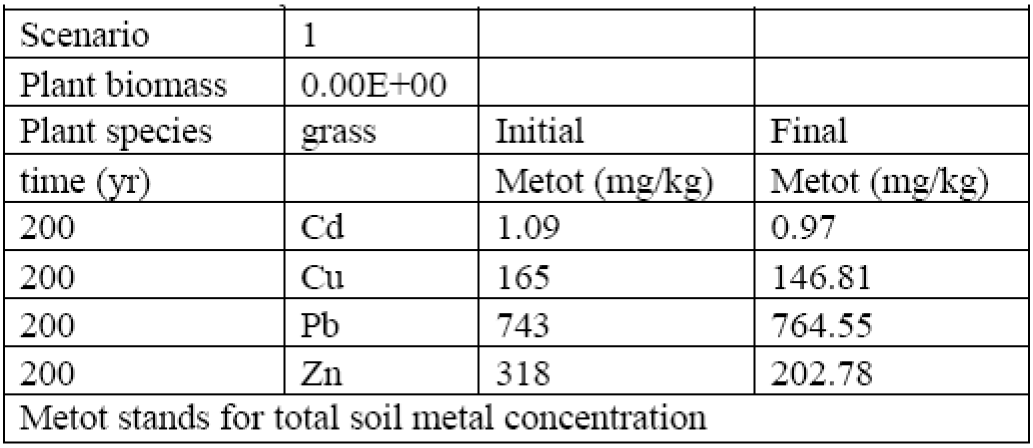
It is not specified whether the flux due to leaching will end up in the water table under the site, or the ditches surrounding the site. Due to the high degree of connection between these masses of water, this question seems academic. The flux due to leaching is evaluated at each time step based on the predicted pore-water concentration and the net infiltration (assumed to be constant for simplification, but easily modifiable). Based on the assumption of first-order kinetics for all four metals investigated (Cd, Cu, Pb, and Zn), the site-specific constant of proportionality is the simple ratio between the total (mg/kg) and dissolved (mg/L) concentrations (Kd value). While questionable on many scientific levels, including the fact that the partitioning coeffecient is expected to vary according to many factors related to metal properties, soil properties, and sampling and handling techniques, lack of better data made this the only possible approach. The mass of each metal removed by leaching at each time step is described by the equation: Leaching flux (kg metal / ha yr) = ((total metal (mg/kg) / Kd (L/kg))/ infiltration rate (L/ha yr))/ 1x 106 (conversion factor, mg to kg). The flux due to plant biomass offtake is evaluated at each time step based on a constant of proportionality derived according to the plant bioaccumulation factor (BAF) and the plant biomass removed from the site. While this method of calculation does not reflect the dissolved metal concentration directly, it assumes that plant uptake is proportional to total metal concentration based on a dimensionless constant, the BAF. This BAF is specific to the site, the metal, and the plant species and its value is based on measurements taken at the site (implicit is the assumption that uptake from the soil is the only factor contributing to the total plant concentration, which is obviously a simplification of reality). It is the ratio of the average aboveground plant metal concentration to the total soil metal concentration. The mass of each metal removed by plant offtake at each time step is described by the equation: Plant flux (kg metal / ha yr) = ((BAF x total metal (mg/kg))/ 1 x 106 (conversion factor, mg to kg)) x plant biomass removed (kg/ha yr) Management strategies were evaluated according to the following assumptions: management strategies do not change the solubility relationship of metals in the soil (linear kinetics apply to all scenarios), physico-chemical conditions remain constant (questionable realism, but necessary simplification due to limitations of time, resources, and quality input data), and the only parameters varying between different scenarios are a) plant biomass removal, and b) plant bioaccumulation factor that varies between species. The structure of the model is a simple mass balance evaluated using a yearly time step for a period of 200 years. Although this time period may seem absurdly long, it has been retained for two reasons: due to the linear nature of the offtake fluxes, it is a simple matter to estimate concentrations at any point between the initial period and the final period, and the rates of metal offtake are so slow for all strategies except excavation that reductions in metal concentrations are almost negligible over shorter (t = 10 or 20 years) periods. The total metal concentration is taken as the average total metal concentration in the upper 10 centimeters (even though it is known that metal concentrations in lower layers exceed those in upper layers by ten times, the concentrations in the upper 10 cm were used for the following reasons: 1) coherence with the analysis of F. Gambazzi, 2) dissolved metal concentrations for the lower soil layers are not available). By using only a single average value the observed spatial variation of metal concentrations is not accounted for; however, it should be remembered that this model is intended for illustrative purposes only. If a realistic evaluation were desired (and the appropriate time and data made available) this kind of mass balance could be evaluated for different hotspots, with varying (rather than one average) metal concentrations. The change in metal concentration at each time step is the simple difference between inputs and outputs (can be positive or negative). The influence of monitoring on scenario evaluations deserves a brief explanation: because monitoring is indicated regardless of which management strategy is adopted, and due to the fact that monitoring activities are not expected to alter site metal concentrations and/or behavior, monitoring is not explicitly included in the model. Scenario 1: change nothing The site continues to be used for grazing by cows and sheep. No biomass removal takes place because it is assumed that all grass that is removed by herbivores is returned to the soil in the form of fecal matter. Data allowing this assumption to be verified or rejected have not been available (no data concerning the rate at which grazers consume grass, grazer density, what proportion of grazer dietary needs are met by the grass of the site, what proportion of time grazers spend at the site, the rate at which grass is removed from the site by other means such as mowing, etc.).

Table 7. Specific input data for Scenario “Change nothing”



Input variable name	Units	Value
Plant biomass removal	kg/ha yr	0
Plant BAF	N/A	N/A
Input rate	kg/ha yr	Default (see Table 6)

Table 8. Summary of scenario results



Scenario	1		
Plant biomass	0.00E+00		
Plant species	grass	Initial	Final
time (yr)		Metot (mg/kg)	Metot (mg/kg)
200	Cd	1.09	0.97
200	Cu	165	146.81
200	Pb	743	764.55
200	Zn	318	202.78
Metot stands for total soil metal concentration			

Table 9. Specific input data for Scenario “Excavation”

Input variable name	Units	Value
Cd total concentration	mg/kg	1.13
Cu total concentration	mg/kg	42
Pb total concentration	mg/kg	95
Zn total concentration	mg/kg	140
Plant biomass removal	kg/ha yr	0
Plant BAF	N/A	N/A
Input rate	kg/ha yr	Default (see Table 6)

Table 10. Summary of scenario results

Scenario	2		
Plant biomass	0.00E+00		
Plant species	grass	Initial	Final
time (yr)		Metot (mg/kg)	Metot (mg/kg)
200	Cd	1.13	0.97
200	Cu	42	40.52
200	Pb	95	121.45
200	Zn	140	97.21
Metot stands for total soil metal concentration			

Table 11. Specific input data for Scenario “Phytoremediation using reeds”

Input variable name	Units	Value
Plant biomass removal	kg/ha yr	1.2 x 10^4
Plant BAF Cd		0.29
Plant BAF Cu		1.9 x 10^{-3}
Plant BAF Pb		9.5 x 10^{-3}
Plant BAF Zn		0.046
Input rate	kg/ha yr	Default (see Table 6)

Table 12. Summary of scenario results

Scenario	3		
Plant biomass	1.20E+04		
Plant species	reeds	Initial	Final
time (yr)		Metot (mg/kg)	Metot (mg/kg)
200	Cd	1.09	0.56
200	Cu	165	146.21
200	Pb	743	749.15
200	Zn	318	184.10
Metot stands for total soil metal concentration			

Table 13. Specific input data for Scenario “Phytoremediation using willows”

Input variable name	Units	Value
Plant biomass removal	kg/ha yr	5700
Plant BAF Cd		2.727
Plant BAF Cu		0.289
Plant BAF Pb		0.019
Plant BAF Zn		2.182
Input rate	kg/ha yr	Default (see Table 6)

Table 14. Summary of scenario results

Scenario	4		
Plant biomass	5.70E+03		
Plant species	willow	Initial	Final
time (yr)		Metot (mg/kg)	Metot (mg/kg)
200	Cd	1.09	0.12
200	Cu	165	109.30
200	Pb	743	749.91
200	Zn	318	25.42
Metot stands for total soil metal concentration			

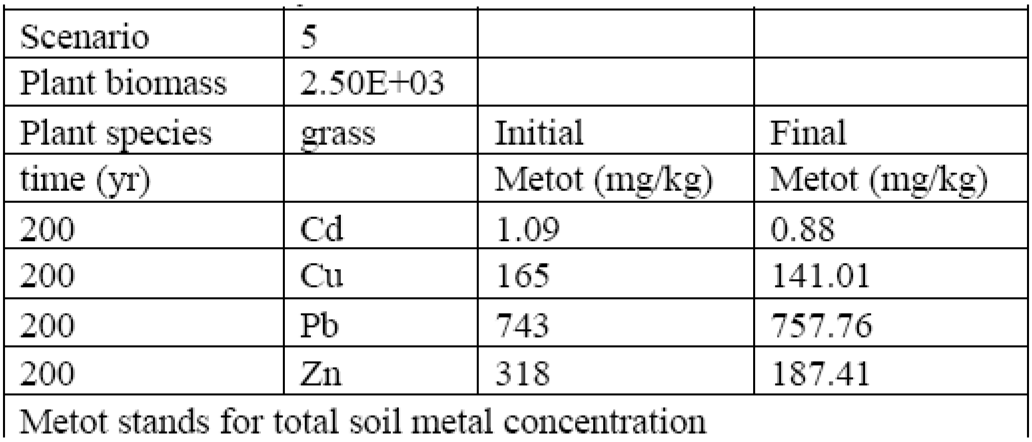
Table 15. Specific input data for Scenario “Existing biomass removal”

Input variable name	Units	Value
Plant biomass removal	kg/ha yr	2.5×10^3
Plant BAF Cd		0.23
Plant BAF Cu		0.09
Plant BAF Pb		0.02
Plant BAF Zn		0.18
Input rate	kg/ha yr	Default (see Table 6)

Interpretation of results

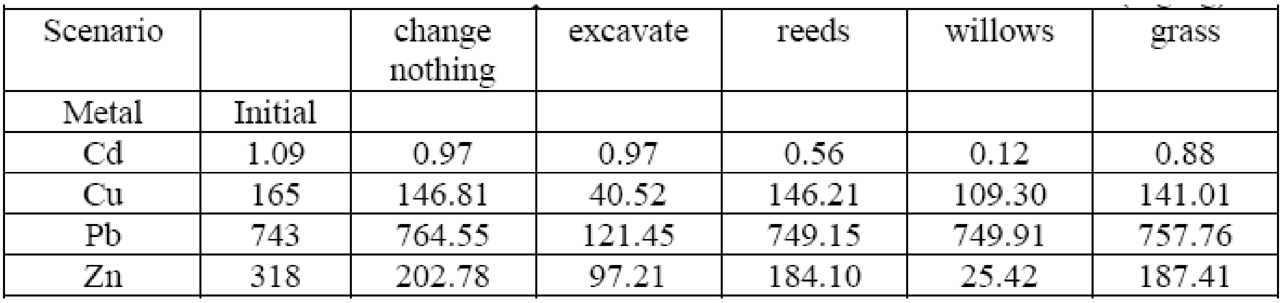
An initial survey of the results of scenario evaluations reveals that due to atmospheric deposition of metals, Pb soil concentrations actually increase under all management strategies except for excavation. It is important to note that the values for atmospheric deposition are taken from a report dating from 1996; if rates of metal deposition have significantly decreased over time, this could alter the results. However, it seems inappropriate to assume deposition rates of zero (or some other arbitrary value), since in reality atmospheric deposition of metals is known to occur. The smallest increase in Pb concentrations is observed for phytoremediation using reeds, although the difference compared to phytoremediation using willows is minimal (and due to the high level of uncertainty associated with the model, probably negligible). The greatest increase is seen with the “change nothing” scenario (although the difference is also quite small). At first glance it may seem strange that the differences between final Pb concentrations for the “change nothing” and all three phytoremediation strategies are so small, because the “change nothing” scenario involves no offtake by plants and therefore more Pb accumulation should take place. However, readers are reminded that the metals removed via leaching are proportional to the total concentration: if the total soil metal concentration is higher (due to less removal by plants), the leaching flux will also be higher.

Table 16. Summary of scenarios results



Scenario	5		
Plant biomass	2.50E+03		
Plant species	grass	Initial	Final
time (yr)		Metot (mg/kg)	Metot (mg/kg)
200	Cd	1.09	0.88
200	Cu	165	141.01
200	Pb	743	757.76
200	Zn	318	187.41
Metot stands for total soil metal concentration			

Table 17. Results of scenarios 1-5 expressed as final total metal concentrations (mg/kg)



Scenario		change nothing	excavate	reeds	willows	grass
Metal	Initial					
Cd	1.09	0.97	0.97	0.56	0.12	0.88
Cu	165	146.81	40.52	146.21	109.30	141.01
Pb	743	764.55	121.45	749.15	749.91	757.76
Zn	318	202.78	97.21	184.10	25.42	187.41

Recommendations for site management

When the results of long-term ecological predictions are combined with an LCA anlysis of management options, does one strategy come out clearly as being the most desirable for this site? The first answer is no, because both tools (EcoRA and LCA) can provide comparative frameworks, but site managers and other stakeholders must still define the priorities by which possible strategies will be judged. If the priorities are to manage the site such that risks are reduced *as much as possible* without a) destroying the site ecosystem, b) generating huge impacts on any of the endpoints considered, or c) costing exorbitant amounts of money both tools indicate that some form of phytoremediation may be the best strategy. Although the differences in final Pb soil concentrations are small between the “change nothing” and phytoremediation strategies, reductions in Cd and Zn concentrations are greater. A lifecycle point of view also indicates that in addition to slowly reducing risks for aquatic and terrestrial ecosystems, phytoremediation may have the benefit of net energy production. Furthermore, when atmospheric deposition is included in the analysis, it becomes clear that Pb concentrations are expected to increase rather than decrease (and more so if nothing changes in the site management strategy). Finally, the fact that much higher Pb and Cu (and sometimes Zn) concentrations are found in the deeper soil layers indicates a potential for higher ecological risks than those visible in the surface soil layers. On a purely social level, strong motivations may exist to both adopt active soil-metalconcentration reductions measures (some form of intervention) and to maintain the status quo. There are groups for whom it is important on a symbolic level to take some sort of action, even if the expected results are small. On the other hand, some stakeholders derive economic value from the fields the way they are and may resist changes that would diminish this earning capacity. It is definitely beyond the scope of this project to evaluate these concerns; however, in the process of sustainable development, the concerns of all stakeholders are considered to be valid. From an academic point of view, the literature is full of speculation as to the efficiency of phytoremediation schemes. Researchers can (and do) model many different aspects of these systems, according to many different assumptions and scenarios. However, none of this speculation or these models replaces real-world experience and data. It may therefore be highly valuable from a research point of view to undertake experimental phytoremediation schemes, especially of sites where urgency is relatively low, few realistic options for site management exist, and space is available due to restrictions on land use posed by existing contamination. This applies most especially to inorganic substances, to which natural attenuation does not apply (even if concentrations decrease over time, they are not gone, but simply shifted). One can even imagine experimenting with different vegetation types in the same soil (plant one field each of reed and willow, and mow one field of grass: could be an interesting long-term (t = 20-30 years) study).

Recommendations for a study site

In attempting to use the Demmerik polder site as a case study for the understanding and development of a terrestrial ecological impact model, several aspects revealed themselves to be practical barriers. The greatest of these was lack of data. This is not intended as a criticism, because the investigations carried out at this site were valuable in many ways and have contributed to the scientific understanding of metals in soil ecosystems and their ecotoxicological implications. Furthermore, these investigations were not carried out with the goal in mind to develop a terrestrial model to be used for the diagnosis or prediction of the effects of substance emissions and/or accumulation on ecosystem structure and function over the short and long term. However, in attempting to put the existing pieces into a coherent framework that could be applied to such purposes, it became obvious that crucial bits of information were missing. Based on this experience, the following list can be elaborated of the data that would be useful (in this researcher's opinion, necessary) for the development of such a model:

- At least one measurement should be taken of concentrations of all substances under consideration in representative organisms at each trophic level (e.g. bacterial films, worms, other invertebrates, small mammals, large mammals if applicable, birds if applicable, plant above and below-ground tissues, fungal mycelia). It has been demonstrated that a kinetic approach is more useful than a static, single measurement (which is unable to account for seasonal variations, and/or sublethal toxicokinetic effects), and if it were possible to make several sampling campaigns and measure contaminant concentrations in different trophic levels at several points in the year, that would be ideal (large funding would be necessary for the necessary lab analyses). In the absence of large funding, even one measurement campaign (provided that it included multiple trophic levels) would permit researchers to calibrate existing accumulation models for site-specific parameters, for example, and reduce the uncertainty associated with trophic modeling. This would improve exposure models, may contribute to extrapolations from the individual to the population level, and may improve the ability of site or regional managers to better understand and protect biodiversity.

- Species surveys and site-specific biomass estimates of all significant trophic groups (and compartments, e.g. litter layer) should be made based on field surveys, and careful observation. For organisms to which this applies, behavioral observation and/or estimation should be reported (e.g. estimates of small mammal burrowing activity). This would aid in the estimation of metal stocks, mobility due to trophic accumulation; it would also give modelers a more realistic impression of the site ecosystem and the ways in which biotic interactions drive/influence its development/evolution. (An additional benefit of field surveys is that as researchers practice observing, their skills of observation and intuition develop).

- A complete soil solid-phase characterization should be reported, including solid phase concentrations of all contaminants under investigation. Mineralogical observations of soil particles, and pedological observations of soil profiles and aggregates should be made and reported. This would contribute greatly to the understanding of how (and if) contaminants age, as well as the understanding and prediction of contaminant solubility relationships and substance availability. If high-tech methods are unavailable (due to equipment or budget restrictions, for example) even simple microscopic observation can yield meaningful information. Typical quantification of soil texture is also important (not to be excluded or replaced) but not sufficient. When possible, laboratory experiments including quantifying adsorption behavior, for example, should be carried out using field soils from the site being used for ecosystem model development.

- Measurements and/or observations of the field hydrologic regime (e.g. soil water level, precipitation) should be made and reported. This would contribute to the understanding of contaminant and soil particle mobility, redox zonation, and perhaps multi-stress situations (for example, combination of metal and drought stress). It would also permit modelers to better predict seasonal effects on fluxes such as leaching to ground and/or surface water.

- A complete physico-chemical characterization of soil pore water should be effectuated and reported (at as many different soil layers as is possible). This should include quantification of contaminant concentrations and pore water pH, as well as concentrations of major anions (e.g. sulphate, phosphate, nitrate, chlorine) and cations (e.g. ammonium, calcium, magnesium), or complete elemental quantification (e.g. complete ICP-MS analysis). Special emphasis should be placed on quantifying major nutrients and elements of major importance for soil redox cycling (C, N, S, Fe, Mn). It should also include at least a quantitative, and at best a qualitative analysis of DOC, which is of prime importance for inorganic and organic substance behavior in soil. The applications for these data include reliable inputs for speciation and transport models, understanding of impacts of nurient (e.g. eutrophication) and non-nutrient (e.g. toxic trace elements, acidification) behavior, and understanding and prediction of site biogeochemistry.

- Monitoring or measurement at the site or at a proximate station of meteorological data including precipitation, dry deposition (especially of contaminants) but also of the data necessary as input for evapotranspiration and energy balance models. This kind of data would be useful for predicting responses to changing climatic conditions, as well as for generating realistic data for simple modeling approaches (such as mass balances). It is obvious that the above list is very extensive. The analysis of the data generated during such sampling/surveying/observation would best be undertaken by an interdisciplinary group made up of experienced researchers (professors, post-docs) and students at different levels (undergraduate, Master's, PhD) from many domains including biology (plant, animal, microbial, ecology), pedology/geology/mineralogy, hydrology, environmental/geochemical engineering, and environmental/social management. The benefit of such an approach would be a sustainable balance of expertise, creative energy, idea exchange, and forward momentum in the research process; as well as tangible benefits for local and regional environmental monitoring authorities, who could incorporate data generated during research efforts into regional or national knowledge and databases.

6. MuSA workshop

The participants in the MuSA project organized a two-day interactive workshop with the goals in mind of promoting international cooperation on this issue, engaging in interdisciplinary exchange, and providing interested parties with up-to-date information regarding the current status of the possibilities for an integrated assessment method for evaluating contaminated site management. A summary follows of the workshop aims, presentations, discussions, and conclusions regarding the integration of EcoRA and LCA for contaminated site management.

Workshop aims

Both Ecological Risk Assessment (EcoRA) and Life Cycle Assessment (LCA) may yield useful information on the potential and actual risk of contaminated sites. Nevertheless, neither of these approaches (currently) is able to address all aspects of the risks/damage linked to pollution. For that reason, the MuSA project ("Integrating Multiple Scale Impact Assessment on Ecosystems for Contaminated Land Management"), funded by the SNOWMAN EcoRANET program, aimed at exploring the possibilities and potential benefits of integrating EcoRA and LCA approaches to contaminated land management, especially with respect to the impacts on ecosystems (rather than human health, for example). This workshop is scheduled at the end of the MuSA project, and wants to reflect on the findings of the project and place them in a broader perspective. The goals of the MuSA workshop therefore are to describe the current status of attempts to link LCA and EcoRA and to propose directions for future efforts based on combining the knowledge gained during the course of the project and the knowledge and experiences from research in other, related domains and projects. With this in mind, detailed aims can be identified:

1. To provide an overview of possible scientific approaches, methods, and models that are potentially suitable for linking LCA and EcoRA

2. To evaluate these approaches, methods and models as to their potential to actually achieve the aim of linking LCA and EcoRA

3. To sum up the problems, defects, gaps of knowledge and lack of tools impeding development of strong links between LCA and EcoRA

4. To outline new approaches, methods and models that will improve on forging links between LCA and EcoRA

5. To discuss some case studies that illustrate the above and that may serve as a proof of principle of what can be achieved by linking EcoRA with LCA

The workshop took place over two days and was divided into four sessions, one for each of the four topics mentioned earlier in this report: spatial differentiation of impacts, biodiversity, ecotoxicology, and mobility and bioavailability of metals in soils. Each session was opened by two presentations discussing aspects of the session topic from and LCA and EcoRA point of view. These presentations were followed by brief discussions, which were in turn followed by in-depth working group discussions whose goals were to outline the key aspects of each topic (as they relate to an integrated EcoRA/LCA approach), and to identify research needs with a focus on future progress in an integrative effort.

Participants in the workshop included a well-balanced mix of individuals from research, regulatory, industrial, and consultancy backgrounds, as well as an impressive (for such a small group, 33 participants) geographical diversity (see workshop closing remarks for a list of countries). Feedback from participants was in general very positive, and all participants felt that they had both contributed something from their own experience, and learned one or more new lessons from having participated in the workshop.

List of participants (in alphabetical order by last name): Hélène Beauchamp, Marcel Bovy, Cécile Bulle, Veerle Buytaert, Marieke de Lange, Ana Maria da Rocha Perigo, Miriam Diamond, Maria Diez, Michael Ditor, Gabriella Fait, Daniel Giesen, Laura Guimaraes, Sébastien Humbert, Vladimir Koci, Stefan Kools, Sirpa Kurppa, Christian Langlais, Yves Le Corfec, Gitte Lemming, Jan Paul Lindner, Miranda Mesman, Mark Mistry, Simon Moolenaar, Jérôme Payet, Massimo Pizzol, Tomas Rakovicky, Sina Ribak, Renaud Scheifler, Jaap Struijs, Helias udo de Haes, Kees van Gestel, Nico van Straalen, Grega Voglar

Workshop presentations

The presentations given in the workshop will be briefly summarized below based on notes taken during the presentations. Some of the slides used during the presentations are included as appendices to this report (this is indicated in the presentation summary). The presentations included as appendices are those produced by members of the MuSA project, and can be considered work products related to the MuSA project. Those not included were produced by researchers participating in other projects/professional activities and are thus not considered to be work products related to the MuSA project. Introduction to the MuSA project:

LCA perspective: Jérôme Payet (see Attached file "Jerome WS introduction LCA perspective Final version.pdf" for presentation handout)

No details are given of this introduction, since readers of this deliverable are already familiar with the MuSA project.

EcoRA perspective: Hélène Beauchamp (see Attached file "Intro to MuSA WS_HB.pdf" for presentation handout)

No details are given of this introduction, since readers of this deliverable are already familiar with the MuSA project.

Spatial differentiation of impacts:

LCA perspective: Sébastien Humbert

An overview is given of the historical development of LCA with time: from a global scale (early) to a local scale (recent). For contaminated sites, LCA includes impacts that are upstream of management decisions (by including resources and emissions of materials used in each management strategy). The question is asked whether or not there is a need for consistency (between EcoRA and LCA), or is it logical to have different tools to answer different questions? There is an increasing demand for site-specific (ecotoxicity) evaluations; for LCA/LCIA there is an increasing trend to site-specific evaluations but these models have not been validated at small scales (area < 10000 m2). The pathway of fate-exposure-effect is already included in LCA (providing a link to

EcoRA). An example is given of existing applications of LCA: LCA of disposal alternatives for excavated soil (similar to our Ronde Venen study). One of the main problems of LCA is the lack of reliable data on emissions from soil (leaching, vaporization). Also, when impacts are evaluated at different spatial scales, the priority of substances changes (relative ranking of influence on impact). Traditional LCA does not consider emissions due to leaching from contaminated sites with "no treatment", or the long-term leaching emissions; there is a high level of uncertainty associated with impact assessment related to site-specific leaching (there is a need for site-specific emission rates). Site-specific models can be run in LCA, but serious questions remain as to their validity.

EcoRA perspective: Cécile Bulle

A specific case study is presented: wood preserving oil in telephone poles in Canada, and potential risks posed to soil and/or groundwater. Interactions between substances, soil microbes, and soil properties are investigated. The wide geographical distribution of these poles means their potential impact is high. The first step is to identify which conditions pose threats of leaching to groundwater, and which pose threats of contaminants staying in the soil?

Interactions between substances (oil, PCP) modify mobility; this complicates fate modeling and estimation. The following input data are included in the assessment: emission rates from poles, infiltration of precipitation, microbial degradation (microbial degradation in the presence of contaminants are measured in the lab for different soil properties). Significantly different behavior (in soil columns used in the lab) is found between organic and sandy soil. Without oil, there is almost no vertical migration. If the concentration present exceeds the microbial toxicity threshold, degradation will not take place. Different soil types, for the same contaminant mixture, will have different most sensitive (most at risk) receptors (sandy soils: groundwater, organic soils: human and soil-dwelling populations).

Biodiversity:

LCA perspective: Helias Udo de Haes

The functional unit is a critical issue of LCA; this has to be defined properly. LCA starts from global level; nowadays models are available to go down to a regional or local scale, and also to include dynamic, temporal aspects. Results of including biodiversity in LCA may become arbitrary, because there are so many ecosystems, each having different properties, it will be difficult to give a real description of biodiversity impacts. Attempting to force such an inclusion may weaken rather than strengthen the existing method: “Scientific development is a motor to extend analytical tools beyond their practical limits of applicability”. EcoRA perspective: Nico van Straalen (see Attached file “Nico MUSA220908_NvS.pdf” for presentation handout)

The degree to which soil microbial diversity is unknown (and unknowable) is explored. What is the definition of a species (especially for microbes)? How can we take account of mutations (e.g. due to environmental contamination) leading to new species (species gain rather than species loss)? Is species richness of bacteria ‘valued’ in same way as that of higher organisms (models for higher organisms were shown to apply also to bacteria)? Why not include diversity of microbial habitats? This is done for many purposes, in fact, we speak of biodiversity at the level of species, landscapes/habitats and genes; but for bacteria, habitat diversity is more difficult to define. Which is more susceptible to perturbations: above- or belowground diversity? Soil seems to be a rather robust system, so belowground diversity is rather redundant. Biodiversity has to be valued for its functional aspects but also for its intrinsic value. From an LCA point of view, the intrinsic value seems more important. For soil functioning especially belowground (microbial) biodiversity is crucial. The same is true for soil restoration (e.g. conversion of agricultural soils to nature). This, however, is beyond the scope of LCA. For LCA, the relevance of biodiversity to society is most important, this concerns both functional and intrinsic value of biodiversity.

Ecotoxicology:

LCA perspective: Jérôme Payet (see Attached file “Jerome Ecotox measure in LCA.pdf”

for presentation handout)

Ecotoxicology fits in phase 3 of LCA: impact assessment. How is the effect factor (EF) quantified? The common model is to define $EF = \Delta C x (0.5/HC50)$, and it is expressed as the fraction of affected species per m3 per year. The EF is derived from the slope of the SSD curve between 0 and HC50. The HC50 is chosen because it can be estimated more reliably than benchmarks in the low concentration range (e.g. HC5). The methodology has been applied to compare three different fungicides. A new application is to apply it to heavy metals, specifically copper in water. For this, information on bioavailability, e.g. by means of the BLM, must be included. Data show that the toxicity of copper is heavily influenced by environmental characteristics, e.g. pH and DOC. In extreme conditions, estimations of HC50 may diverge by two orders of magnitude.

EcoRA perspective: Stefan Kools

Risk assessment of a contaminated site proceeds in three tiers. In step 3 SSD's are used to estimate PAF-values for a specific site. In step 3 a Triad approach is applied. Focus can be placed on species or on functions. Terrestrial model ecosystems can be used to reveal the relationship between the two. Two cases illustrate an EcoRA with a Triad approach. Copper and mercury in a residential area: no effects of copper are found, but accumulation of mercury in earthworms indicates an ecological risk. The three Triad dimensions are scaled to a reference site. One link between LCA and EcoRA is that EcoRA provides input to choose among cleanup options (or treatment options).

Metal mobility and bioavailability

LCA perspective: Hélène Beauchamp (see Attached file “Metals in LCA_HB.pdf” for

presentation handout)

Each substance has its own characterization factor in LCA. The fate factor addresses intermedia transfer and the concentration of a substance over time in each medium. Exposure is considered at 100% but could be expressed as a dimensionless fraction between 0 and 1. Possibilities exist to correct for bioavailability by estimating the exposure fraction for each organism (or group). The necessity exists to identify toxic metal species. Many ecosystem properties influence metal mobility and bioavailability. Current models do not address uptake of metals by vegetation. The possibility exists to use a more detailed multimedia model, and to work with a terrestrial model. For many organisms there is a need to integrate seasonal variation.

EcoRA perspective: Rénaud Scheifler

There is equation to link transfer from soil to organisms. Metals can be present in different parts of the organisms, and depending on where they stored, they can be transferred more or less in the food chain. It is difficult to correlate the level of metal in an organism to another level of biological organization (to do so requires extrapolating to the population level). For this, we need spatial information about the area. STARTT program: addressing the transfer in food web at different spatial scales and for different food webs. For that purpose, we need to identify the exposure route of animals. Metals and organic substances are considered in this project. MetalEurope factory: one ton of cadmium was emitted per year in 2001, sometimes one ton of lead per day. Today soil concentrations of lead in gardens can reach 1000 ppm.

The area of interest is 5 km by 8km and three parcels are considered in it. The purpose of

the study is to observe phenomena at a large scale and to explain them at a smaller scale.

Therefore, two scales are considered at the same time. Higher metals concentrations were found in wooded habitats than in surrounding agricultural soils. Observations are given considering the total concentration (higher concentrations close to the factory) and the extractable concentration (high values of which mainly occur in the forest). Landscape factors have an influence on the availability of Cd. The MetalEurope site is a good opportunity to build up a soil model for impact assessment. The duration of many ecotoxicological tests are not long enough to detect an effect. There is also a problem of extrapolation for the organism test result to the population level. Aging of contaminants can be very important in risk assessment with spiked soil or field soils. To detect field-level effects there is a need for more subtle ecotoxicological endpoints and longer test durations.

The partitioning of Cd between animal (small mammal) liver or soil depends on the age and the sex of the mammals. How is it possible to extrapolate effects from the cellular level to the individual or the population level? There is no straightforward relation between metal body burden and toxic effects. Seven years after the end of the activity, small mammals are observed in the very contaminated area: does this represent recolonization or not? Can genetic description of the population answer the recolonization question? Are they sensitive to the effects of high metal concentrations?

Synthesis and closing remarks

Spatial differentiation of impacts

A simple spatial averaging approach does not seem to be indicated; this would tend to weaken the existing strong points of both LCA and EcoRA. For small spatial scales, EcoRA still seems to be the better tool, whereas LCA gives more information about the global scale. One spatial scale that seems like a logical place for them to meet is the watershed level; a model designed at this level, and taking into account for example an ecosystem typology (forest, grassland, crop fields, residential areas), might be a useful approach that could enrich both tools. The prime research need identified for this topic is that both tools would seriously benefit from real-world validation.

Biodiversity

Each tool has its strengths for assessing biodiversity at different scales (although neither is currently equipped to address genetic aspects of biodiversity – this will probably change as more genetic data become available, and as the tools develop). LCA is strong in assessing the simultaneous impact of many different substances in many different concentrations, whereas EcoRA is strong in addressing questions related to specific species, or to organisms inhabiting specific sites. Species-area relationships have been mentioned, this is an example of a link that may help to address issues of biodiversity and spatial differentiation of impacts. More in depth research in this area would be beneficial regarding baseline biodiversity, and the quantitative relationships between biodiversity and ecosystem function.

Ecotoxicology

This was the area where, surprisingly enough, the two tools seem to have the most similarities, especially in terms of input data. The reliability of the output of both tools depends on high-quality input data; whether these are averages or site-specific values depends only on the approach used (EcoRA or LCA) and the spatial scale of the question being asked. Both tools have the same limits when it comes to addressing issues related to ecological responses to toxicity such as community effects, possible thresholds (nonlinear effects, crash and burn), and the effects of complex mixtures of substances on complex living systems. Neither can adequately address these questions, which makes these issues definite research priorities. Other research needs include development of high-quality ecotoxicology databases that are easily accessible and reliable, and the pressing need for more (and better) data on terrestrial organisms.

Mobility and bioavailability of metals

Recommendations regarding mobility include incorporating parameters that are important for mobility into both site-specific and global multi-media models. The abundance of quality GIS data that already exist means that this effort is definitely worthwhile, and that this goal is within reach. The need was identified to improve the quantification of bioavailability in LCA, perhaps by incorporating speciation modeling. Some wellcharacterized sites have been identified whose further study may help us to better understand mobility and bioavailability of metals at the scale of an ecosystem. These include the MetalEurope site in France and the Budels site in the Netherlands. Overall workshop conclusions: can LCA and ERA be integrated? It is clear that in their current state both tools suffer from large uncertainties that could be reduced with better, higher-quality input data. Priorities for the generation of these data include ecotoxicology data (e.g. chronic dose-response data for sub-lethal endpoints e.g. reproduction, especially of terrestrial organisms), reliable data on exposure routes and rates of uptake, and interaction effects (of substances, stresses, and organisms). Examining these two tools from different viewpoints has revealed that each has its strengths and weaknesses. Which tool to use, or in which order to use them, will depend on the question at hand. Overall, is seems that combining the tools themselves is not as interesting as using both to respond to different aspects of a problem (or question). Some of the analogies used to describe this integration during the workshop are that they should “marry” or “talk to each other”. The analogy of a marriage may be particularly apt to describe the current state of the art, with each tool representing a complete and whole individual, and the integration of these two tools in a combined approach representing the relationship that is a marriage, and that enriches each partner. In this way, LCA and ERA can be applied cooperatively, the strength of one complementing the weakness of the other, and vice versa.

It is hard to believe, but in the small group assembled for this workshop (total: 33 people) there are no fewer than 15 countries represented, which are (in no particular order): the Netherlands, Germany, Canada, Portugal, Czech Republic, Finland, Belgium, Denmark, Slovenia, Italy, France, Brazil, Switzerland, the United States, and Spain.

7. Conclusions

The MuSA project has been successful in three main ways: the identification and elucidation of the current status of possibilities for integration of LCA and EcoRA for contaminated site management, successful dissemination efforts with an emphasis on promoting international cooperation, and the identification of future research that would contribute to an understanding of these issues. The complexity of this question and the quantity and diversity of resources necessary for its satisfactory resolution indicate that a large effort is necessary. This effort is justified by the growing need of environmental regulatory agencies, land developers, and nature conservation professionals for comprehensive coherent tools capable of answering the question: what is the most sustainable management strategy for this piece of contaminated land? Despite vast bodies of knowledge, our understanding remains incomplete of how to sustainably manage our terrestrial ecosystems, and of how to ensure they will continue to provide us with the resources and services we require, including the intrinsic resource that is biodiversity. This remains our challenge, and efforts to rise to this challenge in innovative ways are definitely justified. Further efforts are also justified from a research standpoint. As our understanding of our local and global ecosystems grow, so does our awareness that the tools we need to address increasingly complex situations will be found at the interfaces between classical domains of study. This is perhaps the most valuable lesson to come out of this study. Hopefully, the lessons we learn about managing our contaminated ecosystems will allow us to define and reach goals of healthy, sustainable ecosystems.

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V ANTICIPATED USE AND ESPECIALLY APPLICATION OF RESULTS

Deliverable 5.1:

Assessing LCA and ERA for Sustainable site management in a single Framework

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Abstract

In addition with the exploration of possible methods enabling integration of LCA and ERA, MuSA has also the purpose of applying both tools in a single case study in order to provide indications for using at the same time LCA and ERA for contaminated site management. The case study selected was the Ronde Venen site, which is well known in the Netherlands. Nevertheless, important data was missing to enable an Environmental Risk Assessment of the site itself and it was not possible to get these data in the time duration of the project. As a result, the present deliverable is a comprehensive compendium of recommendations plus a demonstration case study. To enable a broad use, the compendium of recommendations is kept general and highlights some key aspects of the outcome of the project following the structure of the ISO standard for Life Cycle Assessment. The case study compares several remediation alternatives with a special focus on impact on ecosystems. Nevertheless, only four metallic cations are considered and only three remediation strategies are considered. At this time, the work is therefore not a guideline but documents with a demonstration and recommendations.

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1 Feasibility and limitation of partial integration of LCA and ERA for contaminated site

1-1 Definition of the scope and goal of the study

This first part of the Life Cycle Assessment study aims at presenting the context and defining the framework of a study.

The main point that has to be addressed in this part is the functional unit. At this stage we are confronted to the first methodological problem of implementing LCA in a remediation strategy. Indeed, the functional unit will be “remediating a given site”. Nevertheless, the main purpose of an ERA study related to the site is to quantify the risk associated with the site itself without any remediation strategy. Therefore, if a remediation strategy is needed, LCA can say which will generate the lesser impact. There is therefore a need to check if the remediation strategy is not even worth than living the site as it is. One of the remediation strategy considered in LCA will then be “doing nothing”, also called “natural attenuation”. It is a paradox in LCA to have a functional unit which is producing impact without being associated with any industrial processes. In order to enable the assessment, it is necessary to change the system boundaries, considering the site itself as an industrial source of pollution and not as an ecosystem. In terms of LCA, this has a clear meaning in terms of system boundaries, it means the site is therefore part of technosphere and not anymore part of the ecosphere. This is a limitation of the compatibility with ERA where the site is considered as a natural media with its own biodiversity. Still in line with the system boundaries, ERA will especially focused on downstream impacts resulting form the emissions of the site itself. LCA will do the same for the alternative “doing nothing” but it will then be necessary to be able to compare the impacts with alternative scenarios considering both upstream and downstream emissions. Working with emissions integrated over time and averaged over space, LCA will easily provide a simple model for assessing impacts of both upstream and downstream emissions, nevertheless, the downstream emissions will need to be validated on a smaller space scale to ensure the coherence with the ERA approach.

1-2 Inventory building

In building up the relation between the product and the corresponding impact, LCA has to develop a Life Cycle Inventory, associating some resources consumption and emissions to each industrial process associated with the product. In our case, the emissions associated with natural attenuation of the contaminated site will be calculated using ERA models more accurate than LCA models. Therefore, one alternative will be based on accurate data while the others will be associated with a higher uncertainty. It is therefore important to estimate the uncertainty of the dataset used for each scenario, and to integrate this uncertainty in the impact assessment procedure.

1.3 Life Cycle Impact Assessment (LCIA)

This part of Life Cycle Assessment is probably the most tricky part for the integration of LCA and ERA. Indeed, LCIA and ERA are addressing similar targets and at this stage, if the input data and the models used are different, they can give opposite results even if they assess the same target. The complicated points are the estimation of the intermedia transfer. It can be estimate on a large scale perspective, but if the water compartment considered in ERA is very different than the water compartment defined by default in LCIA; it will be necessary to check the coherence of the results.

In terms of model accuracy, it is important to mention here the difference between models. The ERA models are more detailed and closely related to site parameters. It is not the case for LCA models, which even at small scale are working at the watershed scale. In order to ensure the coherence between models, it is necessary to perform a site specific assessment in LCA. It is therefore necessary to check specific data related to the site itself. An illustration of the site parameters that can be considered is presented below.

Table 1 : **Site parameters descritpion**

Soil description	**Mineralogy**	**Hydrology**	**Chemical composition**	**SOM**	**Substance availability**	**Connectivity**	**Land Use**
Depth	Bed rock	Precipitation	pH	Content	Total concentration	Interface Swater	Type of use
dev. Stage	sensitive to erosion	Infiltration	Clay fraction	Composition	Exchangeable fraction	Interface GrWater	
Topography	sensitive to salination	transf. surfW	Humic/Fulvic Acid	Ability to form org comlexes		Via dewlling soil org	
Permeability	Granulometry	Transf. underGW	Ability to form mineral complexes	Diss OM		Via mammals	
Texture (sand, loamy, etc)		Saturation?	Redox potential			Via birds	
Air fraction						Dist connect	
Water fraction							
Soil density							
Bioturbation							
Taxonomy							

These parameters have been indentified in the project as the key parameters that are necessary to develop a detailed soil compartment for assessing the impacts of the contaminated site. Furthermore, the uncertainty associated with the data will have also to be quantified, and for that purpose, the type of variability (Cyclic variation, random, linear change, non linear change), will have to be checked with the average and the standard deviation of each parameter.

Furthermore, assessing the transfer of the chemical in the media is only the first step, the next step being the assessment of the impact on biodiversity. As an indication, the tables below present the type of data that can be required for assessing an impact on the biodiversity at the site scale level.

Table 2 : Presentation of the specificities of the plants exposed in the contaminated sites

Vascular Plants	Size (m)	leaf area/m2soil	N species	dominant species	Biomasss	Protected species	reproduction rate	Strategy r/lk	Roots Area/m2 soil
Spermatophyites									
Ptéridophytes									
Species 1									
Species 2									
Species …									

Table 3 : Presentation of the specificities of the animal exposed in the contaminated site

Animals	N species	dominant species	Biomass	Protected species	Mobility area	Reprod. rate	Protect. species	Strategy r/k	Level in trophic chain	N species below	N Species above	Digestive area
Dwelling soil organisms												
Surface non mammals animals												
Mammals												
Birds												
species 1												
species 2												
species…												

These tables illustrate the need of data from the site itself to develop a site specific assessment in parallel with the ERA. Figure 1 below present how to integrate soil parameters and biodiversity data in a single assessment.

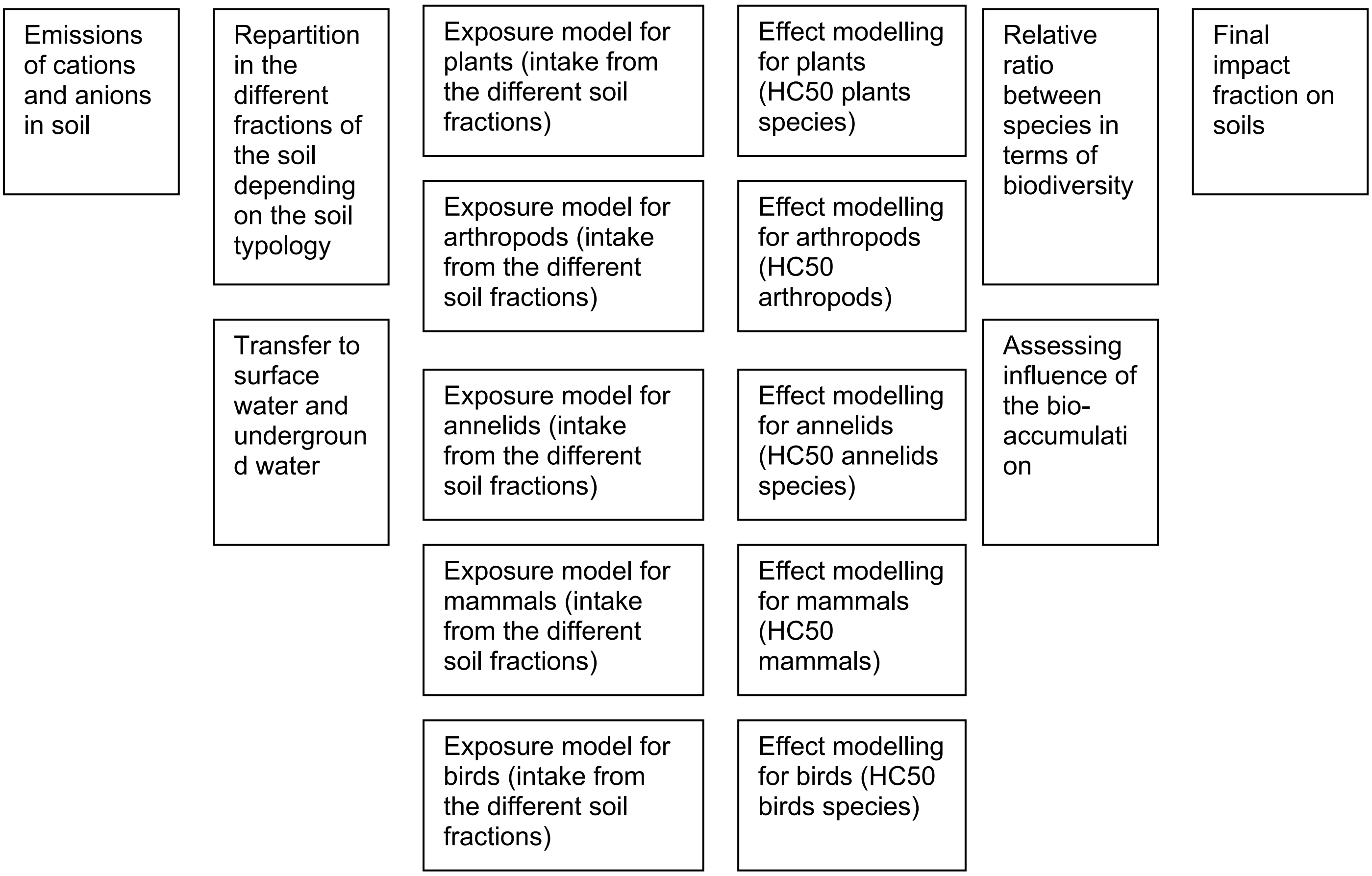

Figure 1 : Integration of the soil and biodiversity data for a site specific assessment in LCIA

The outcome of this assessment can be compared to the results obtained with the ERA approach. So doing, the site specific assessment will provide results interpretable at a local scale.

1-4 Life Cycle Interpretation

The final stage of the LCA has to validate the conclusions of the study regarding the quality of the model used for impact and damage assessment, the uncertainty of the data used for developing the Life Cycle Inventory, and the assumptions considered in the Definition of the scope and goal of the study. In the case where LCA is compared with ERA, interpretation will also have to consider impact assessment on ecosystem based on two different tools. The first limitation will be the number of impact categories covered. Indeed, the scenario “natural attenuation” will only focus on biodiversity and human health while the remediation techniques will have also resources consumption and global warming. This will make the interpretation more complicated. Furthermore, even focusing on the same impact categories for example related to ecosystems, it will be necessary to check the coherence between the different definition of biodiversity that can be considered and to see if the indicators considered by both tools are compatible or not.

2- Site specific study in the Netherlands; life cycle assessment of remediation strategies for Ronde Venen.

2-1 THE NETHERLANDS AND THE POLDERS

2-1-1 Historical and current situation

The central-western part of the Netherlands has been shaped by peat digging and land reclamation in the past centuries and is nowadays marked by the presence of grasslands with remnants of old peat on a floor of old marine and river clays. The region harbours one of the most important low peat marshlands in Europe. The main type of soil use is agriculture (mainly dairy farming), although some protected areas can be found (Rutgers, in press).

The Ronde Venen site belongs to these landscapes. It covers about 115 km^2 and is located in the western part of the Netherlands, about 15 kilometres south of Amsterdam (Bijlmakers et al., 1995). In this area, diffuse heavy metal pollution is found in the top layer of most grasslands, due to a "covering" technique called "toemaaken", which has been applied for hundreds of years, until the first half of the 20th century. It consisted of lowering the water table level in order to gain use of agricultural soils, but it had the effect of oxidising the organic-matter rich soil (when it was not dug to be used as fuel), compacting and degrading the peat layer. Due to this fact, farmers used to spread a mixture of dredged sludge, farm yard manure, sometimes dune sand and large amounts of waste from the surrounding cities like Amsterdam, Utrecht and Leiden on the surface of the soil (Kools, 2006). The mixture was applied to the grassland in a 4 to 7 year cycle (Lexmond et al. 1987). The aim was to amend nutrients and compensate for the lowering of the soils. This agricultural practice, which started around 1600, led to a specific, well moulded, black organic horizon with relatively low clay content, high sand content and a high density. Sand and clay content is extremely variable, depending on the site, as shown by data presented in the next paragraphs. The thickness of this layer goes today from 15 to 50 centimetres. This cover is completely anthropogenic and contains many fragments of pottery, rubble, pipe bowls and other elements that make up household waste (Kools, 2006) and is characterised by high levels of heavy metals, especially lead, copper and zinc. The spreading of metals is patchy and unpredictable with no clear gradient of metal concentration (Rutgers, in press). Analyses in Demmerikse and Blokland polders, in the Ronde Veren area, showed lead concentrations reaching 950 mg Pb per kilo of dry soil (Alterra, 2000) and were compared to Zeevang data (a polder that has not undergone the 'toemaken' process), where lead concentrations around 100 mg/kg were found. In a previous study by Kools et al. (2006) values up to 1400 mg Pb/kg of dry soil were measured in the upper layer (5-10 cm) of Demmerikse polder.

Nowadays, Ronde Venen is a small peat meadow reserve, located close to the town of Vinkeveen. The reserve consists of strips of peat meadow, seprated by ditches that were created to assist peat cutting in the past. The nature reserve was established to protect peat meadow birds like the black-tailed godwit (*Limosa limosa*) and the black tern (*Chlidonias niger*). The water level is strictly regulated and does not vary by more than 10 centimetres through the year (Van der Welle, 2007). The community of Ronde Venen is composed of several smaller centres, and the contaminated site covers several hectares of land. In this study, the focus is on Demmerikse polder, which seems to be the place where most investigations were done and the lead concentrations are the highest.

2-1-2 Brief description of the situation in the Netherlands: general view and legislation

Contamination is defined as the exceedance of the regulatory target value for toxic compounds in an environmental compartment.

For the authorities responsible for contaminated sites, diffuse pollution remains a complex problem, since they can use environmental quality criteria to classify the sites as clean, slightly contaminated or seriously contaminated, but thereafter have access to only a few or perhaps no accepted methods for local risk management (RIVM (c), 2007). In fact, from the point of view of the responsible authorities, the use of quality criteria does not sufficiently solve the risk management problem of ecosystems already contaminated, since cleaning up all sites that are polluted with concentrations exceeding Target Values (TV) (or Intervention Values (IV)) is not realistic due to their huge number and the vast volume they represent in the Netherlands. Other methods are needed to support the authorities for the management of contaminated sites.

When considering both types of regulation (chemical oriented as the target values versus site- or species/ecosystem-oriented as ERA) a possible incompatibility between approaches is suggested (RIVM (c), 2007). Practical methods are needed to assess not only the current status and risk of current site use, but also the ecological perspective for the development and use of contaminated sites in case plans to use the site change. Scenario analyses could have a great potential in this respect, with respect to limitations by local substances as well as ecological potentials in the local context.

2-1-2-1Dutch regulation and Legislation[3]

A brief overview on local regulation is described in order to know tolerable limits and procedures that have to be carried out for contaminated site management in the Netherlands.

In the Netherlands, risk limits for pollutants in soils were studied and implemented in the environmental protection acts, such as the Dutch Soil Protection Act of 1987. The lowest level of risk is defined as the Target Value (TV); the upper threshold of safety levels in the Intervention Value (IV), which means that risk reduction measures are required. Soil properties, like clay or organic matter content, are taken into account in the risk assessment. Between the two values the soil is considered as slightly polluted.

2-1-2-2Environmental quality objectives[4]

The risk policy means that there is a maximum permissible concentration and a negligible concentration (target value), which is in principle 1/100 of the maximum permissible concentration. An

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[3] Based on “Environmental Quality Objectives in the Netherlands”, Ministry of Housing, Spatial Planning and the Environment, 1994

[4] Chapter based on the Dutch legislation (*Environmental Objectives in the Netherlands*, 1994) and the article by Frank A. Swartjes (*Risk-Based Assessment of Soil and Groundwater Quality in the Netherlands: Standards and Remediation Urgency*, 1999)

internvetion value may not be exceeded (unless it is impossible), while exceeding guidance values should be avoided whenever possible.

Target values: environmental quality level of a given compartment at which the risks of adverse effects are considered to be negligible. They indicate the final objectives for the environmental quality to be reached in the Netherlands.

Intervention values: concentration level above which there is a serious environmental pollution which will require clean-up, either in the long term or in the short term.

If a soil contains, for a specific substance, a concentration that is higher than (intervention value + target value)/2 (which gives the so called Intermediate value), it is assumed that there may be serious soil pollution and a further investigation will be undertaken. Consequently, if only the target value is exceeded, no further investigation is needed. The urgency of remediation is present when an average soil volume concentration of at least 25 m^3 is above intervention values. The purpose of determining the urgency of remediation is to distinguish between urgent and non-urgent cases of serious soil contamination. Non-urgent cases are taken up in the provincial soil remediation program without a defined time for starting the remediation, while for urgent cases remediation has to be initiated within one generation (about 20 years) In this category there are priorities based on risk to humans, to ecosystem and of pollutant migration. Finally, economic, financial, social and environmental criteria are considered within the priority group (Swartjes, 1999).

To assess the human toxicological risk, the human exposure to soil is derived by a model called CSOIL (and SEDISOIL for the exposure to sediments), which are based on three main characteristics: pollutant distribution over the mobile fractions of the soil and of the sediment, contaminant transfer from the soil and sediment into contact media and direct and indirect exposure to humans. The ecotoxicological risk assessment is based on two relationships: the first between the soil concentration and the irreversible changes to terrestrial species composition and the second is between soil concentration and negative effects on microbial and enzymatic processes.

The extent of the negative effects will vary among species and range from negligible to severe. An implication of this is that sensitive species are not protected at the level of the (ecotoxicological) intervention value (Swartjes, 1999). Risk for ecosystems and processes, as well as human risks are more-or-less related to the pore water concentration, rather than to the total soil concentration. For this reason, the Intervention Values are corrected for organic matter and clay content.

Because there are no exposure models in use for assessing the risk for ecosystems, a pragmatic procedure has been developed to account for actual risks to ecosystems. In this procedure a matrix has been defined (Table 0.1). Depending on the position in the matrix, an actual risk for ecosystems is assumed to occur if the extent of the contaminated site exceeds a specified surface area.

Table 3 Limit values for surface area as a function of degree of contamination and ecological sensitivity of the area (Swartjes, 1999)

Ecological sensitivity	Soil concentration $< (10 * HC_{50})$	Soil concentration $> (10 * HC_{50})$
High: Nature reserve, 'Ecological network areas'	50 m^2	50 m^2
Moderate: Pasture, Residential areas, including gardens, Residential green spaces, Recreation areas	5000 m^2	50 m^2
Low: City areas without gardens, Arable land, Flower bulb cultivation, Horticulture, Industry, Fallow land, Infrastructures	0.5 km^2	5000 m^2

This limit value for a given surface area is based on the number of species protected as a function of surface area. The surface has been assumed here to represent the space needed to contain the biodiversity of the area category. If this simple procedure does not result in a clear decision on the actual risk for ecosystems, performing measurements (bio-assays) is recommended. However, no standard bio-assays are incorporated in the methodology.

Table 4 shows the target values and the intervention values for a number of metals in Dutch soils. The ecotoxicological data are derived taking into account soil characteristics, correcting for the organic matter and clay content. The formulas can be found in the legislation texts (Environmental Quality Objectives in the Netherlands, Risk assessment and environmental quality division, 1994).

Standard soil (NL)

Composition (%)	Clay 25	Organic 10
	Value	Intervention value
Cd	0.8	13
Cu	36	190
Pb	85	530
Hg	0.3	10
Zn	140	720

Table 5 Soil target and intervention values for standard (NL) and Demmerik soil

Demmerik soil

Composition (%)	Clay 20	Organic 50
	Target value	Intervention value
Cd	1.59	24
Cu	57	301
Pb	120	748
Hg	0.353	12
Zn	185	951

The current environmental situation in the Netherlands is mainly concerned by the problem of the diffuse pollution. Contaminated site and their management need to be supported by new ways of facing the problem and determining the prioritisation of intervention. De Vries (De Vries et al, 2008) writes that to assess current risk and whether current land use is sustainable, tools are needed to evaluate the effects and threats of long term inputs and accumulation of contaminants. In the Netherlands, about 42 500 sites are currently categorised as seriously polluted.

2-2 Site Management

Many technologies to manage and remediate soil and water are nowadays available on the market. In this work the focus will be on the contaminated site of Ronde Venen, more precisely the polder Demmerik, on which available measurements and studies were performed. The brief portrait of the area given in the following chapter helps consider which techniques might possibly be applied.

After selecting the most feasible options, life cycle assessment (LCA) is applied to each scenario, in order to compare the different possibilities and individuate the processes generating the greatest impacts on the environment.

2-2-1 Site characterisation: Demmerikse polder, Ronde Venen

Grassland fields in the Netherlands polder areas are generally several hundreds meters long and around 30 meters wide. They are surrounded by ditches with a width of 1 to 5 meters. Polder Demmerik (or Demmerikse polder) is situated at 52’12’ N and 4’56 E (figure 4.1). It is partly an agricultural area, while another substantial part is a natural reserve. Since waste was spread over the lands, polder Demmerik has a patchy pollution distribution, and previous research has shown there to be high variation in heavy metal concentrations in (Kools, 2006, Bosveld et al., 2000). Detailed study of metal concentrations and spatial distribution in some particular fields (#7, #8 and #10) of polder Demmerik was effectuated by Kools (2006). This project is consequently first based on his data set, and completed by complementary studies and surveys when necessary (references are mentioned).

As mentioned in the previous chapter, in the Ronde Venen case the thickness of “toemaakdek” is variable from 15 to 50 centimetres. The soil lithography is practically homogeneous and organic content dominates its composition. It can be assumed that clay and organic matter content are more or less similar along the soil profile. The spreading of the metals in the polder Demmerikse is patchy and unpredictable, with no clear gradient of metal concentrations. Furthermore, the occurrence of lead concentrations is close to a normal distribution (Rutgers, in press)

A large number of soil samples were analysed to determine main characteristics and their metal concentration. In table 5.1 the results presented by RIVM report (RIVM (c), 2007) are illustrated.

Table 6 Combined data of different soil sampling campaigns in the Ronde Venen area (Demmerikse polder): March 2001, October 2001, march 2003, October 2003 (Rutgers, in press). Mean value with standard deviation (number of samples analysed between brackets).

	Total concentration [mg/kg DW]	Porewater concentration [ug/l]	a concentration [mg/kg DW]
Cd	1.09 ± 0.42 (n=206)	0.93 ± 0.53 (n=21)	0.057 ± 0.043 (n=79)
Cu	165 ± 104 (n=206)	53 ± 22.7 (n=21)	1.228 ± 0. 611 (n=79)
Pb	743 ± 322 (n=204)	12.4 ± 10.8 (n=21)	0.482 ± 0.475 (n=79)
Zn	318 ± 110 (n=206)	365 ± 283 (n=21)	5.792 ± 4.629 (n=79)

Correlations between metal concentrations, soil organic matter content and pH were calculated for the main components at the different sample locations and a positive correlation was found for the presence of lead, copper and zinc, demonstrated by the correlation between metal concentrations in porewater, 0.01 M $CaCl_2$ extractions and total soil content. Oppositely to what can be expected, metals were correlated negatively to organic matter content. RIVM ((c), 2007) explained this fact by the oxidation of peat, which decreases the organic matter content and the bulk matter of soil, lowering surface levels in peaty areas (which is a major concern in the Netherlands).

A study made by Römkens (unpublished data) but used by Salomons et al. (1995) show the measured concentration of cadmium and zinc in soil and soil solution in two situations: in forest and agricultural soils in the Netherlands (averaged values). Under agricultural conditions, the cadmium soil/porewater ratio is 0.35 [mg/kg DW]/ 0.002 [mg/l] (= 175) and for zinc the ratio is 40 [mg/kg DW] / 0.15 [mg/l] = 267.

In the Demmerik polder the ratios are 1172 for cadmium, where although the concentration in the soil is three fold higher, the concentration in porewater is lower than the one presented by Salomons (1995). The ratio for zinc is 871, which means that although the concentration in soil is 8 times higher, the porewater concentration of the contaminated soil is about the double.

To remove all Dutch “toemaakdek” is not considered possible because of the extent and the number of this kind of area and consequently the huge volumes which present these pollution conditions in the Netherlands. In addition, the social aspect is not to be neglected, agriculture is very important, and the landscape management needs to integrate and balance all the interests and the risks concerning these polluted lands (excavation of thick soil layers lead to an outcrop of the water table, creating a pond or a shallow lake).

Different soil management strategies will affect the occurrence of possible effects of metals on terrestrial ecosystems and their leaching through surface water, as is the case today with the hydrological situation, which can lead to oxidation of the peat layer and influence metal mobility when the water table is low. Hypothetically a high water table level would reduce metal mobilisation (RIVM (c), 2007) protecting the soil from the erosion and thus from the destabilisation (as the oxidation of peat) and release of soil components.

2-2-2 Remediation possibilities

Soil (and water) remediation strategies are classified in three general groups: in-situ (no soil disturbance, all processes take place in the contaminated matrix), on-site (movement of contaminated material is limited at the site area, for instance pump and treat technique) and off-site (excavation and transport of the contaminated soil to be treated somewhere else or landfilled).

The preference in a situation such as the Dutch one would be given to the *in situ* techniques as they do not need to move the contaminated soil (which can be considered as special waste and thus has to comply with on stricter laws, depending on the country) and keep intact the original soil structure (even if there are some exceptions as, for instance, some thermal technologies). Despite this, the applicability of every remediation project is strictly linked to site (hydro)morphology, lithography, type and concentration of pollutant, etcetera. On-site (removal of the pollution and treatment on the contaminated site) or off-site (removal and treatment executed in another location) techniques can consequently be more adapted, and often more interesting financially than in-situ ones (remediation without removal). Pollutant thresholds fixed by local regulations are also an important parameter, which always has to be considered when planning remediation.

2-2-2.1 Site physical data

In order to choose the most adapted techniques applicable to the site, a description of the main physical characteristics of Demmerik polder #7, #8 and #10 is the first step.

It is supposed that the thickness of the polluted layer where intervention is needed is 10 centimetres as the measurement campaign was focused on this layer. This is due to the availability of the data for metal concentration in soils, but it does not mean that no pollution is present deeper in the soil. If additional measurements show that the seriously polluted layer (with metal concentrations above intervention values) is thicker, an extension of the functional unit would be possible (increasing the volume of soil that has to be treated). On the other hand, if the deeper soil is not above the values requiring intervention, the system would not change.

Table 7: Main characteristics of Demmerikse polder

Main characteristics	
Surface area to be treated:	25x65m
Thickness of polluted layer:	0.1 m
Soil structure	peaty soil
Bulk density	1100 kg/m3
Soil parameters:	
pH	4.5 - 5
Organic matter (% DW)	50
Moisture (% DW)	70
Clay (% DW)	20
Annual rainfall	700 mm
Net Infiltration	250 mm
Legislation value to achieve	target value, which corresponds to the quality level of a given compartment at which the risks of adverse effects are considered to be negligible
Pollutants	heavy metals (Cd, Cu, Pb, Zn)

The composition of the ‘toemaakdek’ is very heterogeneous, not only in spatial distribution of metal concentrations but also in the size of the particles and objects found, which were present in the city wastes and can still be found in Demmerik soil. If only the most polluted spots are required to be treated, an accurate field measurement campaign should be planned. In addition a first sieving of the excavated soil could be necessary, in order to remove big objects.

2-2-2.2 Retained solutions

To decide which solutions can really be applied to Demmerik polder, some considerations have to be mentioned.

The Dutch landscape is a very particular case because of its origin, often reclaimed from the sea. It means that the water table is very high (near the soil surface) and flooding is avoided only thanks to the drainage system. Without this management, all polders would become small lakes surrounded by the dikes. Figure 0.1 illustrates the situation in the concerned area. It is also important to stress that the shallow depth of the polluted layer avoids the installation of big soil remediation facilities. The Dutch government therefore does not want to excavate the whole soil, because it would engender flooding problems. Despite this, an excavation of top soil layer would be considered as alternative to the other remediation scenarios.

Secondarily, the fact that a part of Demmerik polder is already a protected area, shows that the agricultural use of the land could change to another type of soil management in future years.

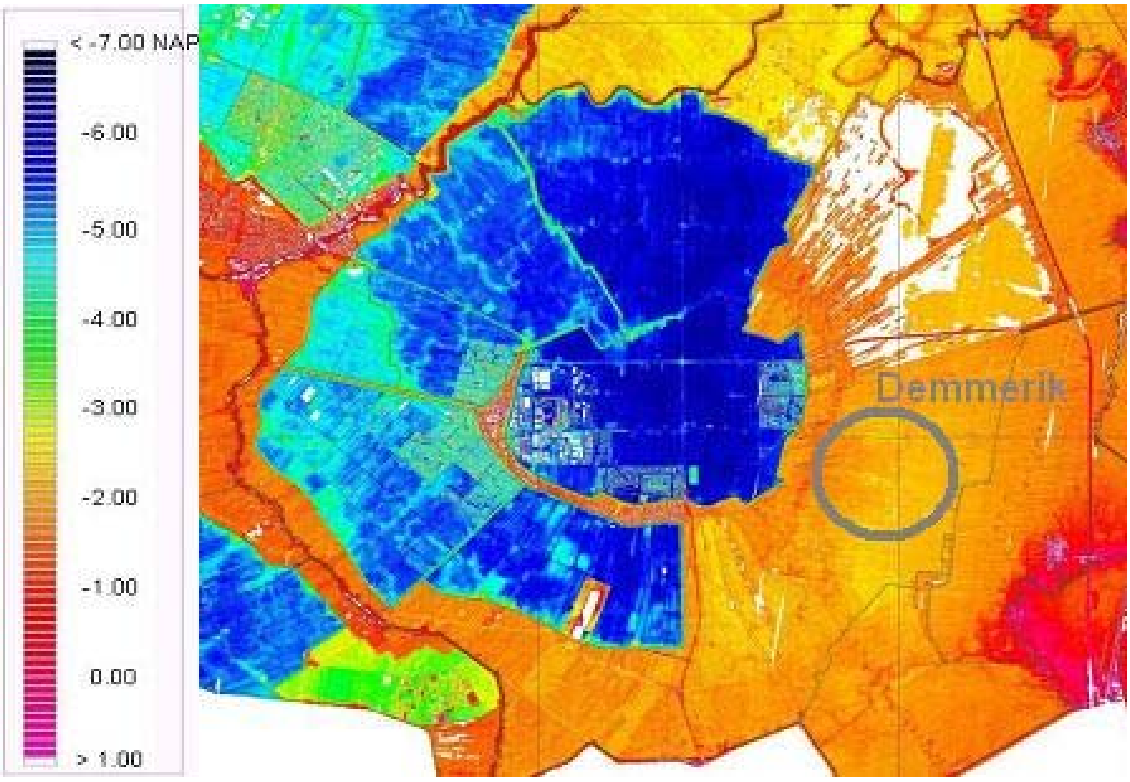

Figure 2: Level of the region around Demmerik site compared to the sea level (Groot Mjidrecht Noord website)

Considering pollutant concentrations found in Demmerik soil, only heavy metal contamination will be considered for this first remediation study. A certain amount of PAHs[5] was also found (RIVM (a), 1996) in this site, but the quantities were low, so that no further research has been done.

Three possible remediation techniques have been identified, which are considered as scenarios in the life cycle assessment in the next chapter: a passive hydraulic procedure, phytoremediation and partial excavation of the most polluted spots, in order to fulfil Dutch regulation limits.

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[5] Fenantrene, anthracene, fluranthene, benzo(a)anthrancene, chrysene, benzo(a)fluranthene, benzo(a)pyrene, benzo(ghi)perylene, Indeno(1,2,3-cd)pyrene

2-3 Excavation Scneario

The first scenario is based on excavation. As no building is present on site, the excavation process does not have any physical obstacle or facility dismantling phase that can constitute a consequent step of the excavation of pollutant soils. The big difficulty in the Netherlands is engendered by the peculiar hydrological management typical of this country. The water table is high, and in the polders the peaty soil shows an important moisture value at field capacity.

The excavation solution has the advantage to be surely efficient (in terms of pollution removal) and rapid. However, as the aim is to achieve target values for all the contaminants, the entire surface polder layer needs to be excavated (lead concentrations are always above 120 mg/kg DW), thus completely changing the soil ecosystem. As the contaminated layer is considered to be shallow (0.1 m), no soil replacement with the cleaned soil is previewed. This is due to the fact that practically any soil cleaning procedure is very destructive regarding the soil structure, which in Ronde Venen is constituted by organic matter.

In this case, as the excavated soil presents contaminant concentrations above the intervention values, it has to be treated as special waste. It is thus hypothesised that the soil after excavation is brought to the special waste incinerator of Rotterdam to be treated and the solid residues remaining after the incineration are landfilled.

The analyses and the monitoring actions are not taken into account for the LCA. The analysis entails only the excavation, transport and disposal proceedings are considered in the reference flux.

The energy put into the burning proceeding of the soil is greater than the heat energy produced by its combustion. No energy allocation is thus calculated.

2-4 Phytoremidiation Scenario

2-4-1 Brief description

Phytoremediation is a technique that presents many advantages: low costs, low energy demand and high public acceptance, which can be crucial for remediation project realisation, depending on the placement of the polluted site (for instance near urban zones or in protected areas). On the other hand, the remediation process is much slower than the intrusive ones (like excavation) and can take up to decades to achieve the desired values.

The plant types that are particularly interesting to use for remediation are those that absorb, store, and under certain conditions, detoxify contaminants.

The aim behind the Ronde Venen site remediation is not to immobilise pollutants, but to remove them from the soil. This entails considering the phytoremediation with the concept of phytoextraction.

Phytoextraction refers to the use of metal accumulating plants that translocate and concentrate metals from the soil to the roots or aboveground plant tissues. Characteristics of plants able to perform phytoextraction include (Gerth, 2000)

- The ability to accumulate and tolerate high concentrations of metals in the harvestable tissues

- Rapid growth rate
- High biomass production

Additional values of this technique are the prevention of soil erosion and the decrease of pollution in the leakage water which can flow to the water table.

The choice of the plant specie consists of making a good balance between its accumulation capacity and its biomass production. Although many hyperaccumulator plants[6] are known, the slow growth rate and the low biomass production are often the limiting factors in heavy metal phytoextraction.

2-4-2 Procedure description

Once the choice of the plant and has been done considering site characteristics, the first step consists of making a simulation in laboratory of the growth of the plant on the studied soil (reproducing site specific conditions). This can be done with more plants species, in order to identify the most suitable plant species or cultivar and determine key factors, such as the total surface removal capacity of the plant for the target pollutant. At the end of the test, information on the uptake per surface unit and per year has to be obtained.

Once the data are found, cultivation of plants can start. To obtain a predefined decrease of metal concentrations, the following calculations can be done

$$A = V_B d_B C_V$$

Where A is the amount of metal removed per hectare [mg/ha], V_B the soil volume [m^3], d_B the soil density [kg/m^3] and C_v the decrease in concentration that is aimed for. To estimate the time period needed for phytoextraction the following formula is used:

$$t = \frac{A}{PB}$$

where A comes from the calculation above, t is the time in years, P is the crop metal concentration [mg/kg DM] and B is the annual biomass production of the crop [kg DM/ha/yr] (Van Nevel et al., 2007).

After plantation, plants require little maintenance. Irrigation can be useful depending on the local climate. In the case of application of phytoremediation techniques, water content plays two different roles: the first one is the water need of the plant, to assure optimal development conditions, the second concerns transport of elements trough the soil. To obtain an efficient pollutant uptake the plant should be in a good situation, and have no lack of macro elements. The uptake rate is also due to evapotranspiration gradient, which creates the flux of liquid and elements from the roots to the aboveground plant tissues. This is why the element uptake would be lower, if the water conditions are not good.

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[6] Plant capable of accumulating more than 1000 mg/kg of dry weight plant for a specific heavy metal. (Roy et al, 2004)

The chosen plant is reed, *Phragmites communis*. Several reasons motivate the choice, but mainly their high performance to absorb heavy metals, nitrogen and phosphorous from the “toemaadek” and the storage of these elements in the aboveground parts of the plants (Gerth 2000). In addition, reed communities grow mostly in fresh but also in brackish and saline water (IHM et al., 2004) practically all over Europe.

The aboveground plants are harvested during winter, and their metal content is analysed and the biomass is brought to a plant for thermal exploitation. In many cases, the first winter season it will not be possible to harvest the plants, thus the harvesting begins only 1.5 years after planting. Normally it takes 1 or 2 years for the plants to achieve the full phytoextraction performance (Gerth, 2000).

Many studies have been carried on this plant, and its wide diffusion all over the world (Allirand et al. 1995). The most similar to the Dutch case is a German case of phytoremediation in Schadlitz, near Leipzig. The site is a former sewage sludge dewatering plant, which received about 300 000 m³ of heavy metal and hydrocarbon contaminated sludge until it closed in 1990. Although in the sequential extraction analysis they found that only 14% of the total metal content was available to plants (adsorbed on surfaces or exchangeable and bound to carbonates, phosphates and sulphides) in the related article by Claus et al. (2007), good efficiency results are claimed.

The approximation described in the following chapter is mainly based on the common characteristics between this case and Ronde Venen.

2-4-3 Scenario description

The phytoremediation scenario is constituted by four main processes, each of them composed by several actions which have to be executed: cultivation on the field, annual harvesting and thermal exploitation of the produced biomass. This scenario is elaborated on the idea of the “best option” scenario, which means that reed plants are acting at the maximum of their accumulation capacity (found in the literature).

2-4-3.1 Starting hypothesis and system boundaries

The system boundaries do not include the growing of test plants in the laboratory in order to test their capacity to grow on the environment of destination. The justification to this choice is the diffused presence of reeds in the polders of Netherlands.

In addition, maintenance, which is normally represented by irrigation and amendments of fertiliser or chelating agents, is considered nil for Ronde Venen, due to the very peculiar conditions of the Dutch polders (drained to avoid flooding) and their eutrophic environment due to past agricultural use. In these very last decades the climate in Europe was changing, especially for annual rainfall and its distribution over the year.

To establish the amount of reed needed to apply the remediation, data about biomass production and metal accumulation have to be found. The literature on phytoextraction is wide, but finding numerical data of an analogous situation is not feasible. The approximation for the Ronde Venen application is thus taken by adapting data of experiments done in the north of France and another in eastern Germany, in order to undergo to similar site climate conditions.

2-4-3.2 Biomass production

A study was conducted on a natural monospecific reed bed located south-west of Paris (France), in the Rambouillet forest (488”N, 1.8”E) along the edge of the ‘Etang Neuf’ mire (Allirand et al. 1995). The time period was three years, and the plants were cut to 5 cm above soil surface during the winter season.

The production levels obtained during the study of Allirand et al. (1995) were between 9 and 13 tons/ha per year, which means between 0.9 and 1.3 kilograms of biomass per square meter. The same study also presented the shoot and root dry matter accumulation in g/plant for one seedling (from Graneli, 1984) showing that November is the most optimal moment to harvest. Summing the biomass production and multiplying by 10 (number of plants for square meters) a smaller result is found (about 0.6 kg per square meter).

Table 8: Shoot and root accumulation in g/plant (Allirand et al. 1995)

	July	August	September	October	November
Above-ground biomass (g/plant)	0.8	9.6	23.0	20.4	16
Roots+rhizome biomass (g/plant)	0	5.0	16	26	32

Similarly, in the German case treated by Claus et al. (2007) in the Schladitz area near Leipzig, the biomass annual production was about 0.1 kg per square meter. It is said that harvesting in December leads to a lower collected biomass, about 0.4 kg per square meters, than if the plants are harvested during October or November (about 1 kg/m^2).

As this option considers the best case solution, a value of biomass production of 1.2 kg per square meter is retained (in the range of the values presented by Allirand et al.,1995).

2-4-3.3 Metal accumulation

To estimate the heavy metal removal by aboveground biomass, the ratio between removal and metal concentrations in soil, data taken from Schadlitz, were extrapolated and compared to other cases discussed in the literature.

Another study was done by Deng et al. (2004), who studied the behaviour of 12 wetland plants in metal contaminated soils. The results for *Phragmites communis* are summarised in the table below.

Table 9: Deng at al 2004, concentration of metals in sediments and in phragmites communis

			concentration mg kg^{-1}		
concentration in sediments mg/kg			underground	aboveground	
Cd	27		10	7.9	
Cu	2171		60	4.2	
Pb	4854		2300	46.0	
Zn	3377		1300	155	

Although the metal concentrations present in the soil studied by Deng (Deng et al., 2004) article are higher than in the Ronde Venen, the same values were taken to quantify the best case time scenario. These values are considered to be under the threshold of toxic effect for the plants.

2-4-3.4 Time estimation

Supposing the ratio between metals present in the soil and the possible uptake as stable (from table 7.2), the estimation of the uptake in aboveground part of the reeds in the Ronde Venen site was calculated. The assumption is that the phytoextraction efficiency does not gradually decrease due to the increasing binding strength of the remaining metals. The results are shown in table 7.3.

Table 10: Estimation of time needed to achieve TV and zero value with phytoremediation

	Cd	**Cu**	**Pb**	**Zn**
Metal concentration in sol [mg/kg DW]	1.09	165	743	318
Metal concentration in porewater [mg/l]	0.00093	0.053	0.0124	0.365
TV corrected for Demmerik soil [mg/kg DW]	1.59	57	120	185
Accumulation capacity for reed plant [mg/kg]	7.9	4.2	46	155
Amount of metal phytoextracted every year [mg/yr]	15405	8190	89700	302250
Initial soil metal amount (whole surface) [mg]	58451	8848125	39843375	17052750
Time to achieve TV [yr]	0	707	372	24
Temps to achieve zero concentration [yr]	4	1080	444	56

2-4-3.5 End of life

After harvesting, the plants are transferred to an incineration centre. In the Netherlands, all incineration plants produce and provide energy (electricity or heating) to the Dutch network. In 2004, 2.535 GWh have been produced and 78% were introduced into the national network, while 22% were used by the plant itself (SenterNovem and Vereniging Afvalbedrijven , 2005).

Phragmites communis harvested in November does not need to be dried to be thermo-exploited. Their calorific potential is around 14’000 kJ/kg (plant moisture content 16 %) (El Bassan, 1998). The amount of electrical energy produced every year by incinerating reed may be:

$$14 \cdot 10^6\ J/kg \cdot 1950\ kg/yr \cdot 1/\ 3600 = 7583\ kWh/yr$$

This is the theoretical production if the yield was 100%. In reality, the yield of thermal production by incinerators is between 70% and 80%, while the same case but with adding cogeneration of electrical power has a yield of 50-60% (Total Company, 2008).

The value entered in SimaPRo , considering that electricity cogeneration is supposed, a global yield of 55% was taken. The kilowatt hours produced are thus:

*7583kWh/yr*0.55 = 4170.65 kWh/yr*

This amount of energy produced is entered in the LCA assessment as a source of co-generated electricity.

2-4-3.6 Limits

The site management decision to apply phytoremediation has to be accurately evaluated within the time scale. Phytoremediation is efficient only if there is no hurry, and at the same time, the maintenance of the processes (harvesting, burning) has to be guaranteed over long periods as well. In reality such a decision is very difficult to defend, especially when the estimated time is longer than 10 years (during that time, the same people can be involved in the decisions concerning landscape management, and thus keep the project running). This is why the evaluation of this scenario counts 24 years of active phytoremediation with reeds and the progressive leaching of the remaining metals in the following years.

2-5 Natural Metal Emission

2-5-1 Adaptation to Ronde Venen

To estimate the evolution of the emission of metals from the studied soil without any action, the theory explained in the Part A of this report is applied to the site of interest: this scenario is called the “do nothing” option. The emitted metals are transferred to the water compartment, where they generate impacts. After having defined a possible evolution of the leaching of metals in time, the LCA method IMPACT2002+ is used, in order to numerically derive the potential damage on the environment and to be able to compare the results with the other scenarios.

2-5-1-1Brief description

In order to estimate the leaching behaviour of the metals in Demmerik polder in the future, the first information to find is: how old is the pollution that has to be treated? To which stage of pollution lifetime does the actual leaching belong?

An historical survey of the related site is absolutely necessary to evaluate which fraction of the pollutant could have already been leached out of the soil. Unfortunately, this is not always possible. Information is not always available and, as in the Ronde Venen case where the past land use is known, the “year zero” is not easy to fix in the time scale.

Visiting and analysing visually the site can give very useful hints. The extremely high heterogeneity and the pieces of ceramics and remains of metallic objects, which are still intact in the polder, support the assumption that Ronde Venen site is a young contaminated site, which has not reached equilibrium yet. As first assumption the “year zero” is thus assumed to be at the present time.

The evolution of the metal emission is based on their interaction with the component (abiotic and biotic) of the soil. An average of several sequential extraction analyses have been considered in order to find the percentage of the metals bound to the specific fraction. The fractionation used is based on literature data and is illustrated in table 8.1.

Table 11: Percentage average partitioning of metals in soils. (Literature data from Salomons et al. 1995, Klaus et al. 2007, Alvarenga et al. 2007)

Element	Easily soluble, exchangeable, carbonate bound	Bound to organic matter	Fe/Mn - Oxides bound	Residual fraction
Cd	31	21	30	19
Cu	5	58	6	31
Pb	9	20	24	48
Zn	22	15	39	24

2-5-1-2First phase (P1)

In the first period easily soluble, exchangeable and carbonate bound metals are leached out of the soil. As the pH of the soil is between 4.5 and 5, it is assumed that the carbonate fraction is also leached out in the first period (see table 3.1). To derive the time required to achieve this value, pore water concentrations measurements on site are supposed to be constant during this first phase. To know the fraction that belongs to this category, several sequential extraction results were averaged (Salomons et al., 1995, Claus et al., 2007, Alvarenga et al., 2007) (Table 8.1).

The annual rainfall in Holland is about 700 mm. The annual discharge can be approximated with the net infiltration rate of 250 mm/year, the rest of the water is mainly evaporated (Van Beek et al. 2007). After an annual cycle, the soil water balance is supposed to be in equilibrium, or presenting negligible differences in porewater stock compared to one year before. Consequently, the discharge per square meter has to be equal to the net infiltration.

The amount of metals present in the study area (cadmium, copper, lead and zinc), porewater concentration and the derived length of the first phase (P1) are shown in table 2.2.

Table 12: Initial concentration in soil and porewater for the Demmerik polder (RIVM, 2007). Fraction of exchangeable/soluble metal and time needed to leach out this fraction

	Total concentration [mg/kg DW]	Porewater concentration [µg/l]	[% w/w] of metal contributing to Phase1	Length of P1 [years] (constant leaching)
Cd	1.09	0.00093	31	48
Cu	165	0.053	5	21
Pb	743	0.0124	9	712
Zn	318	0.365	22	25

Modelling the first leaching period of the site shows that copper would reach the stable value in a relatively short time (with respect to the other metals). In the previous chapter, the landfill model by Obersteiner et al. (2007) was presented and the data measured on the different landfill show the same behaviour.

2-5-1-3Following phases

The second phase concerns the mineralisation of organic matter, leading to a release of the metals embedded in this fraction. As previously mentioned, it is extremely difficult to estimate the lifetime of soil organic matter.

The main phenomenon which influences organic matter degradation is its oxidation. As already seen at other sites similar to the Dutch polders (ex. Plaine de l'Orbe, Switzerland), when the water table level is lowered too much, the oxidation can occur very quickly[7].

The Netherlands Environment Assessment Agency (2005) studied the consequences of climate change in the Netherlands. Even if uncertainties are present, this study gives a range of temperature values that will be achieved in the country in one century and the evolution of precipitation. One to six Celsius degrees higher average temperatures will be theoretically measured in 2100, and the sea level around the Dutch coast will be from 20 to 110 cm higher. Annual rainfall will also increase, and the rain distribution will tend towards an increasing number of extreme events. On the other side summers will be much drier. The combination of all these factors brings to an impact on Dutch territory. It is estimate that, due to the dryness and thus to the shrinking and oxidation of organic matter, the lowering of the surface level on the polder could achieve extreme rates of 1 centimetre per year. Actions to fight this phenomenon have already been studied. Diminishing the drainage could an efficient measure.

Considering that the above mentioned soil dynamic will happen, but with one tenth of the intensity, the first 10 centimetres of soil will be lost in 100 years. This value is taken as representative of the loss of organic matter in the leaching model, and consequently also of the metals which were bound to that fraction. Also Doka (2003) supports the idea that in the long time spans considered, erosion is a probable influence on landfill development. For comparison, Doka ((d) 2007) mentioned a study about the erosion for Swiss soil going from 5 to 100 centimetres in 1000 years (without considering climate change influence).

For oxides the problem is even more complicated. No estimation of Fe and Mn oxides half lives are found in the literature. Here an arbitrary time of 10 000 years was taken as the standard value. As a final point, all the metals present in the soils are assumed to be released after 60 000 years. In the figure 8.1, the curves describing the leaching behaviour are shown.

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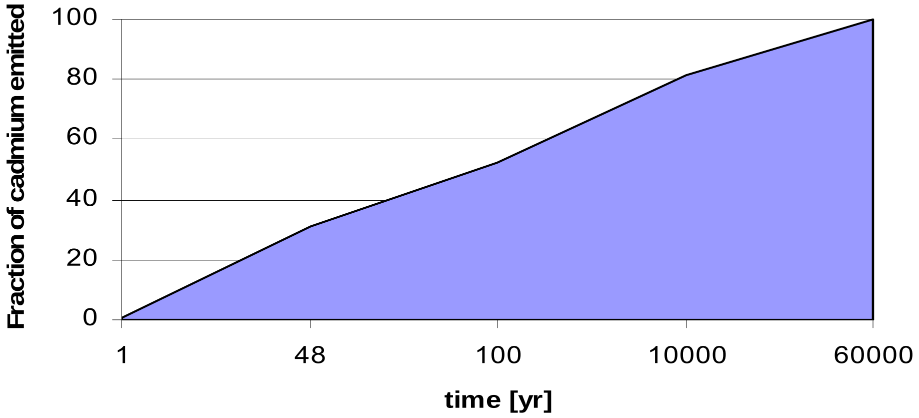
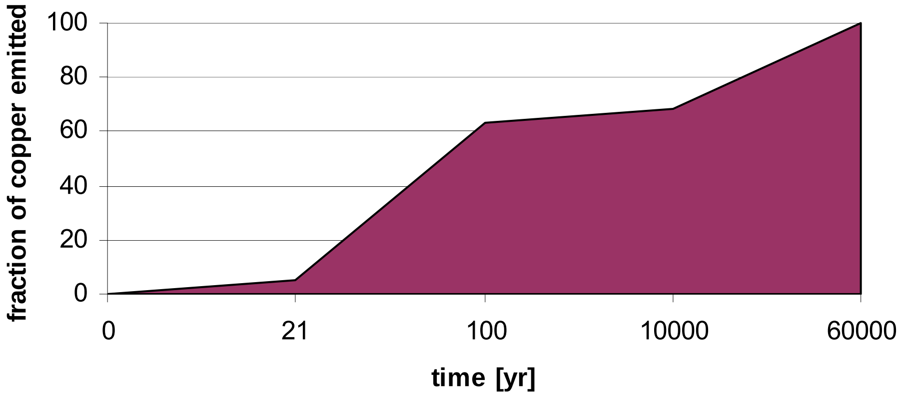
[7] Starting from the drainage installation in 1940, the surface level of the soil in the Plaine de l'Orbe decreased by about one meter in 50 years. This subsidence was due to the oxidation and thus increased mineralization of organic matter (Mermoud, 2006).


Fraction of cadmium emitted
100
80
60
40
20
0
1
48
100
10000
60000
time [yr]


fraction of copper emitted
100
80
60
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20
0
0
21
100
10000
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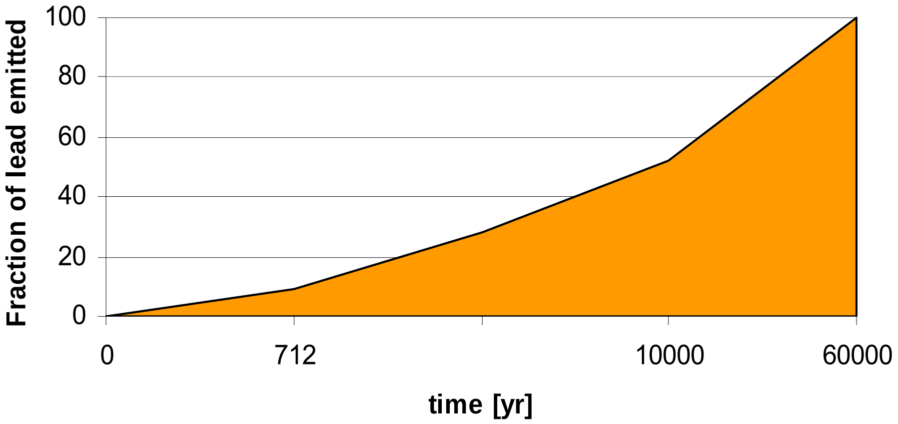
time [yr]


Fraction of lead emitted
100
80
60
40
20
0
0
712
10000
60000
time [yr]

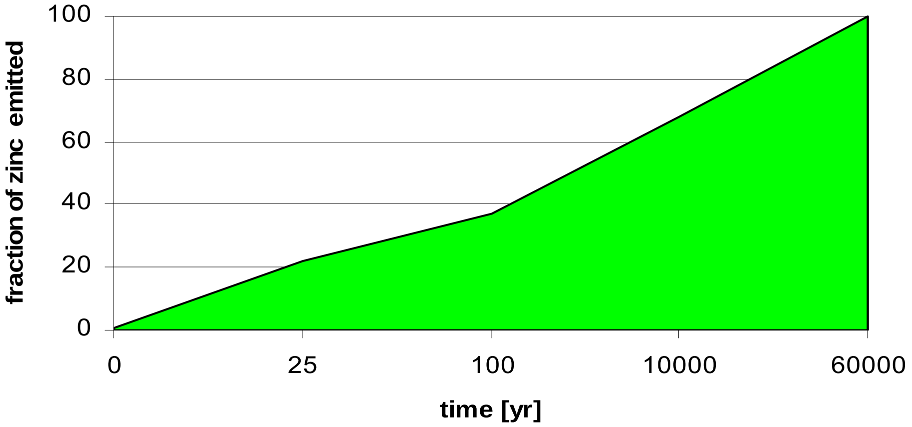


Figure 3: Evolution of the cumulative fraction of total metal emitted during time

Looking at the first time frame (0 – 100 years) it appears clearly that only a minimal part of the lead and zinc content is leached, although cadmium shows a stronger leachability, confirming Obersteiner's model discussed in chapter 2.3. Due to copper's important exchangeable fraction and its affinity for the organic matter, it is emitted in the next 100 years. The steepness of the curves is higher for cadmium and zinc, showing a larger easily extractable fraction, as found with the data of Ronde Venen in chapter 2.1.

In the second (up to 1000 years) period the affinity of copper for organic matter is easily visible, from the steepness of the increase of leaching substance. After 100 years about 50 % of Cu and Cd is still present in the soil, while the fraction is bigger than 70% for zinc and lead. The latter is particularly recalcitrant to leaching and after around 600 year about 90% is still present. After 1000 years between 60% and 80% of all metals except lead have been emitted. Lead is around 40%. After 60 000 years all metals are supposed to have been emitted from the soil to the water.

Figure 8.2 represents the same curves together with a logarithmic scale. The representation allows comparing the global behaviour of the four metals. While zinc has a very straight evolution, copper and cadmium emissions seem to accelerate just before 100 years (due to the organic matter disappearing), lead shows clearly a different trend, and more recalcitrance to emission.

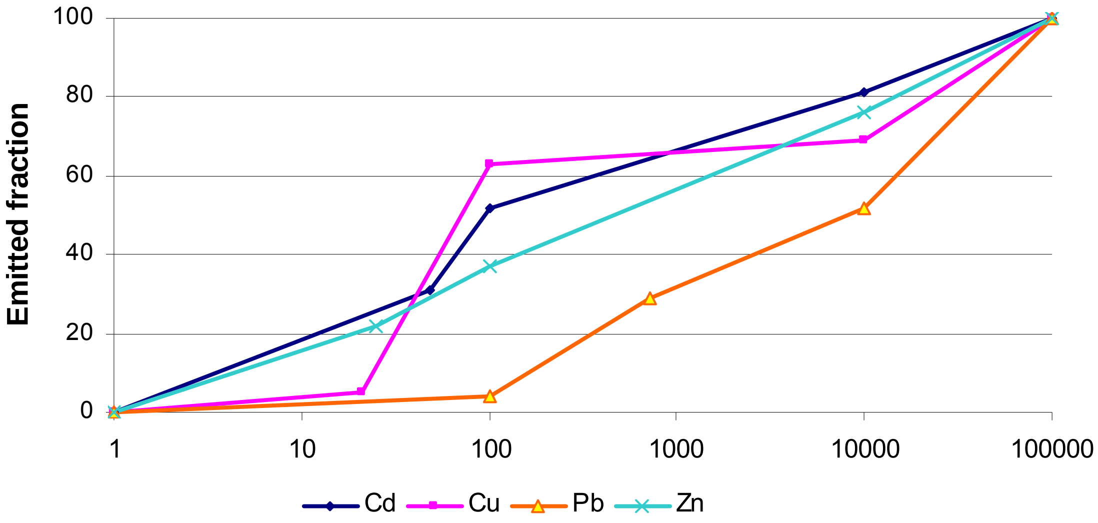


Figure 4 : Representation in logarithmic scale of the evolution of metals emission for the Ronde Venen application

For the time periods shorter than 100 years it is showed that around 40-60% of zinc, cadmium and copper is already leached out of the soil, while most of the lead is still present in the soil matrix.

2-6 Life Cycle Inventory

2-6-1 Purpose

The aim of the application of the LCA to the three remediation strategies established for the Ronde Venen site (the phytoextraction option, the excavation option and the “do nothing” option) is to compare their impact on the environment. Monitoring of the pollution on the site is considered equivalent for the three scenario, and it is thus outside of the system boundaries. Life Cycle Analysis will be performed according to the rules established by ISO (ISO 14040 to ISO 14044).

2-6-2 Functional unit

The first step is to define the functional unit. The remediation of soils is considered a service, which provides a soil not polluted with zinc starting from the soil of the Demmerik polder, parcel #10.

The choice of focusing on zinc levels is based on two main reasons:

- The feasibility of the remediation and the length of time needed by the phytoextraction scenario. It would make no sense to compare the remediation to be applied for more than 700 years (time needed to have all the metals under the intervention value).
- The unchanged excavation procedure. If the soil has to be treated only for one metal or for more, the procedure will be the same.

The functional unit is thus 0.1625 hectares of soil with metal content under the target values on 10 centimetres of depth.

The second step consists of defining and quantifying the reference fluxes (materials and energy needed to achieve the aim) in the life cycle inventory and the third phase is the application of life cycle assessment calculations with the consequent results analysis in order to individuate the greatest impacts engendered by these actions.

Table 13: Main characteristics of the functional unit

Functional Unit		
0.1625 hectares of RV soil	53625	Kg (dry soil)
Surface area to be treated:	25x65m	
Thickness of polluted layer:	0.1 m	
Soil structure	peaty soil	
Bulk density	1100 kg/m3	
Dry density	330 kg/m3	
pH	4.5 - 5	
Organic matter (% DW)	50	
Moisture (% DW)	70	
Clays (% DW)	20	

The functional unit is defined as the service « cleaning 0.1625 hectares of Demmerik polder », where cleaning means the achievement all target values for the studied pollutants. The system boundaries exclude site characterisation, as this first step would be practically identical for all the techniques applied in the three scenarios. What is more, the analyses during the execution of the remediation do not represent a considerable action and contribute to the global emissions in a negligible amount.

Table 14: presents the dimensions of the study area, the polder #10 of the Ronde Venen site.

Width/Length/Depth [m]	peat bulk density [kg/m3]	total polluted volume [m3]	Polluted weight [m3]	avg moisture [kg/kg DW]	dry weight [kg/ m3]
25/65/0.1	1100	162.5	53625	0.7	330

For both plants and polluted soil an incineration is programmed. Rotterdam, which is 60 kilometres north-west from Ronde Venen, is the location of the incineration. This decision has been taken on the basis of the real situation, as Rotterdam provides incineration of municipal waste and special waste.

An important fact that has not been considered is that when the plants are burnt (producing energy) the metals contained are neglected (plants are considered as not presenting metal concentrations exceeding the regulations).

It must be stressed that studies and analyses of the polder soil were done only on the first 0.1 meters of depth (Kools, 2006), and consequently, the whole life cycle assessment is based on the assumption that the pollution is present only in this layer.

As the functional unit is the change from an elevated soil metal concentration to a lower one, the application of different remediation methods do not take the same laps of time. Especially in this case study, the time scales are extremely different depending on the scenario. In fact, for phytoremediation the process to lower zinc concentrations below the intervention value takes 24 years, while the excavation and treatment process takes only a few weeks.

2-7 Reference flows

2-7-1 Excavation

The excavation scenario is composed of three phases, the excavation itself by hydraulic digger, the transport to incinerator of special waste in Rotterdam and the incineration process. Table and figure 2.3 summarise the materials and the processes needed.

Table 15: Inventory for the phytoextraction scenario

Material/assemblies and Processes		
Excavation, hydraulic digger RER/U	162.5	m3
Transport, lorry 28t CH/U	10725	tkm
Adapted disposal hazardous waste	71428	kg

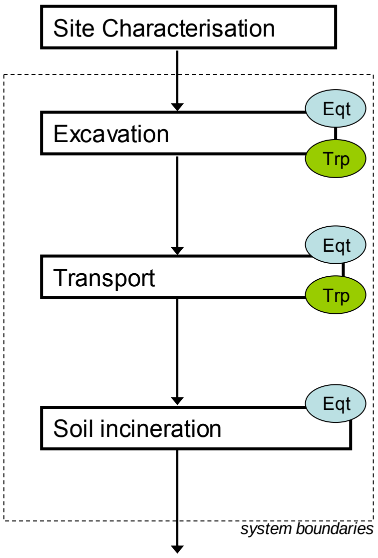


Figure5 : Excavation scenario reference flux

In order to have a representative incineration process, the flux of substances and elements entering the incinerator has to reflect the composition of the contaminated soil which has to be burnt.

The disposal option called "*Disposal, hazardous waste, 25 % water to hazardous waste incinerator*" includes waste-specific air and waster emissions from incineration and auxiliary material for flue gas cleaning. Short term emissions to river water and long-term emissions to ground water are derived for residual material landfilling (from solidified fly ashes and sludge). The treated waste composition as it is composed in the Ecoinvent dataset is (wet, in mg/kg):

Table 16: Waste composition of the process , hazardous waste, 25 % water to hazardous waste incinerator. Ecoinvent dataset.

Element	[mg/kg]	Element	[mg/kg]	Element	[mg/kg]	Element	[mg/kg]
H2O	250000	F	3700	V	N/A	Ag	N/A
O	40000	Cd	0.37	Be	N/A	As	N/A
H	61000	Co	74	Sc	N/A	Ba	N/A
C	416000	Cr	123.95	Sr	N/A	Mn	N/A
S	32000	Cu	267.47	Ti	N/A	Mo	N/A
N	7400	Hg	0.74	Tl	N/A	Al	N/A
P	2200	Zn	2378.3	W	N/A	Sb	N/A
B	7	Ni	126.81	Fe	N/A	Se	N/A
Cl	104000	Pb	296.64	Ca	N/A	Sn	N/A
Si	80425			K	N/A	Na	N/A
				Mg	N/A	I	N/A

Is it considered that 1 kg of this waste produces 0.189 kg of residues, which are landfilled (with 0.07561 kg of cement for the additional solidification, based on Swiss averaged values, Ecoinvent dataset).

The adapted waste incineration scenario is based on the waste disposal called *Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH S.* As the waste scenarios are blocked in SimaPRo it was not possible to modify the treated waste composition. A new waste scenario base was created, copying all the emissions to air, soil and water compartments, and editing only lead emissions, as the input in this study would be higher than the one hypothesised by Ecoinvent database. The factor of change for lead emission was 1.88. In fact, the lead concentration in the soil is 743 mg Pb kg^{-1} DW. One ton of soil is 300 kg/m^3 DW, thus there are 0.223 kg Pb t^{-1} wet. If the soil is let to dry until 25% of the initial water content remains (as required by the process of the Ecoinvent dataset), for 1 ton of soil excavated there are 400 kg of wet soil with 25% moisture that goes into the incinerator. In this case, the lead concentration would be 0.558 kg Pb t^{-1}. As in the dataset the entry Pb concentration is supposed to be 0.3 kg Pb t^{-1}, a correction value of 0.56/0.3 = 1.88 is applied.

2-7-3 Phytoremediation

For the phytoremediation scenario the reference flux for 24 years is illustrated in the figure 9.2 and table 2.5. Planting is the first phase, which is counted in the LCA as the amount of seeds necessary and tillage. Harvesting is composed of mulching and swathing. After the transport to Rotterdam by lorry, reed plants are burnt and electricity cogeneration takes place.

As the Ecoinvent database does not mention “reed seeds”, the product “wheat seed” was taken. Although it seems an arbitrary choice, this assumption is justified by the awareness of the small impact it has compared to the other processes. It is also important to note that if the seeds turn out to be significant in any impact category, the reference flux could be changed with a more precise material for the reed seeds, and the impact re-calculated. It is not the case here, as among the materials or assemblies of the reference flux, it is the group with the smallest impact in practically all midpoints.

Table 17: reference flux for phytoremediation scenario

Material/assemblies and Processes		
Wheat seed IP, at regional storehouse/kg/CH	50	Kg
Tillage, harrowing, by rotary harrow/ha/CH	3.9	Ha
Mulching CH/U	3.9	Ha
Transport, lorry 28t CH/U	2808	Tkm
Swath, by rotary windrower CH/U	3.9	Ha
Wood chips, burned in cogen 6400kWth, emission control/MJ/CH	360360	MJ
Electricity, medium voltage, production UCTE, at grid/kWh/UCTE	-100100	kWh

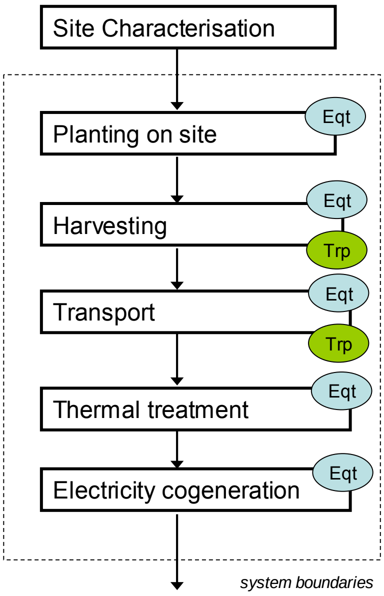


Figure 6: phytoremediation scenario reference flux

2-7-4 Natural metal emission

This option consists in doing nothing. Consequently, the reference flux of this scenario is empty.

2-8 Life cycle impact analyses

In this chapter the application of the LCA to the Demmerik remediation is shown. The Ecoinvent database is used; however three midpoint categories were omitted on purpose: terrestrial ecotoxicity, terrestrial acidification/ eutrophication and land occupation. The reason is that further investigations have to be executed in order to confirm or improve these factors concerning the impacts on terrestrial ecosystems (Haye et al., 2007). The damages are thus considered only on the water compartment. As already mentioned, in the Netherlands, water quality is the big issue for its functions and use in the country (de Vries et al., 2008).

The endpoint "Ecosystem Quality" that is normally found using IMPACT 2002+ method is composed by the midpoints 'aquatic ecotoxicity', 'terrestrial ecotoxicity', 'terrestrial acidification/eutrophisation' and 'Land occupation'. It has consequently been named "Impact on water" because in this report it is composed by the assembly of the midpoints "Aquatic ecotoxicity", "Aquatic eutrophistion" and "Aquatic acidification". In order to characterise these midpoints (i.e. to transform the kilos of SO_2 equivalent of the aquatic acidification, to PDF*m^2*yr), the used damage factor values are based on the latest research in the LCA domain (Tirado 2007, unpublished data) (table 2.6).

Table 18: Damage factors for impact on water Endpoint

Impact category	Factor	Units
Auatic ecotoxicity	$5.02*10^{-5}$	PDF*m^2*yr/kg TEG water
Aquatic Acification	$6.5*10^{-2}$	PDF*m^2*yr/kg SO_2
Aquatic Eutrophication	$3.11*10^{-7}$	PDF*m^2*yr/kg PO_4-P lim

The human health endpoint category is composed of 6 midpoints: substances having carcinogenic or non-carcinogenic effects, organic or inorganic compounds affecting respiration, substances engendering ionizing radiation and substances that are supposed to deplete the ozone layer.

First of all, the analyses of the two scenarios are considered separately, in order to find the major contributions to the impact on the endpoints and the 12 midpoints calculated with the Ecoinvent dataset.

2-8-1 Excavation of the soil

The analysis has been applied to evaluate the impacts on the environment by the excavation option. Table 9.7 shows the endpoints of this scenario for the three main phases involved.

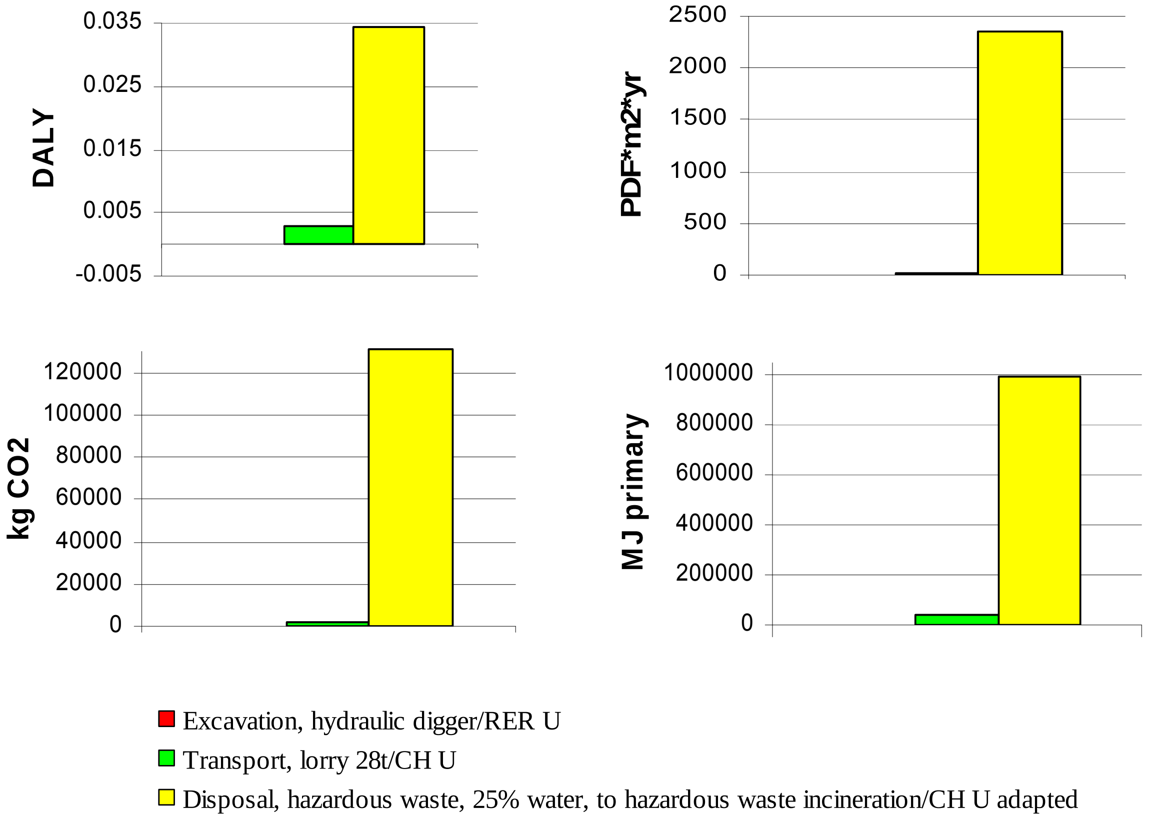


Figure 7: Damages for the three processes involved in the Excavation scenario

Table 19: Impacts generated by the excavation scenario

Damage category	Unit	Excavation, hydraulic digger	Transport, lorry 28t	Adapted Disposal, hazardous waste, 25% water, to hazardous waste incineration
Human Health	DALY	0.0002	0.0027	0.0343
Impacts on Water	PDF*m^2*yr	0.4	11.9	2355.8
Climate Change	kg CO_2	84	2328	131142
Resources	MJ primary	1312	39917	993418

In this case the impact is almost entirely due to the adapted waste disposal of the contaminated soil for all the damage categories, although the contribution of the transport mustn't be neglected, especially for the human health category. Figure 9.4 shows the relative results of the midpoint impacts.

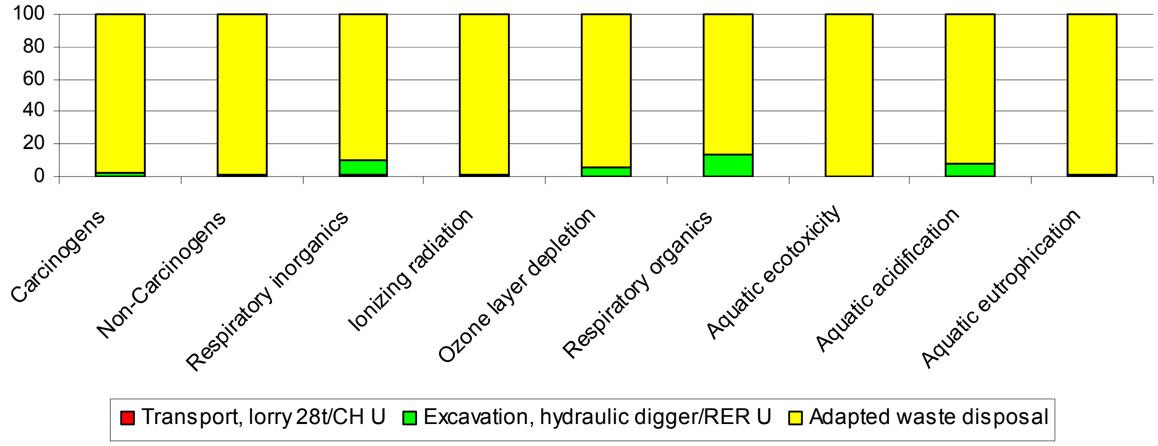


Figure 8: Category of impacts at midpoints, excavation scenario analysis

Human Health

The adapted waste disposal is the process that impacts the most human health. Its impact is at 96% due to the impact of respiration due to inorganic substances, as nitrogen oxides (responsible of 41% for this midpoint category), followed by particulates with diameter smaller than 2.5 µm (30%) and sulphur dioxide (27%) (all emitted in the air compartment). The processes that contribute the most to the impact on human health are a large number of processes related to the waste disposal, and presented in the appendix II, as there is no dominant process; many of them contribute for a small fraction of the impact.

Table 20: Human Health midpoints results for the processes involved in the excavation.

Impact category	Unit	Total	Excavation, hydraulic digger	Transport, lorry 28t	Adapted disposal, hazardous waste, 25% water, to hazardous waste incineration
Carcinogens	DALY	1.9E-03	2.1E-06	4.5E-05	1.9E-03
Non-Carcinogens	DALY	7.7E-03	1.6E-06	8.2E-05	7.6E-03
Respiratory inorganics	DALY	2.7E-02	1.6E-04	2.6E-03	2.4E-02
Ionizing radiation	DALY	3.5E-04	8.1E-08	4.2E-06	3.5E-04
Ozone layer depletion	DALY	8.2E-06	1.1E-08	4.1E-07	7.8E-06
Respiratory organics	DALY	4.4E-05	1.9E-07	5.5E-06	3.8E-05

Impact on water

In the aquatic environment, ecotoxicity generates the most important impact, followed by acidification and eutrophisation, which are negligible compared to the other impact of the on the aquatic ecotoxicity.

Table 21: Impact on water midpoints results for the processes involved in the excavation.

		Total	Excavation, hydraulic digger	Transport, lorry 28t	Adapted disposal, hazardous waste, 25% water, to hazardous waste incineration
Aquatic ecotoxicity	PDF*m^2*yr	2353.1	0.3	10.9	2342.0
Aquatic acidification	PDF*m^2*yr	14.9	0.1	1.1	13.8
Aquatic eutrophication	PDF*m^2*yr	3.57E-06	2.54E-09	4.96E-08	3.52E-06

The adapted waste disposal is again the process responsible for more than 99% of the impact on water and 92% of the impact on aquatic ecotoxicity. In this process there are two substances which represent 43% and 37% of the whole waste disposal impact on water: cobalt and copper (ion), both emitted into the water compartment. Zinc and nickel emitted into water are the following substances in order of contribution to the impact, although they contribute only about 10%.

Summary and comments

In this scenario, the end-of-life step of the remediation activity is the process that creates the greatest impacts on the human health and on the water environment. The variation of the impacts expressed in PDF*m^2*yr is important: aquatic ecotoxicity results are more than 3 orders of magnitude greater than those of acidification and 9 orders of magnitude than those of eutrophication.

Analysing all the substances that play a role in the whole incineration process (whole incineration[8] life cycle, with allocations) it is found that cobalt generates the main impact, followed by copper (almost the same value of PDF*m^2*yr) and zinc (about the half of the impact). Cobalt comes from the fuel and oil combusted in the process linked to the incineration.

2-8-2 Phytoremediation during 24 years

The phytoremediation option is composed of three main processes: phytoextraction, transport, and the incineration of the plants and the co-generation of energy. The impacts generated by this

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[8] 'Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH U' in the Ecoinvent dataset

scenario have been derived and the four endpoints are shown. In table 9.10 the impact on the four endpoints is shown.

Table 22: Impact per endpoint for phytoextraction scenario

Damage category	Unit	phyto24	bonus energie	Burning of reeds
Human Health	DALY	0.00	-0.03	0.01
Impacts on Water	PDF*m^2*yr	7	-141	197
Climate Change	kg CO2	995	-47207	2242
Resources	MJ primary	16846	-1067393	18637

The burning of the plants has a negative impact value (beneficial), which means that it avoids other more polluting processes and consumption of other natural resources to produce the same amount of energy. The impact of the process of combustion of reeds is the most important on water and on human health. Less difference between the remediation phases is found for the climate change and resources use values, where the reeds also present an impact. The difference between the two categories is small, especially for the resource consumption, expressed in primary megajoules.

The focus is put on the impacts on the aquatic ecosystems and the human health. It clearly appears that the burning of reeds has the most relevant contribution to almost all of the midpoints. Exception is made for non-renewable energy, mineral extraction and the respiration of organic compounds, where phytoextraction (especially harvesting) has almost the same impact as the burning of reeds.

In order to find the origins of the impacts, the 12 midpoints used in this study have to be analysed; figure 9.5 helps the visualisation of the midpoints that contribute the most to the impacts on environment.

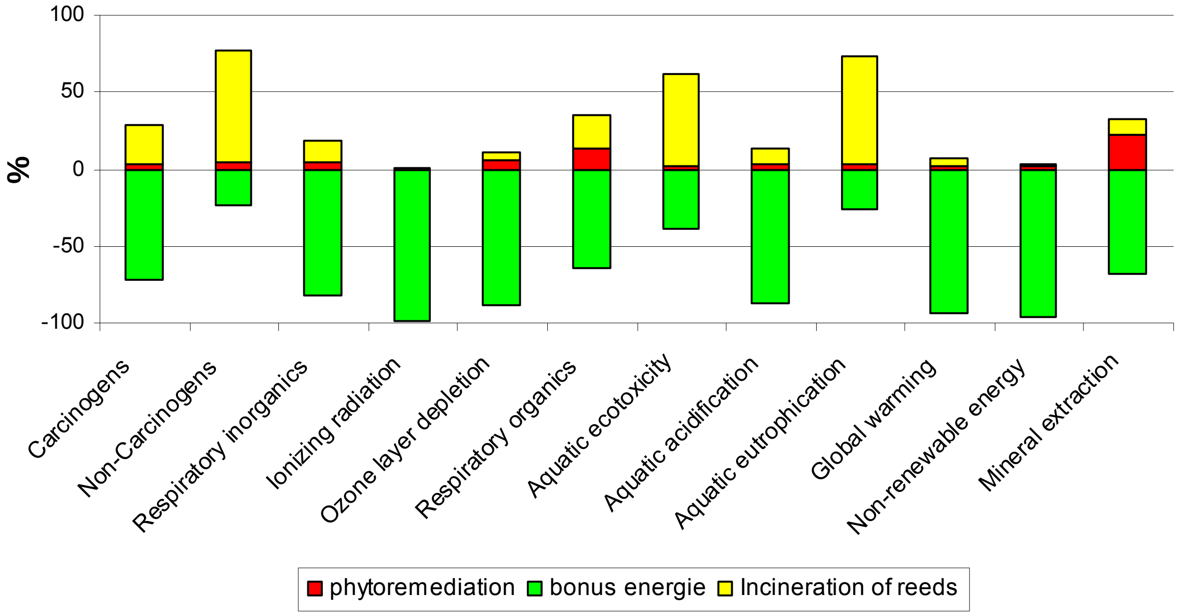

Figure 9: Midpoints impacts engendered by the phytoremediation application

Human Health

In the figure 9.5 it can be seen that non-carcinogenic substances have the greatest impact on human health (see table 9.11). Less important, but still relevant is the impact on human respiration due to inorganic compounds. Both impacts are generated by the incineration and the phytoextraction processes.

Table 23: Human Health midpoints results for the processes involved in the phytoremediation.

Impact category	Unit	Total	phyto24	bonus energy	Burning of reeds
Carcinogens	DALY	-3.1E-04	2.3E-05	-5.0E-04	1.8E-04
Non-Carcinogens	DALY	1.1E-03	1.1E-04	-5.0E-04	1.5E-03
Respiratory inorganics	DALY	-2.0E-02	1.3E-03	-2.5E-02	4.2E-03
Ionizing radiation	DALY	-1.0E-03	2.3E-06	-1.0E-03	7.6E-06
Ozone layer depletion	DALY	-2.0E-06	1.6E-07	-2.3E-06	1.3E-07
Respiratory organics	DALY	-5.3E-06	2.3E-06	-1.1E-05	3.9E-06

Analysing more in detail the non-carcinogenic substances related to the incineration of reeds, zinc appears to be the responsible for 45% of the midpoint impact, followed by arsenic with 25% and dioxins with 19%. The origin of zinc and arsenic is at 99% the disposal of the wood ash mixture to landfarming (*disposal, wood ash mixture, pure, 0% water, to landfarming/kg /CH* in the dataset), dioxins come at 94% from the combustion process.

Non-carcinogenic substances dominating in phytoextraction phase are the same as those responsible for its impact (Zn, As and dioxins). They are emitted by the tillage, the mulching and the swath processes.

In the phytoextraction and incineration phases the respiratory inorganics midpoint is instead dominated by nitrogen oxides and particulates with a diameter smaller than 2.5 µm. They come from the transport and the combustion of the plants.

Impact on Water

Ecotoxicity is the aquatic midpoint which presents the highest impact value. The main contribution to that is the disposal of the wood ash mixture (*Disposal, wood ash mixture, pure, 0% water, to landfarming/CH U*), where aluminium emitted in the soil is the substance responsible for 90% of the process impact.

Table 24 : Impact on water midpoints results for the processes involved in the phytoremediation

Impact category	Unit	Total	phyto24	bonus energy	Burning of reeds
Aquatic ecotoxicity	PDF*m^2*yr	76.1	6.1	-124.6	194.6
Aquatic acidification	PDF*m^2*yr	-13.6	0.49	-16.00	1.95
Aquatic eutrophication	PDF*m^2*yr	4.E-07	3.E-08	-2.E-07	6.E-07

For acidification the sum of the impacts expressed in PDF*m^2*yr is negative. This is due to the contribution of the co-generated energy that prevents producing energy by other techniques that have a greater effect on acidification of the aquatic environment. Despite the total impact having a negative value (net benefit), the incineration process has a large impact compared to the other process phases.

Summary and comments

The phytoremediation process is interesting especially due to the co-generation of energy, which diminishes the impact on the environment of the whole process. However, if no energetic exploitation was possible, this strategy would lose a lot of interest and its impact on the environment would significantly increase.

Major contribution to both human health and aquatic ecosystem impacts derive from the incineration and the ash disposal processes.

The assumption here is that the incineration of reeds is equal to a non-polluted biomass incineration. As they are growing on a zinc contaminated site, the content in the reeds is higher than what is normally found in average biomass and consequently the impact due to zinc emission is under-estimated.

2-8-3 Do nothing scenario

The years necessary for Demmerik polder to achieve target values have been calculated. Legally speaking (in the Dutch regulation context), once these values achieved, this would mean that no impact from the site is considered as influencing the ecosystem. From an ecological and ecotoxicological point of view, this assumption can be refused. For example chronic toxic effects could be found although the concentration of the substance is lower than any legal limit.

The time needed for Demmerik polder to achieve target values of metal concentrations are shown in table 9.13. As a fixed time scale, with fixed steps, was established the value was approximated to the higher time steps (the division is: 1,5,10,100,1000,10 000, 60 000 years). For instance, cadmium in Demmerik reaches the target value in less than 5 years. As it is between the two moments 1 year and 5 years, that were taken for the time scale, it is considered that Cadmium in the Demmerik soil will achieve the value in 5 years (excess approximation).

Table 25: years needed by Demmerik to achieve target values under natural conditions of metal emission. Values rounded up to the following time step.

Cadmium	5 years
Copper	10 000 years
Lead	60 000 years
Zinc	10 000 years

This evaluation will be needed after the calculation of the impact of the emission of metals during time, because once the legal concentration is achieved, the impact will be considered as zero (consequently, the PDF*m^2*yr value of the cumulative emission will be considered as the one found for the moments presented in table 9.13).

The analysis of the impact of the metal emissions is considered only in two endpoint categories out of four. The two neglected categories are climate change (quantified as kg of carbon dioxide emitted in the air) and resources (quantified in the needed mega joules (MJ) of primary energy). This is due to the fact that the “do nothing” option does not need any activity/process to be executed.

Impact on Human Health

The impact generated by the natural emission of the four considered metals from the soil on human health was derived following IMPACT2002+ method with Ecoinvent coefficients. The impact on human health is calculated as the sum of the damages in 6 characterisation categories: carcinogenic substances, non-carcinogenic substances, inorganic substances affecting respiration, organic substances affecting respiration, ionising radiation inducers and substances involved into ozone layer depletion (Humbert et al., 2007).

Following the Ecoinvent database, metals emitted in the water compartment generate an impact only as non-carcinogenic substances, except for cadmium, which is also considered to be carcinogenic. The metals are converted in the amount of vinyl chloride equivalent with the characterisation factors for carcinogenic and non-carcinogenic midpoints. After that the amount of vinyl chloride is converted with the damage factor to estimate the impact. The unit used to express the impact on human health is the disability adjusted life years (DALY), which represents the sum of the years of life lost due to premature mortality and disability. The characterisation and damage factors are listed in the table below.

Table 26: Characterisation and damage factors for the four metals emitted in water. Data from IMPACT2002 method.

	Characterisation factor [kg C2H3Cl / kg of metal]		Damage factor [DALY/ kg C2H3Cl]
	Carcinogens	non carcinogens	Carcinogen and non-carcinogen
Cadmium	6.96E-14	713.820727	2.80*10-6
Copper		4.12829302	
Lead		10.7434227	
Zinc		133.393433	

The impacts on human health were derived and are shown in table 9.15.

Table 27: Impact on human health calculated with IMPACT2002+ method for diffent time periods (cumulated values).

	Impact on human health expressed in DALY						
	1	5	10	100	1000	10000	60000
Cd	7.6E-07	3.8E-06					
Cu	2.5E-07	1.2E-06	2.5E-06	6.4E-05	6.5E-05	7.0E-05	
Pb	1.5E-07	7.6E-07	1.5E-06	1.5E-05	1.3E-04	6.2E-04	1.2E-03
Zn	5.5E-05	2.8E-04	5.5E-04	2.4E-03	2.6E-03	4.3E-03	

The number of DALY's found was taken for the metal concentration in the Dutch legislation considered to be non-contaminated. As the impact derived is calculated on the cumulative metal emission, it could be said that once the target values for metals are achieved (table 9.13), the impact will not increase any more. It means that the impacts of the following period are considered to be zero. It is important to see that this limit is strictly linked to the legal value for soil contamination in the Netherlands, and could be different if the study was applied to a different country.

Table 28: Final impact of the four metals on human health when the metal level reaches taget values.

Cadmium	At 5 years	$3.8*10^{-6}$ DALY
Cupper	At 10 000 years	$7*10^{-5}$ DALY
Lead	At 60 000 years	$1.2*10^{-3}$ DALY
Zinc	At 10 000 years	$4.3*10^{-3}$ DALY

The sum of the impact of the different metals gives the global impact on the aquatic ecosystem for the “do nothing” option

Impact on human health = $3.8*10^{-6} +7*10^{-5} + 1.2*10^{-3}+ 4.3*10^{-3} = 5.6*10^{-3}$ *[DALY]*

Impact on water

Calculating the impact using IMPACT2002 coefficients, PDF*m^2*yr value for the impact on aquatic ecotoxicity (as it is supposed that all the metals are transferred to water compartment) are derived for each metal, during 60 000 years. The characterisation factors given by IMPACT 2002 dataset are shown in table 9.17.

Table 29 : Characterisation factors for the Cd, Cu, Pb and Zn emitted into the water compartment (IMPACT 2002 dataset).

Compartment	Substance	CAS number	Factor	Unit
Water	Cadmium, ion	022537-48-0	2916028.29	kg TEG water / kg
Water	Copper, ion	017493-86-6	20550296	kg TEG water / kg
Water	Lead	007439-92-1	263702.723	kg TEG water / kg
Water	Zinc	007440-66-6	1402499.82	kg TEG water / kg

The damage factor is the same used in chapter 6.3, $5.02 * 10^{-5}$ PDF*m^2*yr/kg TEG water, and the results of the cumulative emission are shown in the following tables.

Table 30: Aquatic ecotoxicity derived by IMPACT2002+ method, expressed in triethylene glycol equivalent in water (kg TEG water/kg) . The amount of metal in water is converted into the amount of TEG equivalent.

Time [yr]	1	5	10	48	100	1000	10000	60000
Cd	1102	5509						
	1	5	10	21	100	1000	10000	60000
Cu	442474	2212368	4424736	9091579	114553900	115504384	125009217	
	1	5	10	100	712	1000	10000	60000
Pb	1328	6642	13284	132840	945613	1122625	5463539	10506806
	1	5	10	25	100	1000	10000	60000
Zn	207964	1039822	2079644	5261625	8908888	9577463	16263206	

Table 31: Impact on water derived with the IMPACT2002+ method. The amounts are expressed in the Potentially Disappeared Fraction per square meter and per year (PDF*m^2*yr).

Time [yr]	1	5	10	48	100	1000	10000	60000
Cd	0	0						
	1	5	10	21	100	1000	10000	60000
Cu	22	111	222	456	5751	5798	6275	
	1	5	10	100	712	1000	10000	60000
Pb	0	0	1	7	47	56	274	527
	1	5	10	25	100	1000	10000	60000
Zn	10	52	104	264	447	481	816	

Table32: Impact of the emitted metal on acquatic ecotoxicity. Values are rounded off.

Cadmium	At 5 years	0.28 PDF*m^2*yr
Cupper	At 10 000 years	6275 PDF*m^2*yr
Lead	At 60 000 years	527 PDF*m^2*yr
Zinc	At 10 000 years	816 PDF*m^2*yr

The sum of the impact of the different metals gives the global impact on water ecosystem for the “do nothing” option:

$$\text{Impact on water} = 0.28 + 6275 + 527 + 816\ [PDF*m^2*yr] = 7618.28\ [PDF*m^2*yr]$$

The results show the impact on aquatic ecotoxicity of cadmium, which is practically negligible compared to the other metals. In effect, this is the only element that is present at time zero on the site, with a value under the Dutch target value.

As lead does not reach target values but at the end,its impact was expected to be highest. The element which dominates the possible damaged fraction per square meter every year is copper, which presents an impact value higher than 6 000 PDF*m^2*yr.

A source of error in the interpretation is the impression that the results show that zinc, which after 10 000 has already reached the theoretically non-impacting level has in reality a greater impact than lead, which is considered to have no more impact only after 60 000 years!

In reality it is false. In fact, the impact depends on the amount of metal that has been leached out from the soil, and not the amount that is remaining. If the same zinc amount over the target value would have been leached out slower or faster, the impact calculated with this method would have been the same.

2-9 Comparing the Results of the Scenarios

2-9-1 Resulted potential damages

The previous chapter gives the results to compare the impacts of three approaches to the management of contaminated sites. Phytoremediation aims at decontamination using the natural properties of plants, excavation is the most concrete and rapid approach, while the “do nothing” option consists of the simple observation of a natural decrease of metal concentrations due to their emission toward another environmental compartment.

To be correct from a LCA point of view, the phytoremediation scenario should take into account also the emissions after the application of the remediation strategy (24 years). In fact, the impact of the fraction of metals that remains in the soil and that leaches during time should be calculated (until it reaches the target values). In this section, the impacts of copper and lead amounts above target values (the metals remaining after the 24-years phytoremediation) are derived with the same coefficients calculated with the same factors used for the “do nothing”.

The endpoints for the three scenarios are illustrated in figure 10.1 to 10.3. The numerical results are presented in the appendix III:

Impact on climate change and energy consumption

As the option “do nothing” does not need any product or service, its impact on energy consumption and climate change is nil as seen in figure (10.1). The excavation scenario presents the greater impact in both categories. The impact on climate change and energy resource consumption are positive for the excavation scenario and negative for the phytoremediation, thanks to the possibility to co-generate energy burning the plants.

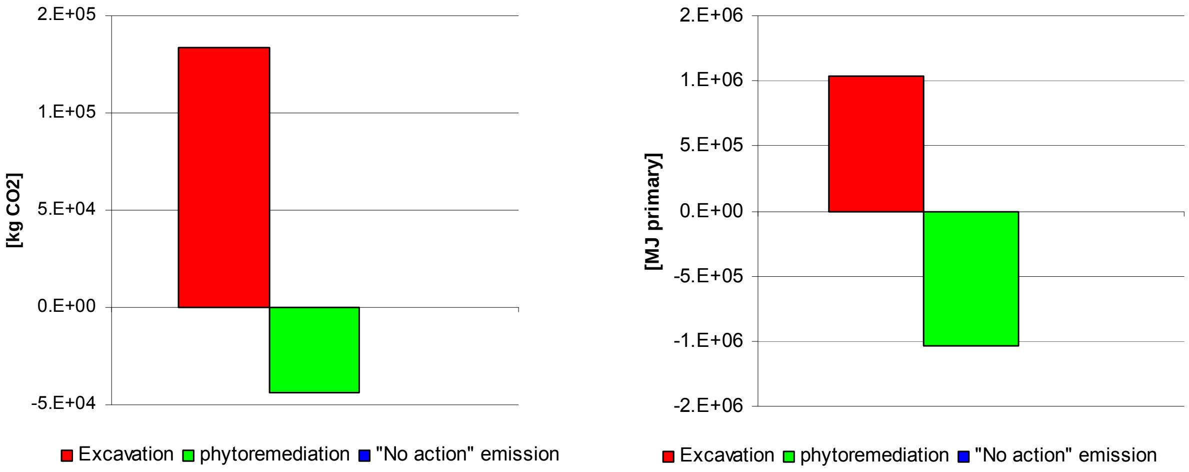

Figure 8: Impact on climate change (in kg CO_2 emitted) and primary energy consumption (mega joules) for the three scenarios.

In the excavation scenario it is the incineration that creates the carbon dioxide responsible for climate change. The major impact on the energy resources of this scenario is the crude oil production to obtain the light fuel oil burned in the waste disposal process. In the case of phytoremediation the most contributing substance is dinitrogen monoxide produced during the combustion, but it is largely compensated by the avoidance of the production of carbon dioxide thanks to the co-generation.

Human Health

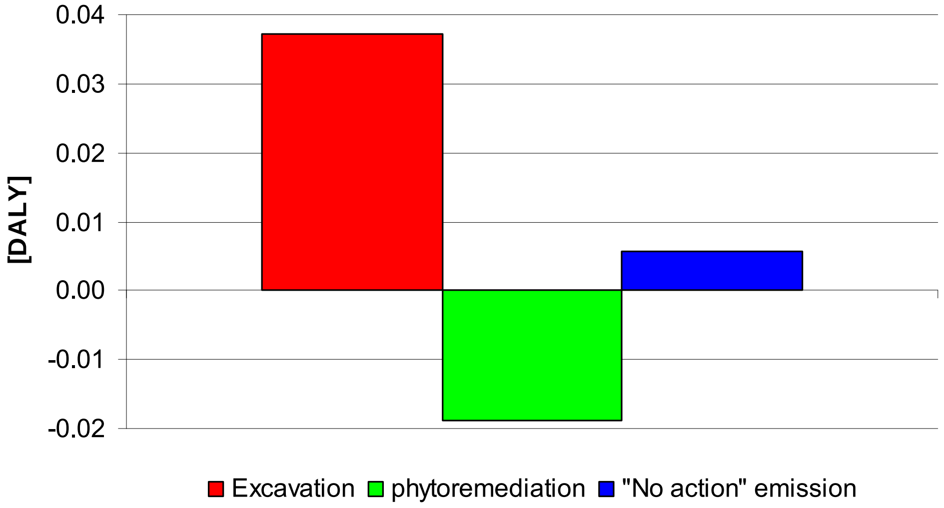

Figure 9: Impact on human health for the three scenarios. Unit DALY

The results for the comparison of impacts on the human health show that excavation has the greatest impact. Phytoremediation, thanks to the co-generation of energy during the plant incineration, has a negative value. The impact engendered by the “do nothing” scenario is less than 1% of the excavation. This latter is clearly the worst scenario for the impact on human health. These results show that the potential impact of the site management options can be responsible of the loss of maximum 0.037 years of life distributed over the overall population, compared to the “gain” 0.019 years of life of phytoremediation.

Impact on water

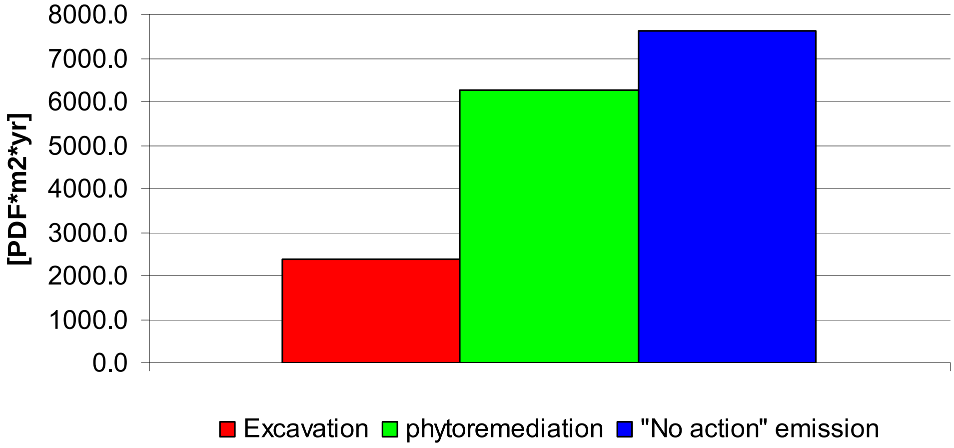

Figure 10: Impact on water for the three scenarios. Unit PDF*m^2*yr

In this endpoint the situation is different. The direct emission of metals in water generates the greatest impact on the potentially disappeared fraction of species per m^2 per year.

The 98% of the impact of phytoremediation is due to the emissions of copper and lead remaining in the field after 24 years, otherwise the impact is mainly due to the aluminium emitted into the soil coming from the disposal process. Excavation is, for this endpoint, the option which damages the water ecosystem the least.

2-9-2 Result Interpretation

The global results show that excavation is the scenario that generates the highest impact on the environment, except for the water compartment, where it generates less than half of the impact of the two other methods. The “do nothing” scenario is interesting from the point of view of the zero energy consumption or global warming contribution, but it is revealed to have a great impact on water ecotoxicity. Phytoremediation is interesting thanks to the possibility to co-generate energy, which compensates for its impact on the environment. Despite this, it presents the same problem as “do nothing” on the long term, as after the phytoremediation period the releasing of the metals in the water would have a very important impact on the aquatic ecotoxicity.

Since the results in the different damage categories do not present one clearly best option, no real conclusions about the most suitable site management scenario is possible. Two main parameters are at the base of the difficult interpretation: time and space.

The length of time needed to apply the different options is far from being comparable on a practical scale. Government and site owners should think in a long time scale for the management of their site, but thinking with a time horizon of 60 000 years make no sense, as there are too many uncertainties about the social and environmental situation in such a long time span. From a social perspective two questions can be formulated:

What is the length of time of a site management strategy? How long could it be assured by the authorities that the occupancy of a site would not change?

If time is a constraint for the choice of the remediation strategy, excavation would be the best option. From the environmental point of view, the main question is: what is the life span of the considered ecosystem? Can is be assumed that the ecosystem that undergoes the current pollution today, will be the same (i.e. be affected by the pollutant in the same way) in 1000 years? If the answer is yes, deriving the impacts on the ecosystem as it was done in this chapter is justified. If not, the results could be under- or overestimated.

Space reminds of the problem regarding the fate of the emissions. Particulate matter emitted in a zone with low population will not have the same impact on human health as if it was emitted in the centre of a city. The same could be said for the emissions in water. If the emissions are located in the upper area of a watershed, near the river outpouring in the ocean or in a lake, the impact would change. For instance, lakes have a key influence on the spatial sensitivity of the impact due to their large hydraulic retention time when compared to rivers.

Despite the problems due to the temporal and spatial resolution, the approach to estimate the impact of the “do nothing” option is justified from the life cycle assessment perspective, although it is difficult to estimate the fraction that will actually have an impact and the fraction that could be considered as “stored ecotoxocity” as the study done by Birgisdóttir presented in the part A, chapter 2.1.2 (Birgisdóttir et al., 2007).

2-10 Sensitivity Analysis of the LCA Scenarios

2-10-1 Atmospheric deposition

The influence of atmospheric deposition on the emission of metals from a soil would increase the time needed to lower metal concentrations until the target values.

To evaluate the influence of this parameter on the impact of metals, data for the Netherlands are considered based on the RIVM report (RIVM (a), 1996). The contaminated site treated in this study has a surface area of 0.1625 hectares, and the annual atmospheric deposition for the four metals is calculated and illustrated in table 11.1.

Table 33: Atmospheric deposition of metals in the Netherlands (RIVM (a), 1996).

Element	Deposition in g ha-1 yr-1	Deposition on Demmerik field in g yr-1
Cadmium	0-3	0- 0.49
Copper	0-100	0- 16.25
Lead	150 – 200	16.25 – 24.37
Zinc	0 - 200	0 – 32.5

The amount of metals leached out of the field due to their porewater concentration during the first years presents similar values. In fact, in the first phase of leaching (chapter 8.1.1), cadmium decreases with 0.4 g/yr, copper 21 g/yr, lead 5 g/yr and zinc 148 g/yr. The influence of the atmospheric deposition if occurring with maximal values during the fist phase of the metals leaching should thus be considered in the mass balance, especially for cadmium, copper and lead, where the emission and the deposition values are similar.

This fact has two main consequences: 1) the scenario of natural emission could be meaningless if the deposition occurs in the maximal values. No significant diminution of the global metal content in the soil would be found. 2) Lead could actually increase during the years due to the atmospheric deposition.

In both cases, more precise measurements should be effectuated as the range of values given is large, and the data could have significantly changed since 1996.

2-10-2 Willows instead of reeds

To test phytoremediation sensitivity on resulted impacts, a new case has been studied, using willows instead of reeds. Willows have been suggested has they are fast to propagate and achieve high annual biomass production. Several studies have demonstrated that willows have the capacity to accumulate elevated levels of cadmium and zinc. This capacity may be applied for removing metals from the topsoil (Meers et al., 2007).

French (French et al., 2006) in the United Kingdom made an estimation for off-take by willows and found that despite a relatively large proportion (20-45%) of lead in soil being extractable with EDTA, very little is available to plants.

Meers et al. (2007) analysed five willow species on three different soils, and in particular a heavily polluted sediment derived from surface soil. The annual biomass production is 5700 kg DW per hectare per year and the willows were planted with a density of 1000 plants per hectare. The accumulation rates are shown in table 11.2. In the article, concentrations for stems and leaves are given. Shoot accumulation of Cd, Cr, Cu, Ni, Pb and Zn was low and insufficient to consider for phytoextraction of these metals, as the derived time needed are too long.

Table 34: Estimation of time needed to achieve TV and zero value with phytoremediation with willow

	Cd	Cu	Pb	Zn
Metal concentration in sol [mg/kg DW]	1.09	165	743	318
Metal concentration in porewater [mg/l]	0.00093	0.053	0.0124	0.365
TV corrected for Demmerik soil [mg/kg DW]	1.59	57	120	185
Accumulation capacity for willow plant mg/kg	40.0	8.2	4.1	520.0
Amount of metal phytoextracted every year [mg/yr]	76000	15580	7790	988000
Initial soil metal amount (whole surface) [mg]	58451	8848125	39843375	17052750
Time to achieve TV [yr]	0	372	4289	7
Time to achieve zero concentration [yr]	1	568	5115	17

The time needed to achieve target value concentration for zinc is much lower than the one estimated using the reeds, but it is higher for copper and lead. The calorific value for willow wood is 19.7 MJ/kg (Ledin, 1996). Converting it to kWh/kg:

19.7 [MJ/kg] * 24 [yr] * 926.25[kg/yr] = 437931 MJ (= 121648 kWh)

Assuming that the yield of energy cogeneration is the same as for the reed case:

437931 MJ * 0.55 = 240862 MJ

The hypotheses used are the following:

The machinery use in the chopping of the maize is very similar to the one that can be used for young willows. The process of the willow phytoremediation is calculated over the same time period as the process with reed plants.

Table 35: reference flux for phytoremediation

Material/assemblies and Processes		
Chopping, maize CH/U	3.9	Ha
Transport, lorry 28t CH/U	1334	Tkm
Planting CH/U	0.1625	Ha
Wood chips, burned in cogen 6400kWth, emission control/MJ/CH	240862	MJ
Electricity, medium voltage, production UCTE, at grid/kWh/UCTE	-66906	kWh

Table 11.4 shows the results of the damage categories. Willow results are higher than the reed ones, with exception for the impact on the water ecosystems. In this category aluminium coming from the disposal of the ashes (see chapter 9.2.2) is the element dominating the impact for both scenarios, but as the biomass produced by willows in smaller, the impact coming from this process is also smaller. The order of the responsible substances/processes is the same for the two scenarios.

Table 36: Comparison of damage results for willow and reed scenarios

Damage category	Unit	Willow	Reed
Human Health	DALY	-0.01	-0.02
Impacts on Water	PDF*m^2*yr	54	63
Climate Change	kg CO2	-28535	-43970
Resources	MJ primary	-675546	-1031910

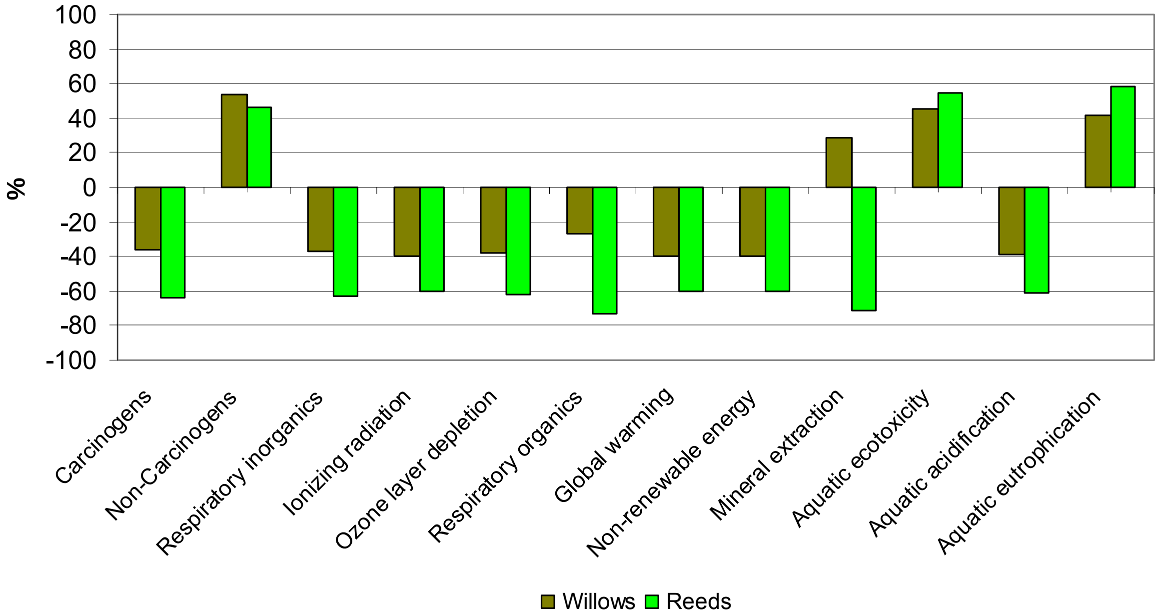

Figure 11: Comparison of the damages generates by the two phyoremediation scenarios.

The results are shown in the figure 11.1. Willow option seems to be more damaging (although it is still in the negative values) for human health, climate change and resource. The impact on water is in this case smaller than with the reed use.

It is interesting to notice the different results obtained for the mineral extraction category. Willow option presents a positive value while for reeds is negative. Looking deeper in the process inventory, it is found that for the willow scenario there is the need to use aluminium coming from bauxite[9] used in the production of the trailer of the chopping machine, while for the reed scenario this source is avoided (negative value of impact). The bigger value for willows is due to the co-generation of energy that lowers the total impact on the mineral extraction midpoint category with a more important value for reeds than for willows. The importance of the fraction of copper and lead for the impact is due to the relatively short time during which the phytoremediation is applied. The option to let the willows grow also after the period of extraction of zinc should be considered, as they still can have a beneficial effect, uptaking small quantities of Cu and Pb or reducing the amount of water seeping in the soil, thereby lowering the metal leaching phenomenon. In this way, the impact on the aquatic environment due to metal emissions can be retarded and no big maintenance is required.

2-11 Spatial Differentiation with IMPACT2002 EUROPEAN RESOLVED MODEL

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[9] Bauxite at mine/GLO U

It has been realised for more than a decade that the pattern of impacts predicted by site-generic LCIA is some cases deviates significantly from the actual pattern of impacts, and that this may have serious implications for the validity of the conclusions drawn from the LCA (Potting and Hauschild, 2006).

A spatially differentiated version of the IMPACT 2002 fate model has been developed for Western Europe, merging multimedia modelling with air and water transport modelling, accounting for 136 watersheds (figure 9.1) zones and 156 air cells (2 x 2.5 longitude/latitude degree air grid, about 200 x 250 km, figure 9.2) nested in a non-spatial global model (Pennington et al., 2005). A watershed zone includes air, soil, vegetation, water and sediment compartments. Oceanic zones match the air grid and consist of multiple sea layers. This model calculates population exposure at the appropriate level of spatial resolution as a function of chemical properties and landscape characteristics (Joillet et al., 2002).

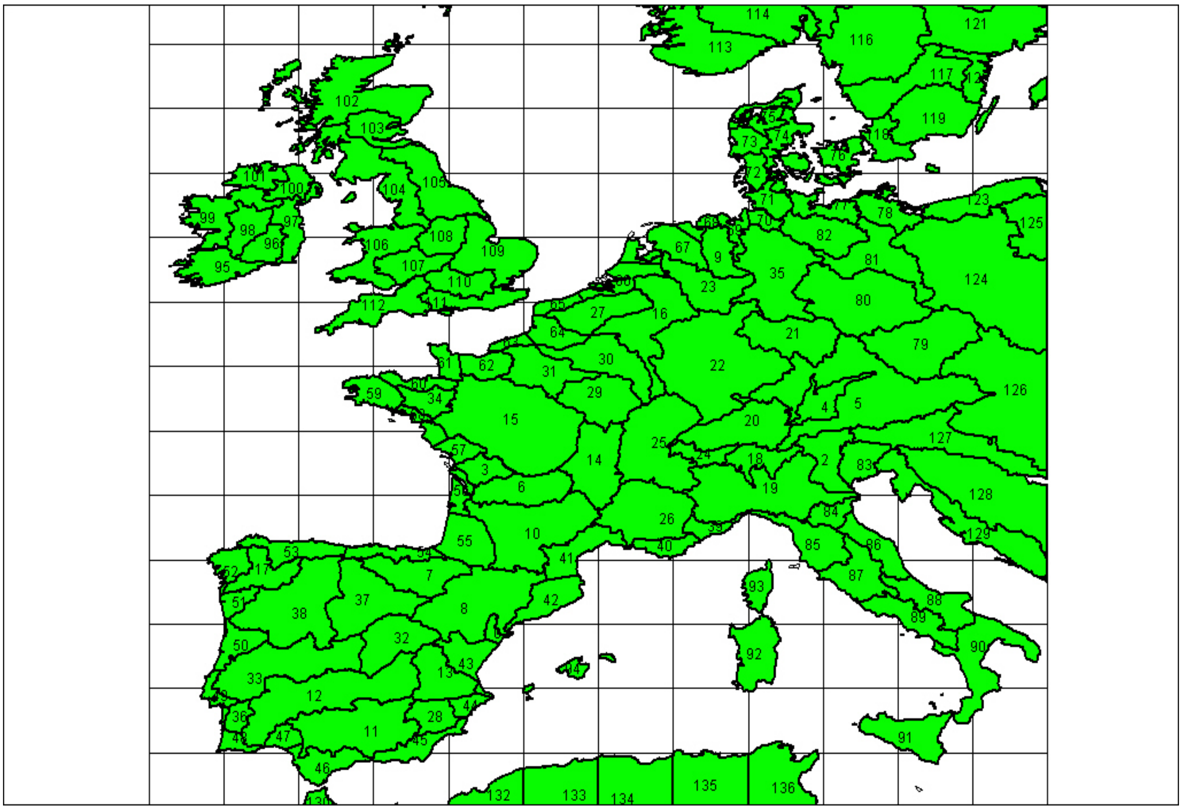

Figure 12: Watershed cells for spatial IMPAT 2002 european model (Joillet et al., 2003)

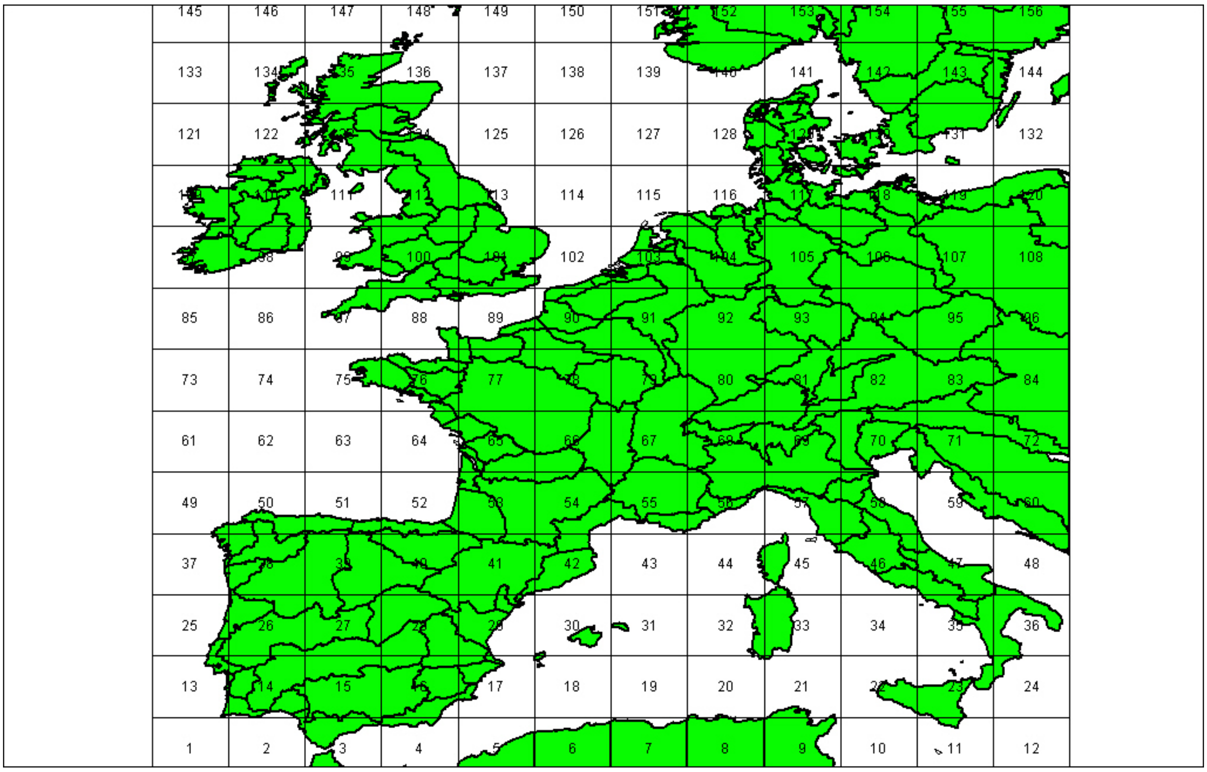

Figure 13: Air cells for spatial IMPAT 2002 european model (joillet et al., 2003)

Some of the watersheds had to be separated into smaller zones, as is the case of the Rhine watershed. In fact, since it has a surface area of more than 160 000 km^2, it was therefore necessary to select a smaller watershed resolution in this case (Pennington et al., 2005). The damage categories “Climate Change” and “Resources” are not illustrated, as they are not modelled with the regionalised version of IMPACT 2002, therefore the results are the same as the ones derived from the global model.

To model the Ronde Venen case study, the watershed W66 and the air cell A103 are considered. W66 encompasses the Dutch regions of Zeeland, South Holland, Utrecht, the western part of North Barbant and most Gelderland. A103 is wider and includes the whole North Barbant, North Holland, Flevoland, Gelderland but not the entire Zeeland.

Tables 12.1 and 12.2 report the results for human health and impact on water damages. They have been derived following the same reasoning as done for the global impact in chapter 9.3.3.

Cadmium	At 5 years	0.0047 PDF*m^2*yr
Cupper	At 10 000 years	10212 PDF*m^2*yr
Lead	At 60 000 years	558 PDF*m^2*yr
Zinc	At 10 000 years	541 PDF*m^2*yr

Table 2.1 Impact evolution on the Impact on Water damage category with the characterisation coefficients derived for the watershed cell W66.

A difference between IMPACT2002 (regional) and IMPACT2002 (aspatial, used in the previous chapters) is the unit used in the damage category “aquatic ecotoxicity”. In IMPACT2002 the new characterisation coefficients are given in PAF*m^3*yr per kilo of substance emitted. PAF means

Potentially Affected Fraction, and it is integrated over time (years) and space (cube meters of water). In SimaPRo and in this work, the characterisation factors are expressed in PDF (Potentially Disappeared Fraction) integrated over time and surface area. It can be noted that in IMPACT2002 PAF are based on EC_{50}, whereas in Eco-indicator 99 (another LCA dataset model) PAF are based on NOEC values (Humbert et al., 2007, Pennington et al., 2005). The relation between PAF and PDF is the following: PDF = PAF/2. This is because of a multiplication factor of 5 between NOEC and EC50 and a division factor of 10 between "Affected" and "Disappeared", providing a net division factor of 2 (thus PDF = (PAF/10)*5 = PAF/2) (Humbert et al., 2007). To pass from cubic meters of water to square meters the following relationship is valid: m^2 of water = m^3 of water / mean depth of fresh water (in the considered region). For the watershed 66 the average fresh water depth is 0.1 m[10].

Table 36: Impact evolution on the Human Health damage category. Derived with the characterisation coefficients for the watershed W66.

Cadmium	At 5 years	$2.1*10^{-6}$ DALY
Cupper	At 10 000 years	$2*10^{-4}$ DALY
Lead	At 60 000 years	$2.2*10^{-3}$ DALY
Zinc	At 10 000 years	$1.0*10^{-2}$ DALY

The damage results given by the two models (aspatial and regional) for the three scenarios are shown in figure 12.3 and 12.4. Regionalised impacts tend to be bigger than the ones calculated with the global model. An exception to this is the impact on water due to phytoremediation.

Human Health

For human health the observed increase of the impact is especially due to the high population density of the Netherlands, which increases the intake fraction of damaging substances. The scenario that presents the greatest change is the "do nothing" scenario, which more than doubles. The excavation scenario augments by more than 80% its impact on human health, while the increase for phytoremediation is around 10 % (its damage is still under zero). The difference is due mainly to the increase of the impact of non-carcinogenic substances (figure 12.4), which in the regionalised model is almost twice that in the global (aspatial) model.

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[10] In the European one box-model the continental average depth was considered to be 17.8 m (value of IMPACT2002). It has recently decided to update the value to 2.5 m (Payet, personnal communication)

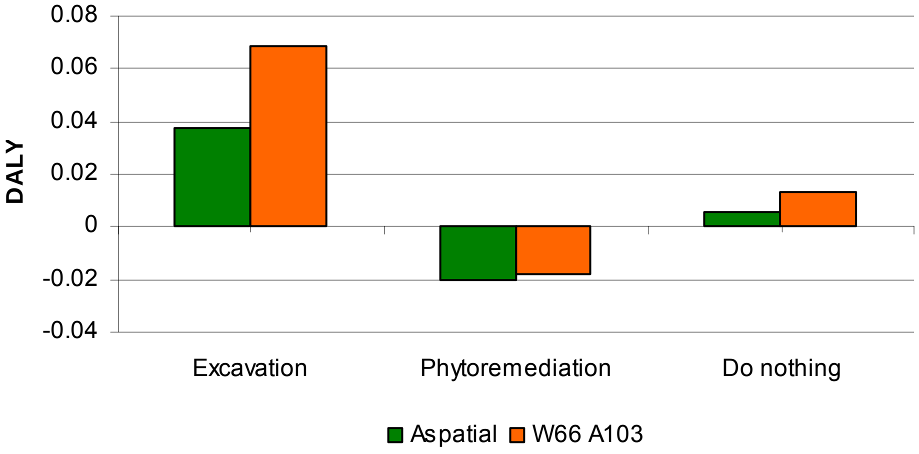

Figure 14: Comparison of impact on human health derived with the global aspatial model (Aspatial) and the regional model for Netherlands (W66 A103)

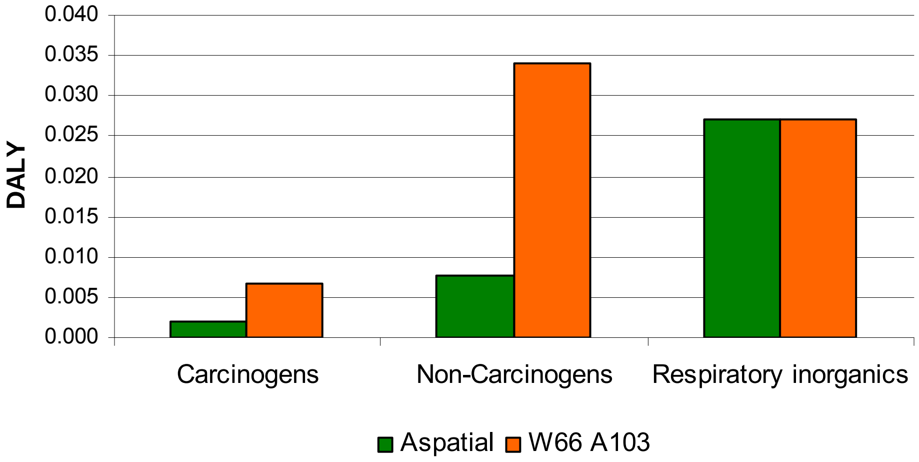

Figure 15: Comparison of the characterisation factors for the three midpoints contributing most to human health damage, derived with the global aspatial model (Aspatial) and the regional model for Netherlands (W66 A103)

With the regionalised approach the weight of the midpoints changes. Whereas the inorganic substances affecting respiration were the most important for the evaluation of the excavation scenario of the impact with the global model, the regionalisation approach says that the non-carcinogenic substances are supposed to bring the biggest contribution to the impact on human health.

As the impact on human respiration is due to substances (mostly nitrogen oxides and PM2.5) emitted in the air, the meteorological conditions of the Netherlands can confirm that no increment in this category should be found the region does not present conditions of still air. This situation could happen in some valleys or in regions with natural air circulation barriers, where the influence of the emission in the air of these substances would have a bigger impact on human health because of the increased exposition. It can be the case for the region between the Swiss Alps and the city of Milano, where the air pollution (especially particulate matter and ozone[11]) is a serious problem and the limit values of the swiss air quality standards are often exceeded.

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[11] Nitrogen oxides are, with volatile organic compound, a precursor of troposphere ozone formation (Van den Bergh, 2001)

On the other hand, the impact of carcinogens and non-carcinogens is strictly linked to the high population density, about eight times higher than the European average (data taken from IMPACT2002).

Impact on Water

The impact on water is illustrated with the figure 12.5. As already noticed with the impacts on human health, the divergence among the results of the two models is important. The damage due to the ecotoxicity on the water environment does not present the same trend for the three scenarios.

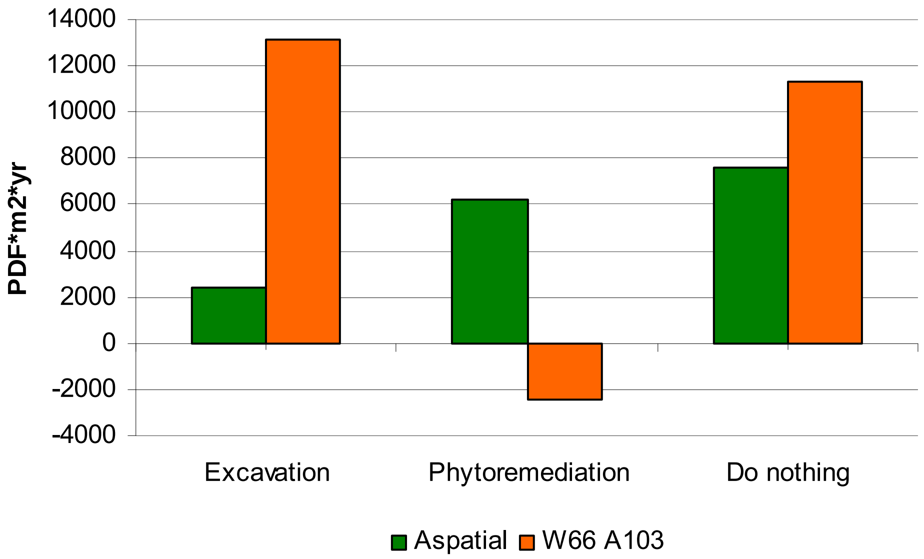

Figure 16: Comparison of impact on water derived with the global aspatial model (Aspatial) and the regional model for Netherlands (W66 A103). The aspatial value for phytoremediation is 62 PDF*m^2*yr, the value is too low to be seen n the figure.

The impact of excavation increases by more than 5.5 times, and the “do nothing” impact, which is generated only by four metals, is about 1.5 times bigger. This lead to the situation where the most impacting scenario has changed, from the “do nothing” with the aspatial model, to the excavation with the regionalised model. Compared to the global situation, aluminium emitted in the air has become in this regionalised analysis responsible for the impact on the aquatic environment, followed by copper and cobalt emitted in water, which were the most contributing substances in the previous analysis. In fact, the characterisation factor for aluminium in the air has increased by almost 400 times, while the characterisation factor for copper has doubled and cobalt has not changed. The process that is at the origin of the aluminium emission is blasting (Blasting RER/U). This latter is a phase considered in the modelling of the incineration and the following disposal of the contaminated soil.

The impact of the “do nothing” scenario increases with the increase of the characterisation factor of the metals illustrated in the table below.

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Table 37: Comparison between aspatial and regionalised characterisation factors (CF) and the ratio between the values.

Compartment	Substance	CF Aspatial	CF W66 A103	Unit	Ratio
Water	Cadmium	2916028	250271.8	kg TEG water / kg	0.09
Water	Copper	20550296	33441995	kg TEG water / kg	1.63
Water	Lead	263702.7	279031	kg TEG water / kg	1.06
Water	Zinc	1402500	1928671	kg TEG water / kg	1.38

It is interesting to notice that while copper, lead and zinc characterisation factors augment, for the watershed W66 the factor for cadmium decreases.

The impact of phytoremediation on the aquatic ecotoxicity decrease, on the contrary to the two other scenarios. In general, the phytoremediation has not decreased its impact, the different trend is due to the avoidance of use of the aluminium that would have been involved in the process of producing energy, if no co-generation were done.

2-11-1Localisation of the impacts

The advantage with the regional model IMPACT2002 is the possibility to investigate where the estimated impact would happen (if inside the considered cell or in others). With the impact on the water environment, this evaluation is relatively easy, especially because the watershed division has a hydrological meaning for the water and groundwater flow and practically does not change in time. In this case the impact on the water would depend essentially to the possibility of the emitted substance to reach the water.

For the impact on human health things are more complicated. The scheme illustrated in the figure 12.6 shows the information and data needed to evaluate the damage starting from the emission of the substance. The intake fractions (fraction of pollutant released into the environment that results in human exposure at a population level) have been calculated for about 1000 substances (Joillet et al., 2004).

If at least one of these factors varies due to the regionalisation, the characterisation factor used to derive the impacts on the human health or the water environment, would vary as well.

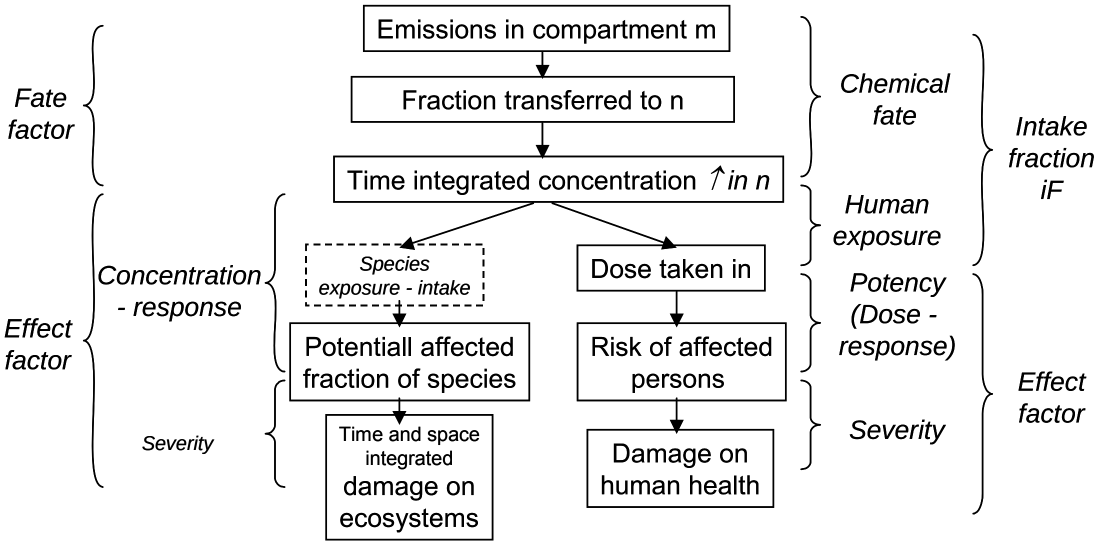

Figure 17 : Scheme of the model IMPACT2002 from the emission to the damage for aquatic ecotoxicy, carcinogen and non-carcinogens substances (from Pennington et al., 2005 and Joillet et al., 2003)

A parameter particularly difficult to insert in the model is the influence of the substances which are present at different trophic levels. In fact, the farming produces food that is most times exported to other regions or countries. In addition to this, the movement of people, travelling for professional reasons of for leisure, for a medium-long time period during the year could influence significantly the results. Because of this, no analysis of the distribution of the carcinogenic and non-carcinogenic substances emitted has been done.

The analysis for the localisation of the damages on aquatic ecotoxicity showed that only four substances are supposed to act on other watersheds: chlorothanol, dinoseb, linuron and metolachlor. The first is a fungicide, the following three are herbicides. They are emitted during the remediation processes but in very small amount, and contribute to less than 0.001% of the impact on any of the considered midpoint categories. Their presence is found in the watershed W67 (north of the Netherlands) and W68 (point in the north-east of Germany), but only for fractions smaller than 0.2 % of the original emitted mass.

In the later version of IMPACT2002 regionalised for European regions the modelling for the oceanic (open seas) compartment has not been done yet. Depending on the geographical situation of the region of interests, this can be very important in the estimation of the impact of emissions to water. This could be the case of the Netherlands, and the influence that the emissions coming from this country have on the North Sea.

2-11-2 Conclusions about the regionalised model

This field of LCA still has to be studied more in detail and the dataset has to be completed. Not all the substances present in IMPACT2002+ dataset have been inserted in the regionalised model. In addition to this, some local characteristics that would give more realism to the emission fate have not been edited yet or are provisionally only roughly estimated (i.e. adsorption coefficients for metals in soil or sediments).

The weight of the impact of non carcinogenic substances on human health increases significantly. In the excavation scenario, this augmentation leads to the change in the highest impact category for human health damage: in the global model inorganic compounds affecting respiration were the dominant category, and now, with the regionalised model, non carcinogenic substances have the biggest impact.

This conclusion is only partially true, as the regional model can derive new characterisation factors only for 2 categories of human health damage (carcinogens and non carcinogens) and aquatic ecotoxicity. What is clear is the tendency to obtain greater impacts (due to greater characterisation factors) when analysing the whole remediation process. The characterization factor is itself obtained by multiplying a fate factor (FF; describing the fraction of substance transferred from the compartment of emission to the compartment of reception, and its residence time in it) and an effect factor (EF) expressing the effect of the substance on organisms per concentration of exposure (see figure 12.6). As already mentioned before, the high population density and the shallow average water depth in the Netherlands are most probably responsible, with a greater exposure (and intake) of substances present in the environment compared to the rest of Europe.

Looking more in detail at the four metals that account for the pollution in the Ronde Venen site and how their CF's evolve, an increase is found for all metals.

Table 38 : Characterisation factors comparison Aspatial model and regional model for non-carcinogen and aquatic ecotoxicity categories, from IMPACT2002+. unsp: unspecified

Impact category	Non-Carcinogens	kg C2H3Cl	Aspatial Model	Regional W66 A103	Unit	Ratio
Air	Unsp.	Cadmium	548	2290	kg C2H3Cl / kg	**4.2**
Air	Unsp.	Copper	2	9	kg C2H3Cl / kg	**4.2**
Air	Unsp.	Lead	3	15	kg C2H3Cl / kg	**5.8**
Air	Unsp.	Zinc	93	433	kg C2H3Cl / kg	**4.7**
Soil	Unsp.	Cadmium	4408	13676	kg C2H3Cl / kg	3.1
Soil	Unsp.	Copper	16	51	kg C2H3Cl / kg	3.2
Soil	Unsp.	Lead	16	40	kg C2H3Cl / kg	2.5
Soil	Unsp.	Zinc	745	2724	kg C2H3Cl / kg	3.7
Soil	agricultural	Cadmium	17932	17932	kg C2H3Cl / kg	1.0
Soil	agricultural	Copper	61	61	kg C2H3Cl / kg	1.0
Soil	agricultural	Lead	35	35	kg C2H3Cl / kg	1.0
Soil	agricultural	Zinc	3068	3068	kg C2H3Cl / kg	1.0
Water	Unsp.	Cadmium, ion	714	1956	kg C2H3Cl / kg	2.7
Water	Unsp.	Copper, ion	4	12	kg C2H3Cl / kg	2.9

Water	Unsp.	Lead	11	20	kg C2H3Cl / kg	1.8
Water	Unsp.	Zinc	133	323	kg C2H3Cl / kg	2.4

Impact category	Aquatic ecotoxicity	kg TEG water	Aspatial	Regional W66 A103	Unit	Ratio
Air	Unsp.	Cadmium	427516	4926467	kg TEG water / kg	**11.5**
Air	Unsp.	Copper	2940562	553767799	kg TEG water / kg	**188.3**
Air	Unsp.	Lead	40064	7063234	kg TEG water / kg	**176.3**
Air	Unsp.	Zinc	203985	36742390	kg TEG water / kg	**180.1**
Soil	Unsp.	Cadmium	2913297	250058	kg TEG water / kg	0.1
Soil	Unsp.	Copper	20440752	33275685	kg TEG water / kg	1.6
Soil	Unsp.	Lead	263691	279021	kg TEG water / kg	1.1
Soil	Unsp.	Zinc	1395894	1920205	kg TEG water / kg	1.4
Water	Unsp.	Cadmium, ion	2916028	250272	kg TEG water / kg	0.1
Water	Unsp.	Copper, ion	20550296	33441995	kg TEG water / kg	1.6
Water	Unsp.	Lead	263703	279031	kg TEG water / kg	1.1
Water	Unsp.	Zinc	1402500	1928671	kg TEG water / kg	1.4
Water	Unsp.	Zinc, ion	1402500	1402500	kg TEG water / kg	1.0

The significant increase of the impact for Cd, Cu, Pb and Zn is found for the emissions in the air (table 12.4). The ratio varies up to 5.8 for the lead as a non-carcinogenic substance, but it takes a value of 188.3 for copper in the aquatic ecotoxicity.

The only element decreasing its toxicity in the regionalisation of the impact is the characterisation coefficient for the aquatic ecotoxicity of cadmium emitted in soil and in water, where the value is one tenth that of the aspatial model.

Despite these increasing of the characterisation factors of the four metals, excavation becomes more damaging that “do nothing” in water compartment. This is due to the high augmentation of the aluminium characterisation factor. This result has to be analysed more in detail, as only one substance play a significant role in the determination of the rank of the different options. This problem is discussed in chapter 14.3 where the uncertainties of the inventory data are treated.

2-12 Spatial and Temporal Differentiation of the Impacts

After having analysed the spatial influence on the potential impact generated by the different scenarios, it is useful to compare the two different spatial scales with the distribution of the impact during time.

The study performed by Birgisdóttir et al. (2007) (already discussed in chapter 2.2) considers a new impact category, Stored Ecotoxicity to water/soil for the analysis of the emission of metals from road material or landfills. This new category takes into account the presence of heavy metals and very persistent organic compound that might leach in the long term period. The author found that stored ecotoxicity to water is the most dominating environmental impact when impacts are assessed for more than 100 years, in both cases. It justifies the choice to try to evaluate the impact on water due to metal emission.

To illustrate the allocation of the impact in time, the concept of the stored additional impact (SAI) is introduced. SAI aims to visualise the fraction of the impact that is acting for 100 years since the beginning of the management strategy application, compare to the fraction that will accumulate only after this period.

For example, landfill sites can be considered as pollutant reservoirs that will release their content one day in the future. Neglecting the facilities, now-a-days they generate little damage on the environment, but once that the impermeable layer will be degraded, some elements could reach the water table. In this case, there is an impact, but is make no sense to average it over an infinite time period (or very long one).

Figures 13.1 and 13.2 illustrate the comparison of the two spatial scales over two different periods. Excavation SAI is zero, because the process is very fast (a few weeks depending on the site size). This is not exactly true, because the fraction remaining after the incineration of the polluted soil is supposed to be landfilled, and thus stored in the soil under a stabilised form, which would mean that also this scenario should have a SAI, even is small.

Human Health

Figure 13.1 shows the potential damage on the human health. The major impact, as already seen previously, is generated by the excavation. From this graph it appears clearly that the impact of the “do nothing” option. Half of it is present before 100 years, and the other half is added after 100 years. The impact of phytoremediation is still negative, but the contribution of the remaining metal emissions from the soil appears in the stored additional impact after 100 years.

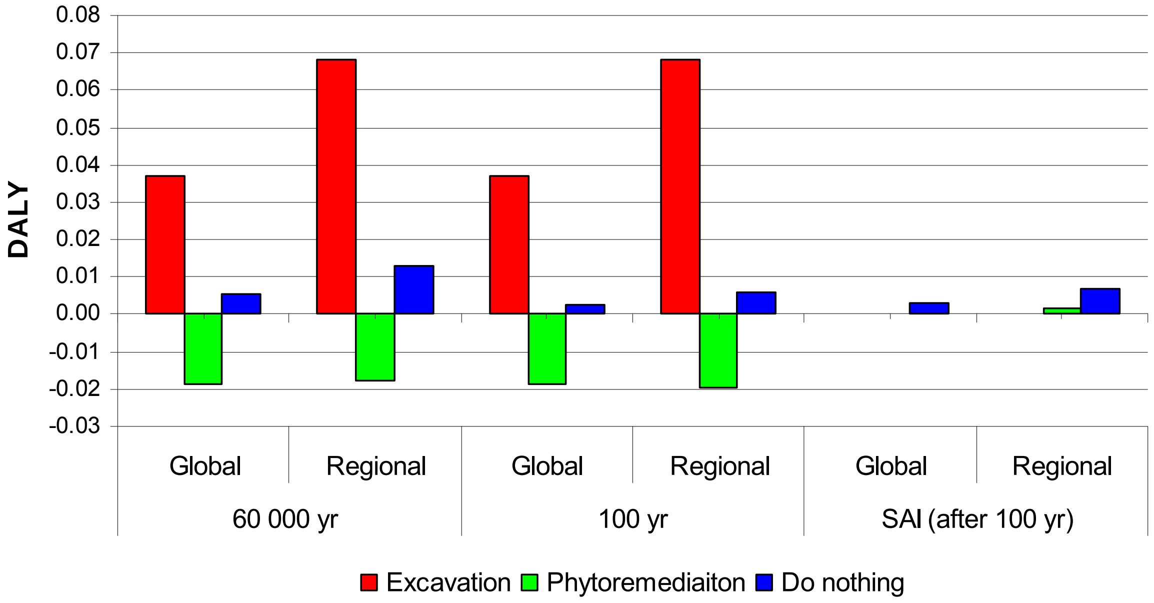

Figure 18: Impact on human health for the global and regional models, for a time horizon of 60 000 and 100 years. Stored additional impact (SAI) represents the fraction of the impact that will be cumulate after 100 years.

Impact on water

The analysis of the distribution of the responsibility over the three midpoint categories revealed that aquatic ecotoxicity gives the major contribution to the damage on water. In figure 13.2 the results of the regionalisation and the temporal sharing of the impact are shown. The information of this graph are more difficult to extrapolate, above all in the case of the phytoremediation, which changes the sign of its impact depending on the model (aspatial and regionalised) that is used.

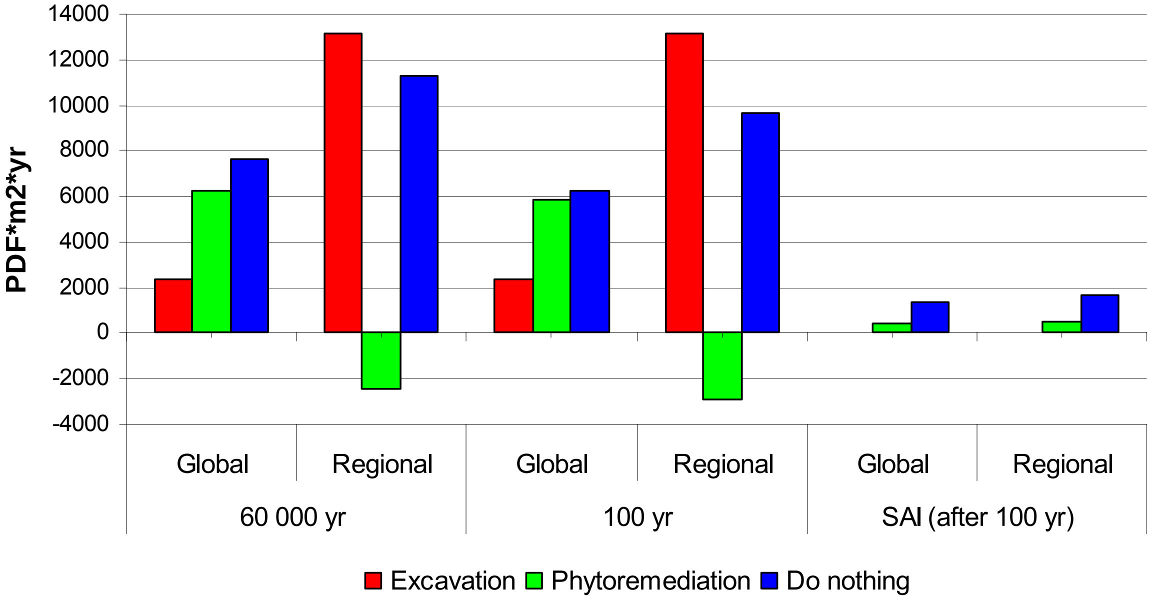

Figure 19: Impact on aquatic ecotoxicity for the global and regional models, for a time horizon of 60 000 and 100 years. Stored additional impact (SAI) represents the fraction of the impact that will be cumulate after 100 years.

The concept of stored additional impact should be studied more in detail in order to be used as communication tool of LCA studies. It could be crucial information for authorities dealing with contaminated sites that have to undergo remediation. In fact, most national regulations impose an intervention value based on metal concentrations in the present in the soil, giving a delay of a few years to start the remediation. A prioritization of the actions has to be effectuated in order to organise the works and focus on the most urgent cases.

This analyse is in addition justified on one hand by the amount of work and the costs that are behind the remediation proceeding and on the other hand by the need to know if a remediation action is actually necessary and beneficial for the ecosystem.

The SAI depends strictly on how the evaluation of the pollutants leaching is defined. As discussed in the end of the chapter 9.3.3, the leaching rate does not influence the impact calculated with this method. In fact, the derivation is based on the amount of metal that is leached, not on the time needed. SAI can thus bring the temporal dimension, sharing the damages occurring in the short term and long term. Landscape management projects are not normally planned with more than 100 years time horizon, but this fact does not mean that the SAI can be neglected, especially in the frame of sustainable projects, were the future impacts generated by actions undertaken today have to be taken into account and weighted in the decision-making procedure.

2-13 Uncertainty Analysis

2-13-1 Model uncertainties: space

The regional and the aspatial models express the same partition coefficient values for each metal for the partition of the metal in suspended soil/water and in the sediment water. In the followings table the data taken from the literature (U.S. EPA 1999 and RIVM 1997) have been substituted in the regionalised IMPACT2002 model, in order to study the influence of this single parameter on the damage on the environment, expressed in PAF*year*m^3 (potentially affected fraction over one year in one cubic meter) per kg of TEG equivalent. The relationship to derive the PDF*m^{2}*year is the following: PDF = PAF/2. This is because of a multiplication factor 5 between NOEC and EC$_{50}$ on which these units are based, and a division factor 10 between “Affected” and “Damage”, providing a net division factor of 2 (Humbert et al., 2007).

Table 39: sediment solids / water partition coefficient (l-water/kg soil) considered for the sensitivity analysis of IMPACT2002 regional model

Metal	IMPACT2002	US EPA (1999) minimal values	US EPA (1999) Maximal values	RIVM (1997)
Cadmium	85114	3	19952623	85114
Copper	33884	5	1584893	33884
Lead	430000	100	10000000	426580
Zinc	72444	32	11584893	72444

Table 14.1 illustrates the values coming from different databases that were used to analyse the sensitivity of the IMPACT2002 regionalised model on the sediment/water partition coefficient on the calculation of the damage to the water environment. It is seen that for the four metals analysed, the result obtained are practically the same, although the sediment/water partition coefficient can vary up to four orders of magnitude.

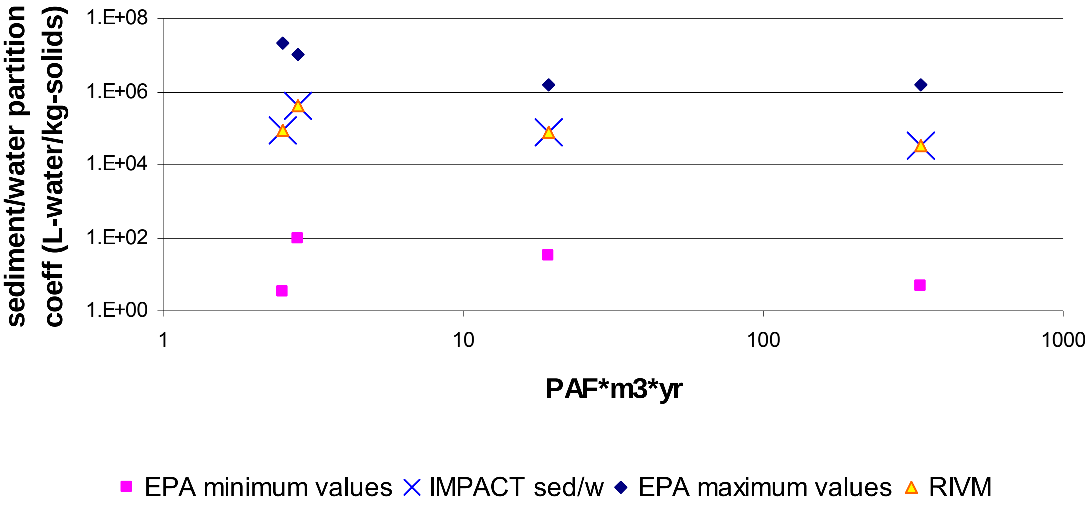

Figure 20: variation of the sediment/water partition coefficient to test the sensibility of the derivation of the impact on water. Every colour corresponds to a different dataset.

The same analysis was effectuated on the variation of the suspended solids – water partition coefficient. Data are present in lower number in the literature compare to the sediment/water partition coefficient, but the U.S.EPA gives us a range of data also for this value. The results are shown in figure 14.2. Is to be noticed that in a study about the derivation of normative values for the Netherlands Crommentuijn (Crommentuijn et al., 2000) selected suspended soil/water partition coefficient equivalent to the values of the IMPACT2002 regional model.

Table 40: Suspended soil / water partition coefficient (l-water/kg soil) considered for the sensitivity analysis of IMPACT2002 regional model

Metal	IMPACT2002	US EPA (1999) minimal values	US EPA (1999) Maximal values
Cadmium	128825	631	1995262
Copper	50118	1259	1258925
Lead	650000	2512	3162278
Zinc	109648	3162	7943282

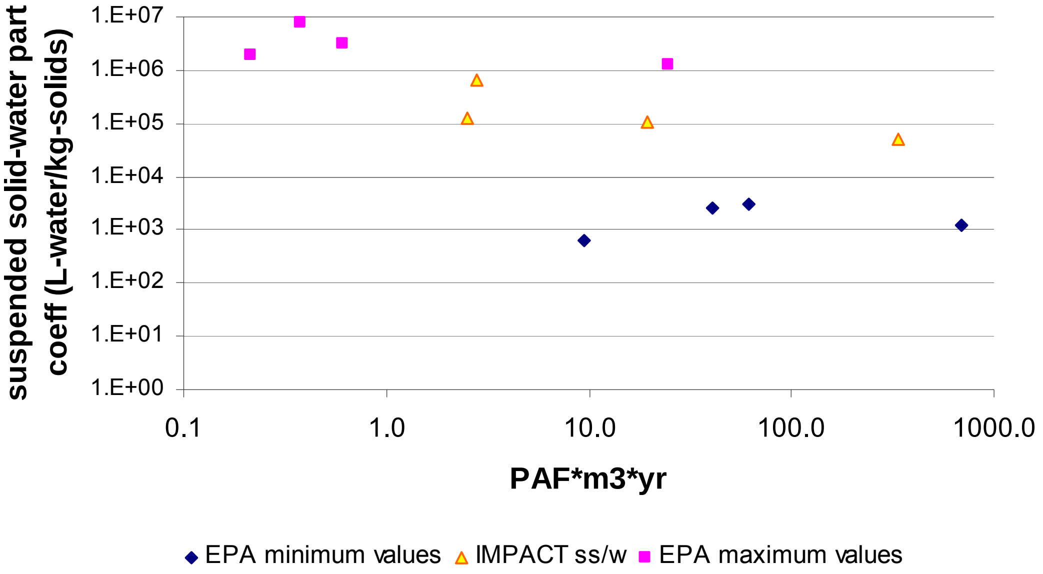

Figure 21: variation of the suspended soil/water partition coefficient to test the sensibility of the derivation of the impact on water. Every colour corresponds to a different dataset.

The results of the variation of the suspended soil/water partition coefficient can go up to two orders of magnitude on the resulted PAF*m^3*yr score. The metal with the most impact is copper, for the three groups of data. Cadmium and lead have very similar results, while the values for lead vary greatly.

2-13-2 Model uncertainties: time

To analyse the sensitivity of the time period considered for the impacts on the environment it would be justified to consider the "long-term" characterisation factors of IMPACT2002+. Treating the impacts on water it would mean that all the emissions going to the ground water or to the lakes, should be transferred to groundwater long-term or to the ocean compartment.

The state-of-art of the method does not allow this, because even the groundwater emissions don't have their characterisation factors, nor does the emission toward the ocean. Hellweg (Hellweg et al., 2001) mentions the gap of information about emissions to deep soil layers and groundwater in the LCA. In this moment, the part of impact due to landfilled waste (coming for instance from the incineration process) for long-term periods (over 100 years) is not taken into account. The emissions from the landfill for long-term period exists in the Ecoinvent dataset (chapter 2.4), but the characterisation coefficients in IMPACT2002 are still missing. This leads to an underestimation of the impact of the scenarios entailing actions with long-term emissions.

2-13-3 Inventory uncertainties

Blasting turns out to be the process that contributes up to 90% of the presence of aluminium, which is the substance responsible for most of the impact on the aquatic environment in the regionalised model.

Blasting in the dataset appears as Blasting (RER/U under the category Processing/Others). In the appendix IV the contribution for several processes, represented in the network of the excavation scenario, are represented.

For phytoremediation, aluminium is also the substance responsible for the impact on the aquatic environment. It appears in the three phases of the phytoremediation: 1) in the phase of plant maintenance[12] as aluminium emitted in the soil, 2) in the co-generation[13] phase as aluminium emitted in the air, and 3) in the burning phase emitted in the soil[14]. Looking further to see in which action of the processes the aluminium plays such an important role, it appears that although the disposal process contributes to the impact, some emissions are coming from the blasting action. Its contribution is found for the other phases of the phytoremediation scenario, as well. Ecoinvent process dataset, used by the IMPACT2002 method to derive the impacts, presents the following remark about the blasting: "Only the information on the raw material is handed over in written form. Sampling procedure: *Emissions are based on stochiometric calculations and all other In- and Output modules are estimations. This module should not be used if its relative importance would be high in a certain environmental inventory.* "(Ecoinvent dataset, comment by process authors).

Blasting is defined by the use of 100% of the explosive Tovex[15], which also presents a great uncertainty regarding environmental impacts. It is explicitly recommended in the comments on the Ecoinvent dataset not to consider the blasting if its contribution to the damage is too important, due to the low certainty of its impact on the environment. The scenario calculations were consequently executed again, subtracting the contribution of the blasting process.

It contributes in the following categories: non carcinogens, inorganics affecting respiration, organics affecting respiration, aquatic ecotoxicity, aquatic acidification and climate change. The new results, compared to the previously obtained, are shown in the figures 14.3 and 14.4. The changes are small for the impact on human health, but for water environments the situation is completely different.

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[12] In the process *disposal, drilling waste, 71.5% water, to landfarming/kg/CH*

[13] In the process *Blasting RER/U*

[14] In the process *disposal, wood ash mixture, pure, 0% water, to landfarming/kg/CH*

[15] Tovex is a high performance explosive which is replacing dynamite all over the world because it is safer to manufacture, transport, store and use.

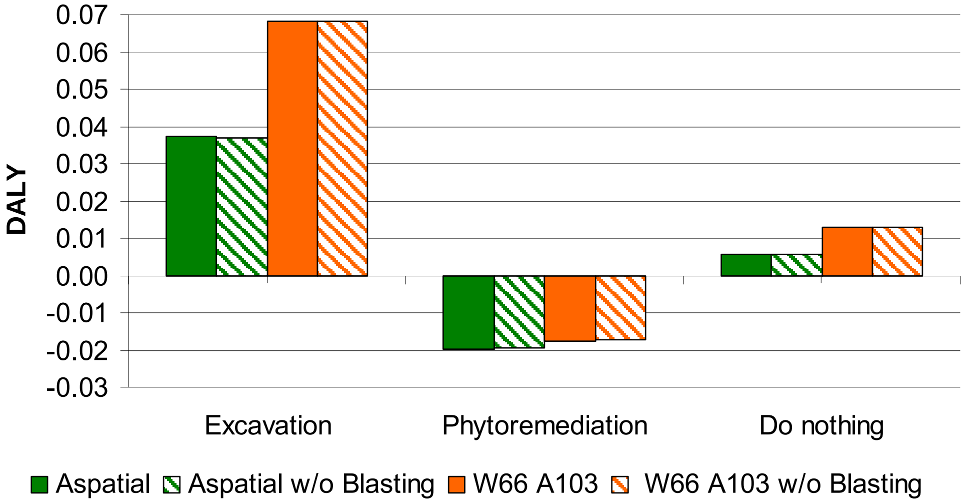

Figure 22: Impact on the human health of global and regional scale, with and without Blasting process contribution.

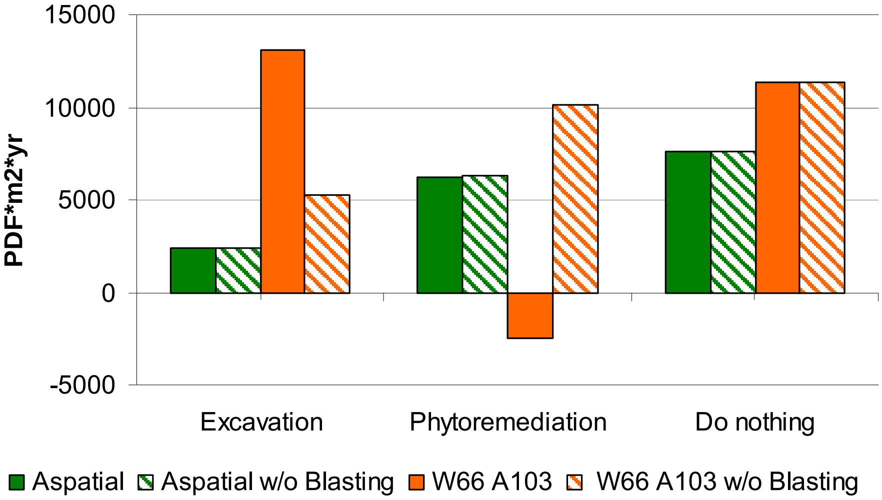

Figure 23: Impact on the water environment of global and regional scale, with and without Blasting process contribution.

Excavation diminishes its impact by more than a half, and phytoremediation increases in a very important way. It confirms that the avoidance of emission of aluminium due to the avoided blasting counted for a very large part in the value lower than zero.

The current impacts on the water for the different options have a different origin now. Copper and cobalt in water and air are the substances responsible for the impact of the excavation scenario, as in the global model. In the phytoremediation scenario the important contribution of the avoided blasting has disappeared, increasing importantly the impact of the whole scenario. As the process is not involved in the “do nothing” there is no variation between the results.

Globally, excluding the blasting process, the impact on the environment is underestimated, but its allocation on the different processes and substances is more representative of the reality. The conclusions deriving from the elaborated results can be different from the conclusions of the previous chapters. Particular attention has to be paid while using this data: as the blasting process contributed in different processes that constitute the scenarios, the damages found for the three remediation options can not be compared. In fact, looking at the figure 14.4, the situation shows that phytoremediation has a bigger impact than excavation for the regional model without blasting, but it is still smaller than the “do nothing” scenario damage.

It has to be noticed that the process was not contributing to the scenarios with the same quantity. It is thus correct to look at the responsible substance generating the impact within the scenario, but not compare the endpoints of the three options between them.

2-13-4 Final site quality: how to evaluate differences?

The three scenarios analysed with the LCA in this study, show different techniques that require diverse time lengths. The state of the site that undergoes the remediation is also strictly linked to the applied option.

A contaminated site that gets cleaned by remediation has theoretically a higher ecological value, and an increased exploitation potential (as normally contaminated sites are limited in use). It could be assumed that a remediated site can not be worth less than the same site with contamination.

This augmentation in the value (ecological, economical and social) due to the site management should be taken into account in the decision-making process. For example, if the economic potential of the site after the remediation is important, the motivation of people for applying a slower or more expensive approach would be greater.

Although it is difficult to quantify the positive impact of the remediation, a qualitative evaluation should be introduced and added to the description of the options available for the site management. It could be simply a three-level scale:

- 0 if the state of the site would substantially not change (except for the pollution management). This could be the example of the application of only monitoring.

- 1 if the improvement of the site is significant, but its ecosystem is still undergoing the consequences of the pollution (ecological value) or utilisation is still restricted.

- 2 if the remediated site can be allocated to any type of activity or it can develop a healthy ecosystem.

This evaluation scale could even be separated in the three categories of improvement: social, economical and ecological. For the first two the evaluation can be done by site/landscape managers, whom aim should be the sustainable integration of the “new” site in the context of the region. For the ecological value, a more scientific evaluation should be applied, always thinking also to the interactions between the site and the landscape. For instance, a very small area isolated in an urban landscape could never develop a wide space far from the anthropogenic pressure. In addition the remaining contaminants and their influence on the ecosystem could be assessment by the ecological risk assessment, which provides useful information that allows predicting the possible development the ecosystem and its ecological value.

Some peculiar cases of former contaminated sites could serve as examples: the Corwall County Council claims the ecological biodiversity value of metalliferous mining sites, which were abandoned in the past and show nowadays a flora adapted to the high metal concentrations, which gives a surplus value to the touristic value of the historic site. This is a combination of the three interests quoted before in a site that would be categorised as 1 in the evaluation scale of the previous paragraph. If landfills are considered as contaminated sites, the golf fields built once the covering and isolation was done, is also an example of economical improvement of the site.

2-14 Conclusions

This study attempted to integrate LCA as a tool within the contaminated site management framework. The method presented in the part A to assess the impact of the soil contaminated by heavy metals allowed a comparison between the “do nothing” scenario and two site management options, namely excavation and phytoremediation. The allocation of a part of the potential impact for a second period, and the regional model IMPACT2002 brought new elements to the analyses of the results derived by the “classical” IMPACT2002 global model.

Excavation showed the greater impact on human health, but the least for the water environment. It would be an efficient method to prevent the degradation of the environment but it has to be reminded that no possible long term emission was calculated for this scenario due to the lack in the modelling of the characteristics coefficients describing it (groundwater, groundwater long-term, ocean). Its impact on water could thus be underestimated.

On one hand, phytoremediation is very interesting due to the co-generation of energy, which allows avoiding polluting emissions. On the other hand it is not an efficient remediation strategy. The time needed is very long, it would be a long-term project for site management. If the decision-makers' wish is to designate the site as a natural area, the phytoremediation option could be a solution. In that case, phytoremediation could be separated into two periods: the first applying phytoextraction in order to decrease the amount of metals in the soil and co-generate energy, and the second may be phytostabilisation. Several studies showed the possibility to use grasses to phytostabilise metals (for instance *Agrostis tenuis* for copper wastes and *Agrosas tenuis* for acid lead and zinc (US EPA (b), 1997)).

In any case, the scenario needs to be adapted to the context of the country. Cogenerating energy in a plant that is already present in the territory of the region is very interesting, but it would not be the case if the cogeneration plant had to be built on purpose to allow energy production.

The "do nothing" scenario has a smaller impact on the human health compared to excavation, but the aquatic ecosystem undergoes a great pollution pressure. De Vries (De Vries et al., 2008) claims that the accumulation of heavy metals in Dutch soil at present primarily affects the quality of surface waters. In addition to this, the critical limits for copper and zinc in surface water are exceeded throughout the country. This option, put in the Dutch context, would thus not be advisable.

The regionalisation process for carcinogenic, non-carcinogenic and aquatic ecotoxicity characterisation factors leads to an augmentation of the impacts and to a change in the order of contribution to the damage, as seen for inorganic compounds affecting respiration in the excavation scenario. This result stresses the importance of the consideration of the local situation in the comparison of the impacts that can vary significantly depending on the emission watershed. For example, in the case of aquatic impact the depth and the presence of lakes in the region may change importantly the residence time of pollutants, and consequently their movement in the watershed. In the case of the Netherlands, the influence due to the high population density is a key factor for the impacts on human health.

The regional model has not been completed for all impact categories yet, and the results derived with a more completed version could vary due to the contribution of the other midpoints.

Temporal allocation of the damage with the introduction of the Stored Additional Impact (SAI) is done with the only aim of giving a time perception to the stakeholders that might use the information provided by the LCA. The setting up of management strategies involves also the planning in time of different actions, not only as practical work, but also as financial support. Detailing the time dimension gives reality to the information and makes it useful in the case of difficult site prioritisation. In fact, the

Netherlands presents a situation of diffuse pollution, where the financial investments in remediation are important due to the large number of sites that need further pollution investigation or remediation. The EEA (EEA (b), 2007) estimates the annual expenditures for contaminated site management in the Netherland to be about 0.5 % of the Gross Domestic Product (GDP), which represents about 15 euro per capita per year.

If the aim is to consider the impact of this scenario at the same moment in time (e.g. what will the level of pollution be in 30 years) the scenarios will give very different situations. A lower level soil in the case of excavation, a former reed field with developing vegetation and a basically unchanged grassland field are all possible conditions of the site in a few decades. It is thus important that stakeholders using the LCA information already have clear ideas about what is wished for the site, inserted in the landscape and regional context. The natural or leisure value for people of the surrounding has to be considered in addition to the exploitation potential. The development of a willow forest near urban areas might be more valuable for the quality of the environment than a soil exploited for agriculture. The decision makers have to know (or decide) if they need to clean the contaminated site drastically in a short time period or if they can assure a long term maintenance.

The decision depends basically on the target to protect: does human health in this context take priority over the degradation of the environment? To take on this subject it is needed to evaluate if this source of impact contributes significantly to the worsening of the current situation. For instance, if the damage caused by the excavation on human health is negligible compared to the impact on the transport traffic in the region, but it could improve notably the quality of the surface water, the preference could be given to this option. Once again, the importance of the context is essential for the good development of the project and the achievement of the goals.

In the Dutch current situation the long term thinking of the water issue leads to the problem of the rising sea level and the consequent increase of infiltration of salty water. The quality of surface water becomes more and more important for agricultural use (not directly for the drinking water supplying, as the drinking water in the Netherlands is taken by well from the groundwater). Water plays a fundamental role in the Dutch society, and it is given particular attention in the decision making process.

LCA gives comparable results for the impact on the global ecosystems, but no site- specific information. It would be an error not to consider the impact on the area under management. With the methodology developed in this study it is not possible to evaluate the impact of the pollution on the site itself. The possible development of the ecosystem depends on the pollution level and consequently the quality of the environment assessed with the ecological risk assessment has to taken into account.

As already mentioned, the evaluation of the final quality of the ecosystem should be integrated in the analysis. In addition to the “exploitation potential” described in the chapter 14.4, the ecotoxicological assessment should be included.

LCA is a tool that can be applied to the issue of contaminated site management, and can bring additional information to stakeholders involved in the decision making. The Appeldorn declaration (2004) declares that: “even though LCIA can use the models and the methodologies developed for Risk Assessment, LCA is a comparative analysis designed to compare different products and systems and not to predict the maximal risks associated with single substances”. In addition to this it is said that the oceans are deficient in essential metals, therefore additional inputs to the ocean will probably not lead to toxic effects. A sensitivity analysis is advised that excludes the metals from the impact assessment if the contribution analysis of the LCIA shows that metals have a dominant influence on the results and conclusions (Appeldorn Declaration, 2004).

No global agreement is present nowadays on the issue of the impacts of metal in the environment. In this study metals constitute the pollution source contributing to the majority of the impact; it is therefore obvious that not considering the role of metals in this context would be meaningless. This problem has been faced also by Haye, who claims that metals emitted during the life cycle of different products and substances may particularly generate damages on the ecosystems, especially the soil; however, their specific behaviour and impacts on soil organisms have not been adequately considered within the LCA process so far. In fact, in the current situation, effect factors of metal in the soils are derived from the effect factors on aquatic ecosystems for the same substances using the coefficient of partitioning between soil and porewater at equilibrium (Haye et al., 2007).

LCA can be applied to contaminated site management studies, although the results provided by this tool are comparative values and not absolute.

The “do nothing” scenario has to be considered while analysing several strategies. It serves as a reference and can lead to deductions about the real efficiency and utility of the other remediation processes. The approach for the evaluation of its impact presented in the part A of this report is valid to assess the surface soil layer as a pollutant reservoir, but it does not allow the evaluation of the pollutants on the site itself. To do this, a complementary approach that includes perspectives such as those used in ERA is advised.

The spatial and the temporal dimensions are analysed via the comparison between the global and the regional IMPACT2002 model and the introduction of the stored additional impact (SAI). Although the LCA does not give absolute results, the tendency of all impacts values to increase suggests that, compared to the average situation in Europe, the considered processes will have a greater impact. This is especially due to the increased exposure and uptake due to high population density. The temporal allocation helps the visualisation of the repartition of the impact in short-term (before 100 years) and long-term (after 100 years), in order to give a sense of priority if is seen that most impacts are occurring in the first 100 years.

The question of the evolution of the receptor ecosystem is still open. In fact, if long-term emissions continue the provoked damage on the evolved ecosystem, the impact could be different from what is was predicted.

2-15 Perspectives

This report has the aim to approach the problem of contaminated site with LCA as a decision-making tool. Some points have been discussed during its application to the real case of the Ronde Venen area.

The evaluation of the emissions of metal from the soil should be studied more in detail in order to build a general method that could be integrated in the LCIA for European soils. A further step could be the creation of maps representing the different types of soils and their possible (most probable) evolution in time. The derived map should show the main weathering factors such as soil erosion, flooding and sensitivity to climate change. In addition to this, a review of the literature treating the partitioning of metals among the soil fraction should be done, in order to give acceptable approximations methods (equations or specific trends) at the regional scale, in order to be able to integrate it in a mathematical model and derive more precise fate factors for the substances.

The regionalisation at the contaminated site level of the IMPACT2002 model would be an interesting study to assess the variation of the characterisation factors for smaller scale definitions and the consequent impact derived by the LCA.

The addition of the ERA results for the same site to these damages could help to find discrepancies or contradicting conclusions deriving from the two methods. In addition to this, the ERA could also bring the element necessary to evaluate the ecological value of the site after (or during) the management.

Studies to characterise the impact of substances in the groundwater (at both short and long term) and on the ocean should be effectuated, as they are still considered zero. The IMPACT2002 regional method treats only two categories in human health and one in the water environment. Further studies will add new data, giving more complete results.

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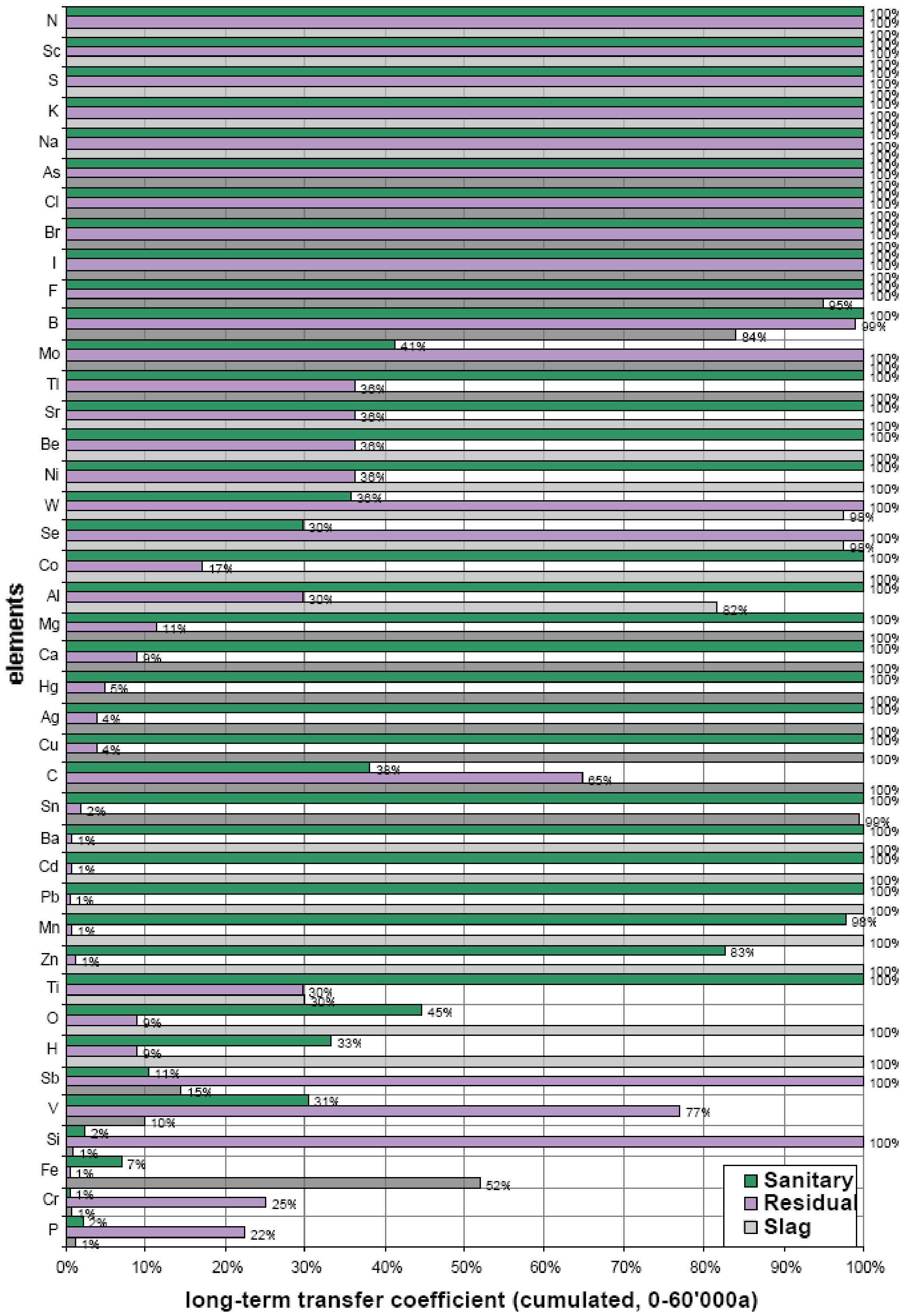
4 Appendix

- Long-term transfer coefficients for MSWI

Long term transfer coefficient for MSWI slag compartments (bottom ash landfill), residual material landfill and sanitary landfill in the case of average municipal solid waste. Sorted according to average over the three values. From Doka (Doka (d), 2007).



- Analysis of the excavation scenario.

Analysis of the excavation scenario. Used method: IMPACT V2.03 / IMPACT 2002+ with modified damage factors for aquatic environment. Indicator: Damage Assessment. Cut-off of the contribution 0.10% (processes contributing to less omitted from the list).

Process	Unit	Total	Process	Total
Total of all processes	**DALY**	**3.7E-02**	Heavy fuel oil, burned in power plant/PT U	1.4E-04
Remaining processes	DALY	1.3E-03	Coke oven gas, at plant/GLO U	1.4E-04
Process-specific burdens, hazardous waste incineration plant/CH U	DALY	9.5E-03	Hard coal, burned in power plant/PT U	1.4E-04
Operation, lorry 28t/CH U	DALY	3.2E-03	Operation, barge tanker/RER U	1.4E-04
Light fuel oil, burned in boiler 100kW, non-modulating/CH U	DALY	2.4E-03	Heavy fuel oil, burned in industrial furnace 1MW, non-modulating/RER U	1.3E-04
Adapted Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH U	DALY	1.5E-03	Lignite, burned in power plant/SI U	1.2E-04
Disposal, cement, hydrated, 0% water, to residual material landfill/CH U	DALY	1.5E-03	Iron ore, 65% Fe, at beneficiation/GLO U	1.2E-04
Natural gas, sour, burned in production flare/MJ/GLO U	DALY	1.2E-03	Nuclear spent fuel, in reprocessing, at plant/RER U	1.2E-04
Diesel, burned in diesel-electric generating set/GLO U	DALY	1.2E-03	Quicklime, in pieces, loose, at plant/CH U	1.2E-04
Lignite, burned in power plant/CS U	DALY	1.0E-03	Copper, primary, at refinery/RLA U	1.2E-04
Hard coal, burned in power plant/ES U	DALY	9.1E-04	Sour gas, burned in gas turbine, production/MJ/NO U	1.1E-04
Heavy fuel oil, burned in power plant/IT U	DALY	8.3E-04	Iron ore, 46% Fe, at mine/GLO U	1.1E-04
Clinker, at plant/CH U	DALY	8.0E-04	Hard coal, burned in power plant/BE U	9.8E-05
Lignite, burned in power plant/ES U	DALY	7.7E-04	Natural gas, burned in power plant/IT U	9.6E-05
Diesel, burned in building machine/GLO U	DALY	7.2E-04	Heavy fuel oil, burned in power plant/ES U	8.6E-05
Operation, transoceanic tanker/OCE U	DALY	6.5E-04	Sweet gas, burned in gas turbine, production/MJ/NO U	8.6E-05
Lignite, burned in power plant/GR U	DALY	5.8E-04	Natural gas, at production onshore/NL U	8.4E-05

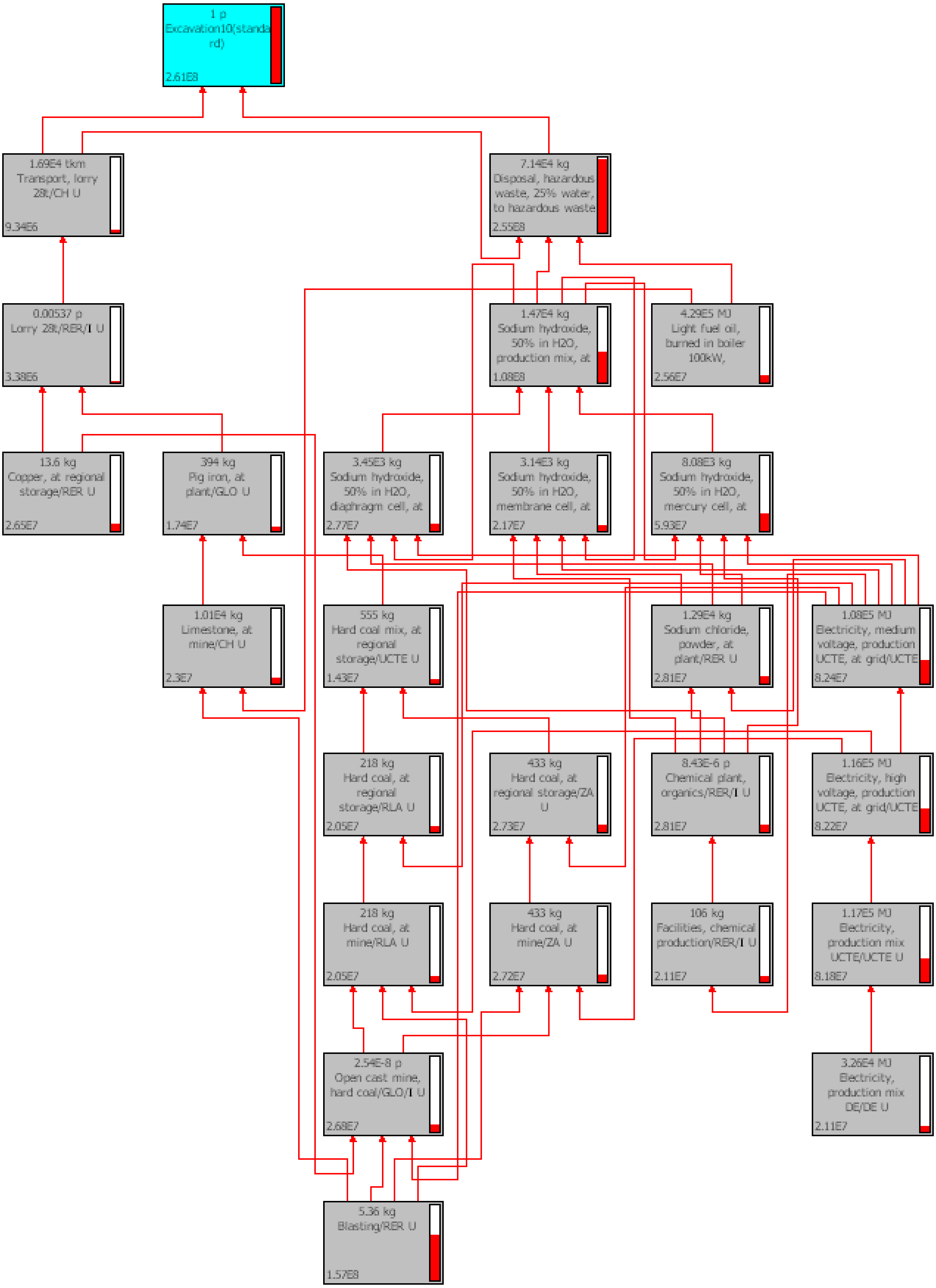
Natural gas, sweet, burned in production flare/MJ/GLO U	DALY	5.7E-04	Light fuel oil, at refinery/RER U	8.2E-05
Heavy fuel oil, burned in refinery furnace/MJ/RER U	DALY	5.3E-04	Disposal, drilling waste, 71.5% water, to landfarming/CH U	7.8E-05
Operation, freight train/RER U	DALY	4.9E-04	Hard coal coke, at plant/RER U	7.6E-05
Operation, lorry 32t/RER U	DALY	4.5E-04	Ferronickel, 25% Ni, at plant/GLO U	7.4E-05
Operation, transoceanic freight ship/OCE U	DALY	3.8E-04	Calcium chloride, CaCl2, at plant/RER U	7.3E-05
Hard coal, burned in industrial furnace 1-10MW/RER U	DALY	3.5E-04	Heavy fuel oil, burned in power plant/FR U	7.1E-05
Lignite, burned in power plant/DE U	DALY	3.3E-04	Heavy fuel oil, burned in power plant/GR U	6.6E-05
Hard coal, burned in power plant/DE U	DALY	2.9E-04	Heavy fuel oil, burned in refinery furnace/MJ/CH U	6.6E-05
Hard coal, burned in power plant/FR U	DALY	2.7E-04	Polyethylene, HDPE, granulate, at plant/RER U	6.4E-05
Lignite, burned in power plant/BA U	DALY	2.4E-04	Refinery gas, burned in flare/GLO U	6.0E-05
Hard coal, burned in power plant/IT U	DALY	2.2E-04	Limestone, at mine/CH U	5.7E-05
Tailings, uranium milling/GLO U	DALY	2.2E-04	Natural gas, burned in industrial furnace low-NOx >100kW/RER U	5.7E-05
Steel, electric, un- and low-alloyed, at plant/RER U	DALY	2.1E-04	Process-specific burdens, municipal waste incineration/CH U	5.6E-05
Lignite, burned in power plant/MK U	DALY	2.1E-04	Natural gas, burned in gas turbine, for compressor station/RU U	5.1E-05
Blasting/RER U	DALY	2.1E-04	Disposal, decarbonising waste, 30% water, to residual material landfill/CH U	4.7E-05
Discharge, produced water, onshore/GLO U	DALY	2.0E-04	Copper, concentrate, at beneficiation/RLA U	4.6E-05
Ferrochromium, high-carbon, 68% Cr, at plant/GLO U	DALY	2.0E-04	Treatment, sewage, to wastewater treatment, class 4/CH U	4.6E-05
Sinter, iron, at plant/GLO U	DALY	1.9E-04	Refinery gas, burned in furnace/MJ/CH U	4.4E-05
Refinery gas, burned in furnace/MJ/RER U	DALY	1.8E-04	Disposal, dust, unalloyed EAF steel, 15.4% water, to residual material landfill/CH U	4.3E-05
Excavation, hydraulic digger/RER U	DALY	1.6E-04	Hard coal, burned in power plant/NL U	4.3E-05
Electricity, at cogen ORC 1400kWth, wood, allocation exergy/CH U	DALY	1.5E-04	Natural gas, burned in power plant/NL U	3.9E-05

- Damages of the three scenarios.

Numerical results of the aspatial model.

Damage category	Unit	Excavation	Phytoremediation	Do nothing
Human Health	DALY	0.03718	-0.01890	0.00557
Impacts on Water	PDF*m^2*yr	2368	6248	7618
Climate Change	kg CO2	133555	-43970	0
Resources	MJ primary	1034647	-1031910	0

- **Blasting in the excavation process network**


1 p
Excavation10(standard)
2.61E8
1.69E4 tkm
Transport, lorry 28t/CH U
9.34E6
7.14E4 kg
Disposal, hazardous waste, 25% water, to hazardous waste
2.55E8
0.00537 p
Lorry 28t/RER/I U
3.38E6
1.47E4 kg
Sodium hydroxide, 50% in H2O, production mix, at
1.08E8
4.29E5 MJ
Light fuel oil, burned in boiler 100kW,
2.56E7
13.6 kg
Copper, at regional storage/RER U
2.65E7
394 kg
Pig iron, at plant/GLO U
1.74E7
3.45E3 kg
Sodium hydroxide, 50% in H2O, diaphragm cell, at
2.77E7
3.14E3 kg
Sodium hydroxide, 50% in H2O, membrane cell, at
2.17E7
8.08E3 kg
Sodium hydroxide, 50% in H2O, mercury cell, at
5.93E7
1.01E4 kg
Limestone, at mine/CH U
2.3E7
555 kg
Hard coal mix, at regional storage/UCTE U
1.43E7
1.29E4 kg
Sodium chloride, powder, at plant/RER U
2.81E7
1.08E5 MJ
Electricity, medium voltage, production UCTE, at grid/UCTE
8.24E7
218 kg
Hard coal, at regional storage/RLA U
2.05E7
433 kg
Hard coal, at regional storage/ZA U
2.73E7
8.43E-6 p
Chemical plant, organics/RER/I U
2.81E7
1.16E5 MJ
Electricity, high voltage, production UCTE, at grid/UCTE
8.22E7
218 kg
Hard coal, at mine/RLA U
2.05E7
433 kg
Hard coal, at mine/ZA U
2.72E7
106 kg
Facilities, chemical production/RER/I U
2.11E7
1.17E5 MJ
Electricity, production mix UCTE/UCTE U
8.18E7
2.54E-8 p
Open cast mine, hard coal/GLO/I U
2.68E7
3.26E4 MJ
Electricity, production mix DE/DE U
2.11E7
5.36 kg
Blasting/RER U
1.57E8

VI Realised or planned publication of project results

Deliverable 5-2:

Publication presenting the outcomes of the project

Date of preparation: 25 October 2008

Start date of project: 10 October 2007 **End date of project:** 9 October 2008

Report author: Hélène Beauchamp

Date: 19th February 2008

Context for the achievement of the deliverable 5.2.

As planned in the deliverable 5-2, the MuSA project proposed the publication of an article presenting the outcomes of the project. This article has been submitted to the the international newsletter of the Society of Environmental Toxicology and Chemistry (SETAC) “SETAC Globe”. It has been published in the January Issue 2009. A second article is currently in preparation.

SETAC Globe, January 2009, vol 10, issue 1, http://communities.setac.net/download/cat-TheGlobe/cat-TheGlobe-1001/TheGlobe-1001-Full.pdf

International workshop at VU University, Amsterdam

Exploring links between Life Cycle Assessment and Ecological Risk Assessment

Sustainable Land Management and Life Cycle Assessment may have more in common more than may be apparent at first view. Such was the main conclusion of the workshop, "Integration of LCA and ERA for contaminated site management: current status and future perspectives," which took place 22-23 September 2008.

The workshop, hosted by the Department of Animal Ecology of the VU University Amsterdam, addressed topics related to the application of Ecological Risk Assessment and Life Cycle Assessment to the evaluation of management strategies for contaminated soil. The workshop came at the end of a year-long European SNOWMAN project focusing on the use of ERA and LCA to compare various management options for contaminated sites. This project, called MuSA, aimed at developing a Multiple Scale Assessment capable of considering site-specific and global impact concerns. Focus was placed on terrestrial sites characterized by historical pollution with metals. There are many of these sites in the densely populated areas of Europe and the U.S., and managers are often at a loss when it comes to decisions about sites that are clearly polluted but do not pose immediate human health risks. Four areas of focus formed the core of the workshop activities: spatial differentiation of environmental and ecological impacts, impacts on biodiversity, ecotoxicology and its application in different assessment approaches, and the bioavailability and mobility of metals in soils.

Several workshop outcomes can be highlighted. Both methods could benefit from the development and use of a detailed soil model for better assessing the fate and effects of different metal species in terrestrial ecosystems. The application of LCA within the process of ERA can give a broader view of the impacts of contaminated sites. Concerning the standard methodology LCA, it is currently impossible to consider land use and its associated impacts with other ecosystem-related impact categories; the need also exists for LCA to explore more precise impact indicators than the current HC50 to be compatible with ERA.

Participants in the workshop hailed from a variety of geographical and professional backgrounds, and included members of the research, policy-making, industrial, and consultancy communities. Promising perspectives for an integrated approach to land-management were identified, based on the conclusions that the tools of ERA and LCA, currently available for evaluating management strategies for contaminated sites, have much in common in terms of input data, current trends towards spatially referenced information bases and international cooperation in the decision-making process. This workshop demonstrated that integrated approaches to decision-making fit into plans of many management entities wishing to continue the pursuit of sustainable development goals. It also demonstrated that existing tools can and should be used to promote transparency in the decision-making progress.

MuSA was funded by the ERA-NET SNOWMAN. More information on MuSA and its results can be found at the following websites: www.snowman-era.net/index.php, www.falw.vu.nl/nl/onderzoek/ecological-sciences/animal-ecology/news/index.asp, and www.ekoconception.eu/web/index.php?category/MuSA-Project.

The SNOWMAN partners are:

Austria	Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft (BMLFUW)	http://www.lebensministerium.at/
Belgium (Flanders)	Openbare Vlaamse Afvalstoffenmaatschappij (OVAM)	http://www.ovam.be
France	Agence De l'Environnement et de la Maîtrise de l'Energie (ADEME)	http://www.ademe.fr
Germany	Umweltbundesamt (UBA)	http://www.umweltbundesamt.de
Netherlands	Stichting Kennisontwikkeling en Kennisoverdracht Bodem (SKB)	http://www.skbodem.nl/
Sweden	Naturvårdsverket (SEPA, Swedish Environment Protection Agency)	http://www.naturvardsverket.se/
United Kingdom	The Environment Agency of England and Wales	http://www.environment-agency.gov.uk/