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BIOLOGICAL FENCE AT THE SITE OF SHELL NETHERLANDS REFINERY, PHASE 2, 3 AND 4

Phase 4: Project evaluation

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Samenvatting

Op 1 januari 1997 is door NOBIS opdracht verleend voor een haalbaarheidsstudie ten aanzien van een biologisch hekwerk op het terrein van Shell Nederland Raffinaderij (SNR), met als doel risicoreductie te bewerkstelligen door middel van biologische afbraak van de verontreiniging met koolwaterstoffen.

In fase 2 en 3 zijn drie prototypen van hekwerken ontworpen, geïnstalleerd, geëxploiteerd en heeft monitoring plaatsgevonden. In dit rapport worden de resultaten van het project geëvalueerd (fase 4).

Trefwoorden

Gecontroleerde termen: aromatic organic compounds, biodegradation, maintenance, organic compounds	**Vrije trefwoorden:** -

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Abstract

On January 1 1997 NOBIS commissioned a feasibiliy study regarding a biological fence at the site of Shell Netherlands Refinery. The aim of the study was to reduce risk of migrating pollution by biodegradation of contaminants (hydrocarbons).

In phase 2 and 3 three test fence were designed, constructed, operated and monitored. This report gives the results of the project evaluation (phase 4).

Keywords

Controlled terms:	**Uncontrolled terms:**
aromatic organic compounds, biodegradation, maintenance, organic compounds	-

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SAMENVATTING

Biological fence at the site of Shell Netherlands Refinery, phase 2, 3 and 4

Dit rapport geeft een overzicht van de resultaten van de haalbaarheidsstudie van een biologisch hekwerk op het terrein van Shell Nederland Raffinaderij.

De enige methode om verspreiding van verontreiniging over de terreingrenzen heen tegen te gaan, was grondwateronttrekking en zuivering. Het zou echter mogelijk moeten zijn om biologische technieken te gebruiken om het uitstromende grondwater in de bodem te zuiveren. Het grondwater stroomt daarbij ongehinderd, terwijl lage vrachten aan verontreinigingen in het grondwater worden afgebroken in een biologisch geactiveerde zone. Het concept is verenigbaar met een strategie van risicoreductie van verspreiding van verontreiniging over terreingrenzen.

Conclusies
De haalbaarheid van het concept is gedemonstreerd door de installatie van 3 prototypen van biologische hekwerken in het veld en te demonstreren dat:

- de hekwerken verspreiding van verontreiniging tegengaan;
- aërobe afbraak van koolwaterstoffen naar alle waarschijnlijkheid heeft plaatsgevonden met een afbraaksnelheid van 0,5 - 0,65 d^{-1};
- het mogelijk is om de processen te beïnvloeden, maar dat controle en sturing van de processen moeilijk is omdat de beschikbaarheid van zuurstof wordt beïnvloed door (de interactie van) verschillende chemische en fysische processen;
- als gevolg van de hoge initiële verontreiniging ter plaatse van de hekwerken in november 1999 nog geen acceptabele concentraties waren bereikt;
- het eindresultaat kan worden voorspeld door modellering.

De algemene conclusie is dat een biologisch hekwerk de verspreiding van koolwaterstoffen kan voorkomen wanneer voldaan is aan bepaalde randvoorwaarden.

Drie verschillende hekwerken zijn aangelegd en geëxploiteerd, te weten:

- *Hekwerk 1*
  Luchtinjectie door middel van horizontaal geboorde drains.
- *Hekwerk 2*
  Luchtinjectie door middel van horizontale drains, aangelegd met een drainmachine in een grindkoffer.
- *Hekwerk 3*
  Luchtinjectie door middel van 20 gepulsde verticale injectiefilters.

Monitoring
Het monitoringssysteem van de drie hekwerken bestond uit:

- 3 meetdrains van 15 meter lengte, met drie meetpunten in iedere drain. De drains zijn aangelegd met een draineermachine waarbij de omringende grond is vervangen door grind;
- 12 verticale grondwaterfilters.

Resultaten
Over het algemeen genomen zijn de concentraties koolwaterstoffen met 50 % gedaald. Over de hele periode zijn de concentraties (gemiddeld) gereduceerd met 2000 µg/l (hekwerk 2) tot 7000 µg/l (hekwerk 1). Als gevolg van de hoge initiële concentraties is de reductie echter nog niet voldoende om acceptabele concentraties te bereiken. Modellering geeft aan dat acceptabele

concentraties kunnen worden bereikt na 1 jaar (hekwerk 3), 3 jaar (hekwerk 2) en 6 jaar (hekwerk 1). De afname kan niet worden verklaard door de instroom van schoon water of verdamping. Een verzadigd grondwater- en transportmodel is gebruikt om de afname te analyseren. Op basis van de modellering mag worden geconcludeerd dat de afname in alle 3 de hekwerken het gevolg is van biologische afbraak. De gefitte afbraaksnelheid in de hekwerken is vergelijkbaar met waarden tussen 0,5 en 0,65 d^{-1}. Deze waarden komen overeen met de waarden die zijn bepaald met de kolomexperimenten van fase 1.

Optimalisatie van zuurstofsuppletie

Metingen van de zuurstofconcentraties en redoxpotentiaal laten een grote variatie zien in tijd en plaats, zelfs bij een constant injectieregime (augustus - november 1999). De zuurstofconcentraties in hekwerk 2 en 3 zijn vergelijkbaar. De beste resultaten zijn behaald bij hekwerk 1, met duidelijk verhoogde gehalten zuurstof (> 5 mg/l) in de bovenstroomse meetdrain. Over de hele periode genomen lijkt de zuurstof overal in de hekwerken te worden verspreid, maar een structurele toename van de zuurstofconcentratie of redoxpotentiaal is niet gemeten.

Tracertests met helium hebben aangetoond dat bij hekwerk 2 de geïnjecteerde lucht rechtstreeks naar het oppervlak wordt getransporteerd. De grindkoffer bij hekwerk 2 kan als een grote preferente stroombaan worden beschouwd. Bij hekwerk 1 en 3 wordt de lucht horizontaal naar de haven getransporteerd. Boorprofielen geven aan dat daar dunne kleilagen aanwezig zijn. De hogere heliumgehalten bij hekwerk 2 gaan niet samen met hogere zuurstofconcentraties. Dit is een aanwijzing dat de zuurstofuitwisseling van lucht naar grondwater een lage efficiëntie heeft, mogelijk door het transport van lucht in de vorm van bellen in combinatie met een korte verblijftijd. Luchttransport en zuurstofoverdracht van lucht naar grondwater zijn gevoelig voor de bodemheterogeniteit en de korrelgrootteverdeling.

Technische vergelijking

De flexibiliteit voor wat betreft de aanleg en luchtinjectie is hoog bij het gebruik van verticale filters en laag bij horizontale drains. Verticale filters blijken de meest kosteneffectieve techniek te zijn. In de aanwezigheid van gebouwen en ondergrondse infrastructuur kan een horizontaal geboorde drain echter weleens het enige alternatief zijn.

De kosten van biologische hekwerken zijn vergelijkbaar met die van conventionele systemen die zijn aangelegd op het terrein van Shell Pernis. Biologische hekwerken hebben minder negatieve milieueffecten.

Ontwerp van een biologisch hekwerk

Gecombineerde modellering van grondwaterstroming, stoftransport en biologische afbraak in de verzadigde zone, zoals is toegepast in fase 1, is een krachtig hulpmiddel om de voorwaarden op te stellen voor een hekwerk, omdat het de informatie van verschillende processen combineert.

SUMMARY

Biological fence at the site of Shell Netherlands Refinery, phase 2, 3 and 4

This report gives the results of the feasibility study of a 'biological fence' at the site of Shell Netherlands Refinery.

For a number of years now the sole method used to combat the spreading of pollutants across the boundaries was the construction of a system of groundwater extraction and the purification of the water thus pumped to the surface. It should, however, be possible to give biological degradation processes an opportunity to 'filter' the outward-spreading groundwater in the soil. The groundwater is allowed to pass unimpeded while the (relatively) low loads of pollutants are degraded in an activated biological system. This concept is very compatible with a strategy of risk reduction with the effort aimed at preventing the spreading of pollution across site boundaries.

Conclusions

The feasibility of the concept was demonstrated in this project by constructing three prototype biological fences in the field and demonstrating that:

- the biological fences can prevent spreading of contaminants;
- aerobic degradation of hydrocarbon is likely to take place at a degradation rate of 0.5 - 0.65 d^{-1};
- it has been possible to influence the process but controlling and optimizing was very difficult as oxygen availability is influenced by several physical and chemical processes;
- due to high initial concentration at the test site acceptable levels were not yet reached. Modelling showed that adequate end result is likely to be reached;
- the end result can be predicted by modelling.

The overall conclusion on the applicability of a biological fence is that when certain conditions are met a biological fence can prevent spreading of hydrocarbons.

Three different fences were constructed and operated during the pilot test. The different fences are:

- *Fence 1*
  Air injection through horizontal drains, installed by horizontal drilling.
- *Fence 2*
  Air injection through horizontal drains, installed by a drainage machine. The soil above the drain was replaced (100 %) by gravel. At the column experiments the highest degradation rate was measured when 80 % or more gravel was used as filling material. From this it is expected that fence 2 will perform best.
- *Fence 3*
  Air injection though 20 vertical injection filters installed by pulsing. The filters are placed in a serrated line. This design is expected to have a wide range of application.

Monitoring

The monitoring system per fence consisted of:

- 3 measuring drains of 15 m, with three sampling tubes in each measuring drain. The drains are installed with a drainage machine. The soil surrounding the drains was replaced by gravel;
- 12 groundwater monitoring wells.

Performance
Overall, the hydrocarbon levels showed a decrease of 50 %. Over the total period the reduction of the average total hydrocarbon concentration ranges from about 2000 µg/l (fence 2) up to 7000 µg/l (fence 1). However, the reduction is not yet sufficient as acceptable levels are not reached yet due to high initial levels of contamination. Modelling shows that acceptable levels will be reached after 1 year (fence 3), 3 years (fence 2) and 6 years (fence 1). The decrease is not likely to be caused by inflow of non-contaminated groundwater or volatilization. A saturated groundwater and solute transport simulation model including a degradation module was used to analyse the results. From the modelling it is concluded that it is likely to suppose that biological degradation is proceeding in all fences. The rate of degradation between the fences is comparable, and has a value between 0.5 and 0.65 d^{-1}.

The simulated values of biological degradation in the fences are about the same as the one determined in column experiments of phase 1. The rates of mineral oil degradation in the fences, make sure that we are dealing with aerobic biodegradation: anaerobic biodegradation of mineral oil can not be this fast.

Optimizing oxygen supply
Measurements of oxygen level and redox potential show a large variation in place and in time, even at a constant flow regime (August 1999 - November 1999). The oxygen levels in fence 2 and fence 3 are considered alike. The best results are reached at fence 1 with considerably higher (> 5 mg/l) oxygen levels in the upstream measuring drain. Over time oxygen seems to be distributed through out the fences. However, from the zero measurements until now, no structural increase of oxygen level or redox potential was measured.

Tracer tests with helium at the soil surface indicated that at fence 2, the injected air was transported vertically to the surface of the fence. The gravel ditch at fence 2 can be considered as 'one preferential flow path'. At fence 1 and 3, the air is transported horizontally to the harbour. Soil profiles indicated that small clay layers are present at the fences 1 and 3. From research by Elder and Benson [1999] we also know that injected air in finer media of about 0.2 mm and smaller (like fine to medium sand), passes nearly horizontally by way of air channels through the soil. The higher recovery of helium at the tracer tests did not result in higher oxygen levels. This indicates that oxygen transfer from air to groundwater is less efficient, probably because of the 'bubble' air transport in combination with a relatively short residence time of air before emergence at the surface of the fence. Air transport and oxygen transfer from air to groundwater are sensitive to soil heterogeneity and pore size distribution.

The construction of fences 1 and 2 went with serious problems. The construction of fence 3 went without problems. During the test period of the pilot no maintenance problems occurred for the three different fences.

To ensure an equal air distribution over the whole length of the drain, it is necessary always to have a high injection flow rate for fence 1 and 2 (relatively with respect to the oxygen demand). Lower air injection rates at fence 3, vertical filters, don't have an impact on the distribution of the injected air.

Technical comparison
The flexibility with respect to construction and air injection of fences with vertical filters is high. The flexibility of fences with horizontal injection drains is low.

Based on the technical comparison the fence with vertical filters, is the most cost-effective technique to construct and operate a full-scale aerated biological fence. However, in the presense of buidings and infrastructure horizontal drilled drain can be the only alternative.

The costs of biological fences are comparable to conventional systems installed at the site of Shell Pernis. Biological fences have less negative environmental influences than conventional techniques. Failing equipment is likely to cause an unacceptable spreading faster with conventional systems than with biological fences. The reliability of conventional systems is regarded higher as the influence of these fences can be easier assessed.

Design of a biological fence
Combined modelling of groundwater flow, solute transport and degradation in the saturated zone, as is performed in phase 1, is a powerful instrument to put up the conditions for the design of a biological fence. It brings together the information from the preliminary investigations about the various processes.

Modelling could be improved when the transfer of oxygen from air to groundwater is better accounted for. The implementation of biological fences for aerobic degradation of hydrocarbons would be improved when more knowledge is gained about transport of air and transfer of oxygen from air to groundwater.

CHAPTER 1

INTRODUCTION

In January 1997, NOBIS commissioned a feasibility study regarding a biological fence at the site of the Shell Netherlands Refinery. The aim of this study was to reduce risks of migrating pollution by applying biodegradability techniques.

The Shell Netherlands Refinery site is part of Shell's overall Pernis site, part of which houses a refinery and storage facilities for petroleum products (Shell Netherlands Refinery: SNR), while the remainder houses chemical plants and storage facilities (Shell Netherlands Chemical: SNC). The site is generally covered with a landfill layer varying between 2 to 5 metres in thickness. This upper layer rests on a layer of clay, which contains a layer of sand at approximately 8 m below the surface. The aquifer starts at a depth of approximately 20 - 25 m below the surface. At the boundaries of the site the phreatic groundwater flow (in the landfill layer) is directed outward in the direction of the first and second 'Petroleumhaven' docks and a polder area. The centre of the site is mainly characterized by infiltration into the aquifers. At a number of locations along the boundaries of the site it has been found that pollution (mineral oil and Volatile Aromatic Hydrocarbons: VAH) is being transported across the site boundaries. The site can be regarded as representative of industrial sites in the Botlek area.

For a number of years now the sole method used to combat the spreading of pollutants across the boundaries was the construction of a system of groundwater extraction and the purification of the water thus pumped to the surface (Pump and Treat). It should, however, be possible to give biological degradation processes an opportunity to 'filter' the outward-spreading groundwater in the soil. The groundwater is allowed to pass unimpeded while the (relatively) low loads of pollutants are degraded in an activated biological system.

The potential of microbiological degradation of VAH and mineral oil has been adequately demonstrated by now; both in the laboratory and in the field. In general, the limiting factor is not the quantity of micro-organisms present but rather the conditions. Much more research has been carried out into the effectiveness of the technique and the application and optimization of biorestoration over an entire site, than into activating biological activity in a small zone or linear structure such as for instance along the site boundaries. The present project is directed at the concept of preventing the spreading of pollution by stimulating biodegradation in a narrow zone along the site boundaries.

This concept is very compatible with a strategy of risk reduction with the effort aimed at preventing the spreading of pollution across site boundaries. The feasibility of the concept is demonstrated by constructing a number of prototype biological fences in the field and demonstrating the achieved degree of the reduction of risks.

This report describes the evaluation of the study (phase 4). The report contains:

- a summary of the results of the former phases (see chapter 2);
- the objectives and research questions of this phase (see chapter 3);
- a description of the three test sections (see chapter 4);
- an evaluation of the three test sections (see chapter 5);
- a comparison and evaluation of biological fences versus conventional fences (see chapter 6);
- drafting a guideline for application of a biological fence (see chapter 7);
- conclusions (see chapter 8).

As evaluation is a central theme, this report will be an integration of phase 1 (preliminary investigations), phase 2 (construction of the fences and optimizing air injection) and phase 3 (monitoring).

CHAPTER 2

SUMMARY OF FORMER PHASES

The feasibility study that NOBIS commissioned is divided into four phases:

- phase 1: Preliminary project design studies, from January 1997 till January 1998 [CUR/NOBIS, 1999];
- phase 2: Construction of the biological fences and optimization of air injection, from September 1999 till March 1999 [Heijnen and Vis, 1999];
- phase 3: Operation of the fences and monitoring, from March 1999 until December 1999 [Heijnen and Praamstra, 1999];
- phase 4: Evaluation, December 1999.

Phase 1: Design studies

Phase 1 was completed and is reported in the CUR/NOBIS report 'Characterization and design of a biological fence' [CUR/NOBIS, 1999]. In this phase, characterization of the site and designing took place. Summarizing, the following aspects have been concluded and/or worked out:

- ***Experimental*** (details are given in the TNO report on the column study [Van Liere et al., 1998]):
  - from various filling materials gravel is selected on physical properties as the optimal backfill material;
  - column experiments showed that best results were obtained with backfill consisting of more than 80 % gravel;
  - column experiments showed that degradation is limited by oxygen;
  - iron precipitation in the column did not result in pressure build up.
- ***Field*** (details are given in the CUR/NOBIS report 'Characterization and design of a biological fence' [CUR/NOBIS, 1999]):
  - the site is polluted with mineral oil and BTEX. The mineral oil consists largely of light components (C6 - C12);
  - the groundwater flow is directed straight to the harbour;
  - the groundwater is anaerobic;
  - on indicative calculations of oxygen demand an injection rate of about 0.38 - 0.5 m^3 air/hour mfence was calculated;
  - an outline of the monitoring was given, in which a distinction was made between process parameters (for instance O_2) and hydrocarbons. The idea behind this was that aeration of the fence was expected to be a faster process than decrease of contaminant concentration by degradation. The process parameters would give an insight in the operation of the fences more quickly. Beside the conventional vertical standpipes for monitoring, horizontal drains should be tested.

On the basis of the results of phase 1, NOBIS decided to proceed with phases 2, 3 and 4 of the project. These phases are outlined in more detail in the Basic Project Plan [Heijnen, 1998].

Phase 2: Construction of the test sections

This phase involved:

1. design of the test sections;
2. the construction of test sections by installation of the air supply systems and the monitoring system;
3. an optimization period of the air supply.

The construction of the system started in September 1998. A description of the installation is given in chapter 4.

Base line measurements were performed at December 7th and 8th 1998, after construction and 1 week before the air injection started. The aim of these baseline measurements was to have a reference before influencing the chemical and biological conditions in ground and groundwater. A description is given in chapter 4.

The air injection into the test sections started at December 14th 1998. From this moment on air injection regimes were evaluated and optimized on the basis of the process parameters: the oxygen level and redox potential.

Monitoring during this period was to show whether or not it is possible to influence and optimize conditions for aerobic degradation of hydrocarbons (HC): an oxygen level of about 8 mg/l and a corresponding high (positive) redox potential.

Until February 3rd 1999, 5 different injection regimes (flow rates ranging from 8 to 16 m^3/hour, continuously as well as intermittently) were performed to try to maximise the oxygen level. From February 3rd until May 7th a constant flow regime was used. Injection drain 1B (on valve 7) of fence 1 did not work anymore since May.

The monitoring results during this optimization period showed in IWACO report 1082750 [Heijnen and Vis,1999], that no significant and consistent oxygen level or redox potential rise was measured. Although small changes were noticed, it was not clear at that moment whether or not it would be possible to optimize the conditions for aerobic degradation of hydrocarbons. That is why in addition to the monitoring proposed for phase 3 in the Basic Project Plan [Heijnen, 1998] additional measurements and calculations were performed to find an explanation for the apparent lack of oxygen in the groundwater.

Phase 3: Monitoring

This phase originally involved actual operating and monitoring of the test sections. This monitoring should answer the following questions:

1. What is the performance of the fences:
    - does hydrocarbon content decrease?
    - is this decrease a result of biological degradation?
2. Can conditions be controlled with air injection:
    - is the air really injected into the soil?
    - where does the air go once it is injected?
    - is the injected amount of air sufficient to rise oxygen level?
    - is it possible to optimize conditions for aerobic degradation of hydrocarbons?

Performance of the fences

Overall, the hydrocarbon analysis show a decrease of hydrocarbon content. Increases of hydrocarbon content are seen upstream measuring points, indicating an inflow of contaminated groundwater. The decrease is seen for non-volatile components as well: at several points the decrease of these non-volatile components is larger than the decrease of volatile components, indicating that volatilization is not the (only) explanation for the decrease. Although the trends from the analysis are hardly confirmed by the FOC-sensors (see appendix C), we conclude that the decreasing concentrations are due to biological degradation. The relatively high carbon dioxide content of the soil air confirms this conclusion.

As air is injected into the soil whereas little oxygen is measured, it is possible that the supplied oxygen is used by aerobic degradation. Another possibility is that the degradation proceeds under less anaerobic conditions (with iron as electron acceptor) as has been seen in the column experiments of phase 1. In phase 4 this was further investigated.

Optimizing conditions

The measurements show that (until now) we did not succeed to measure a structural increase of oxygen level or redox potential. So, although we know that injected air reaches the soil, we do not have clear evidence that we are able to optimize conditions for aerobic degradation of hydrocarbons. As the oxygen level varies temporally, over time oxygen seems to be distributed throughout the fences.

CHAPTER 3

OBJECTIVES AND RESEARCH QUESTIONS OF THIS PHASE

The objective of this phase is to evaluate and conclude on the project. The results of the study were evaluated from the point of view of the questions regarding the feasibility of biological fences, which were asked at the start of the project [Schipper and Satijn, 1996]:

- Does degradation take place?
- How fast does degradation take place?
- Is the end result adequate?
- How does one predict the end result?
- Is it possible to influence and optimize the process?
- Is it difficult to measure and track?

These questions are subject in chapter 5: evaluation of the performance of the three tested biological fences and the optimization and controlling of the processes.

Other research questions that were defined [Schipper and Satijn, 1996] are:

- What does the basic design of a biological fence look like? In chapter 4 a description is given of the design of the three tested biological fences.
- To what extent is it possible to operate a biological fence at a large-scale (see section 5.3)?
- What is the practical added value of a biological fence compared to conventional systems and to what extent is a biological fence also competitive in financial terms (see chapter 6)?
- To what extent is a biological fence reliable (see chapter 6)?
- How can the operation of a biological fence be monitored; what added value do field measuring techniques offer (see chapter 7)?

CHAPTER 4

DESCRIPTION OF THE THREE TEST FENCES

The three different fences, constructed and operated during the pilot test, are being compared in this chapter.

4.1 Injection system

For a specific overview we refer to appendix A.

Fence 1
Air injection by means of horizontal drains. Two drains (length 30 m) were installed by the use of a horizontal drilling machine. For one drain, a normal perforated drain with casing was used. This drain has been out of order since spring of 1999. The other drain was a so-called 'no-dig drain', with a perforated casing. This drain is still functioning. In this fence the soil remains undisturbed. This fence is not expected to give the best performance, but is applicable on a wider range of sites.

Fence 2
Air injection by means of horizontal drains. Two drains were installed by a drainage machine. The soil above the drain was replaced (100 %) by gravel (diameter of 4 - 16 mm) up to the groundwater level. It is expected that after a while original soil will be washed into the gravel ditch because of the flow of water from up gradient to down gradient. At the column experiments the highest degradation rate was measured when 80 % or more gravel was used as filling material. From this it is expected that fence 2 will perform best.

Fence 3
Air injection by means of vertical injection filters. Twenty filters are installed by pulsing. The filters are placed in a serrated line, with perforation from 4 to 4.5 m bgl (metre below ground level). This design is expected to have a wide range of application. By applying this design together with a horizontal system the differences between horizontal and vertical air supply can be investigated.

An overview of the installation is given in appendix A. The drains were installed at about 4 m bgl on top of a confining clay layer. Soil profiles of the implementation area indicated that at the north side a shallow clay layer was present that would disturb air transport. As the drainage machine, used for the construction of fence 2, cuts through the clay layer this difficulty is overcome. That is why fence 2 is located at the north side of the area. The air injection rate can be adjusted per drain at the fences 1 and 2 and per set of five injection filters at fence 3. Air injection rates were determined and adjusted with a flow meter. Since August 17th 1999 the following flow rates have been injected:

- fence 1: 10.3 m^3/hour (1 drain);
- fence 2: 24.7 m^3/hour (12.35 m^3/hour each drain, 2 drains);
- fence 3: 8.2 m^3/hour (about 0.4 m^3/hour each filter, 20 filters).

These injection rates were determined based on:

- the results of the start test;
- technical reasons (pressure build up for reaching end of the drains);
- to minimize bubbles in the harbour (fence 3).

4.2 Monitoring system

An overview of the monitoring system is given in appendix B.

Groundwater

The monitoring system per fence consists of:

- 3 measuring drains of 15 m, 2 straight above line of injection, 1 downstream of the fences. The drains are installed at 2.5 m bgl. In each measuring drain three sampling tubes were installed, at 2.5 m from both ends of the perforation and in the middle of the drain. The drains are installed with a drainage machine. The soil surrounding the drains was replaced by gravel;
- 9 groundwater monitoring wells (1 - 9) with perforations from 2.5 - 3.5 m bgl;
- 3 groundwater monitoring wells (12, 11 and 10) at 1.8, 2.6 and 3.8 m bgl respectively with 0.2 m of perforation.

From this monitoring system, groundwater samples were taken periodically for field measurements (oxygen content, redox potential etc.) and chemical analysis in the laboratory (C6 - C12, C10 - C40, BTEX). Besides that, in the measuring drains as well as in the monitoring wells, the total hydrocarbon content was continuously monitored by Fibre Optic Sensors. However, the sensor values turned out to be insecure. We refer to appendix C for an evaluation of the Fibre Optic Sensors. For the interpretation of the operation of the fences we therefore only use the results of chemical analysis.

Soil air

For soil air measurements at depths of 0,5 to 1,0 m bgl, lances as well as permanent monitoring filters are used. The samples were analysed with a PID (total hydrocarbon content) and a landfill analyser (carbon dioxide, oxygen and methane).

4.3 Initial contamination present at test sites

With an average total hydrocarbon concentration in the groundwater of about 17000 µg/l, fence 1 is the most polluted fence. The average total hydrocarbon concentration in fence 2 is about 5000 µg/l and in fence 3 about 4000 µg/l. In all fences the highest concentrations are mainly formed by the volatile fraction of mineral oil (C6 - C12), upstream as well as downstream. With increasing depth (filters 12, 11, 10) concentrations decrease.

The average total hydrocarbon concentration per fence, based on all measurement points, is shown in table 1. For a complete overview of the initial hydrocarbon concentrations we refer to appendix D.

Table 1. Average total hydrocarbon concentration initially present in groundwater (December 1998).

	average total hydrocarbon (µg/l)		
	fence 1	fence 2	fence 3
December 1998	16800	4900	3900

Fence 2 shows a decrease in pollution from up gradient to down gradient. In the other fences 1 and 3 this is not the case: the degree of contamination varies throughout the fences. This has not changed during the experiment.

CHAPTER 5

EVALUATION OF THE THREE TESTED BIOLOGICAL FENCES

5.1 Performance expressed in hydrocarbon decay

5.1.1 *Development of hydrocarbon content*

Overall, the hydrocarbon levels showed a decrease of hydrocarbon content. Table 2 shows the change in average total hydrocarbon concentration in groundwater from December 1998 until November 1999. For a complete overview of the progress of hydrocarbon concentrations (mineral oil as well as BTEX) we refer to appendix D.

Table 2. Change in average total hydrocarbon concentration in groundwater from December 1998 until November 1999.

	average total hydrocarbon (µg/l)		
	fence 1	fence 2	fence 3
December 1998	16800	4900	3900
September 1999	11500	2700	2000
October 1999	9200	1750	1800
November 1999	9000	2700	1600

Over the total period the reduction of the average total hydrocarbon concentration ranges from about 2000 µg/l (fence 2) to 8000 µg/l (fence 1). The interim increase of hydrocarbon concentration at fence 2 from October to November is an exception to the image of reduction.

In table 3 the changes in total hydrocarbons at individual measuring points are summarized.

Table 3. Change in total hydrocarbon concentration in groundwater at individual measuring points from December 98 till November 99

fence	measuring drains			monitoring wells			total number of points with a clear decrease
	number of points +	number of points +/-	number of points -	number of points +	number of points +/-	number of points -	
1	0	1	8	0	5	7	15 out of 21
2	0	5	4	2	3	7	11 out of 21
3	0	2	4	2	2	8	12 out of 18
total	0	8	16	4	10	22	38 out of 60

In a majority of points (38) a clear decrease (-) of total contaminant concentration has occurred. This trend is most obvious at fence 1: 71 % of the measuring points shows a clear decrease. At fences 2 and 3 this is respectively 52 % and 67 %.

At a few (4) points a clear increase (+) has occurred. Three of these points are upstream, indicating an inflow of contaminated groundwater. At several points (18) the trend is not clear (+/-).

The decrease is that large that it is not likely to be caused by inflow of non-contaminated groundwater: the retardation of mineral oil is too large for that; for a quantitative foundation we refer to section 5.1.3. Besides that, increases of hydrocarbon content are seen in upstream measuring points, indicating an inflow of contaminated groundwater (we refer to appendix D, fences 2 and 3). The decrease is seen for non-volatile components as well: at several points the decrease of these non-volatile components is larger than the decrease of volatile components, indicating that volatilization is not the (only) explanation for the decrease. Implicitly, we therefore conclude that the decreasing concentrations are due to biological degradation.

When it is concluded that degradation is occurring the next questions arise:

- to what extent and at what rate is degradation occurring?
- is this a reasonable degradation rate?
- as the fences differ in initial concentrations and decrease, how can the fences be compared?
- what kind of degradation is taking place: aerobic or anaerobic degradation?
- are the biological fences capable of reducing pollutants to such levels that they cause no unacceptable levels of pollutants outside the site boundaries?

A saturated groundwater and solute transport simulation model including a degradation module was used (see section 5.1.3) to analyse the results and to give answers to the questions above.

5.1.2 *Efficiency of reduction of hydrocarbon levels*

Are the biological fences capable of reducing pollutants to such levels that they cause no unacceptable levels of pollutants outside the site boundaries? In the Netherlands an unacceptable level for an individual component or for groups of components in soil and groundwater is the so-called intervention criterion (I-value). So a remediation should at least meet this intervention criterion. The I-values for the relevant hydrocarbons are shown in table 4. Table 4 shows as well the fraction of points throughout the fences that exceeded this criterion in November 1999.

Table 4. Points with unacceptable levels hydrocarbon in November 1999.

	sum mineral oil	benzene	toluene	ethylbenzene	xylenes
I-criterion	600	30	1000	150	70
	points > I	points > I	points > I	points > I	points > I
all fences	80 %	23 %	0 %	9 %	29 %
fence 1	81 %	0 %	0 %	14 %	52 %
fence 2	86 %	52 %	0 %	14 %	24 %
fence 3	67 %	11 %	0 %	0 %	0 %

From this it becomes clear that acceptable levels are not reached yet. This is caused by the high levels of pollution initially present in the fences matrix. Besides the inflow of hydrocarbons, this initial amount of hydrocarbons in the fences has to be dealt with too.

5.1.3 *Modelling solute transport*

To get an indication of the rate of biological degradation in the fences 1, 2 and 3, the development of concentrations is simulated in a computer model for solute transport. The main advantage of the use of a model for solute transport is that processes like convection, dispersion, sorption and biological degradation are integrated. Because the light fraction of mineral oil is the most dominant one in the field and because this fraction was observed by TNO during the column tests [Van Liere et al., 1998], we modelled solute transport for the fraction C6 - C12. Moreover, it is expected, on base of experience and literature, that BTEX breaks down faster under aerobic conditions than mineral oil does.

For the computer simulation we used the program SORWACO (version 0.03), developed by IWACO. SORWACO is a one dimensional, numerical solute transport model. In SORWACO it is possible to divide the soil into a few cells, for which density, porosity and carbon content can be defined. So, for this case it is possible to split up the flow path into cells which consist of gravel (measuring drains) and cells which consist of clayey sand (rest of the fence).

Furthermore, SORWACO calculates with a first order degradation ($k = \ln(C_0/C_t)/t$). For the model input we refer to appendix E.

We refer to the figures in appendix E for an overview of the initial groundwater contamination used as input for the model, based on the averages of the concentrations measured in December 1998. From the initial situation it can be concluded that the present contamination in the fences is not homogeneously distributed. Partly this can be explained by the presence of the drains in a gravel ditch, which is initially non-polluted and where hardly any sorption of mineral oil takes place.

Calculations
Calculations were executed for the following scenarios:

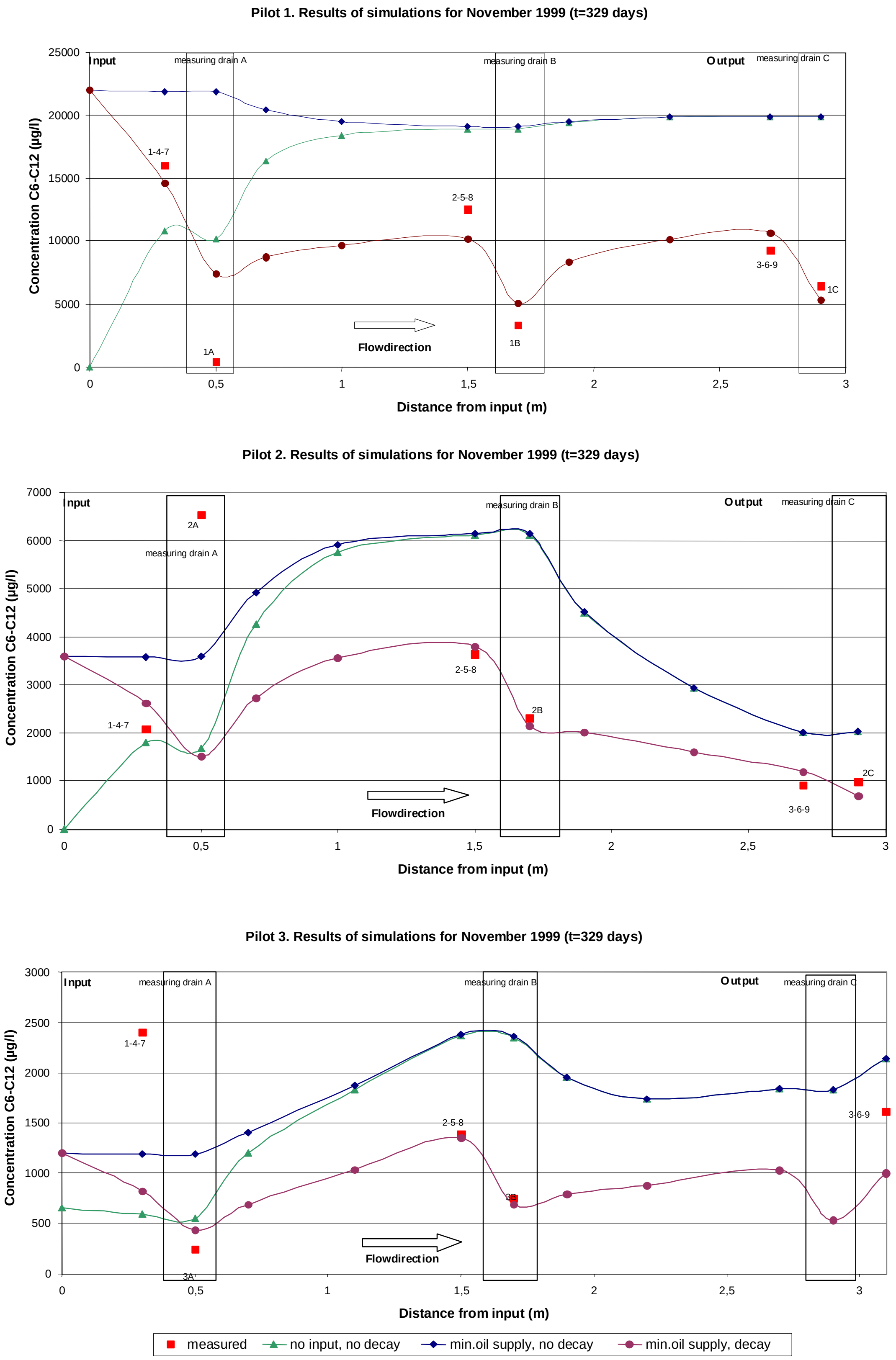
1. no supply of mineral oil to the fences, no biological degradation;
2. supply of mineral oil to the fences, no biological degradation;
3. supply of mineral oil to the fences, biological degradation.

Ad 1.
This scenario is calculated to predict the situation in November 1999 if there is no new supply (input) of mineral oil and biological degradation of the initially present mineral oil does not occur. Is the inflow of non-polluted water the cause of the decrease in mineral oil concentrations in the fences?

Ad 2.
This scenario is calculated to predict the situation in November 1999 if the supply of mineral oil is continuous and biological degradation of mineral oil does not occur.

Ad 3.
This scenario is calculated to predict the situation in November 1999 if the supply of mineral oil is continuous and biological degradation of mineral oil occurs. Is it possible to predict the measured decrease in mineral oil concentrations in the fences by assumption of biological degradation? On base of the knowledge of groundwater flow, organic carbon content of the soil and sorption of C6 - C12, the biological degradation rate was used as a fit parameter to simulate the measured concentration development in the fences.

Results
In figure 1, 2 and 3 the simulation results for November 1999 are presented. At this time the operational period of the fences is 329 days (from December 1998 on). In the figures the averages of the measured concentrations in November 1999 are presented as well.



Pilot 1. Results of simulations for November 1999 (t=329 days)

Pilot 2. Results of simulations for November 1999 (t=329 days)

Pilot 3. Results of simulations for November 1999 (t=329 days)

Fig. 1 - 3. Results model simulations of concentration development in all fences for November 1999.

Overall

From the results it is concluded that the measured concentrations of mineral oil (C6 - C12) can not be simulated without the proceeding of biological degradation. Even an inflow of non-polluted groundwater into the fences can not be the reason for the measured decrease of mineral oil (scenario 1), due to the relatively large retardation of the initially present C6 - C12 fraction in combination with the relatively short operational period of the fences. By assuming biological degradation, the measured concentrations and the shape of the spatial concentration development can be simulated. In appendix F measured and simulated values are compared for each fence and the correlation coefficient is calculated.

Fence 1

At a first order degradation rate (k) of about 0.65 d^{-1} (corresponding with a half life of 1.07 day), the measured concentration development can be simulated quite good, except for the concentration in drain A. It seems that biological degradation is locally higher here. This is a plausible explanation because pilot 1 has got just one operational injection drain (1A) left. The other injection drain 1B is obstructed since May 1999. Without the values of drain A, the correlation coefficient (R^2) between simulated and measured values is 0.89.

Fence 2

At a first order degradation rate of about 0.5 d^{-1} (corresponding with a half life of 1.39 day), the measured concentration development can be simulated quite good, except for the concentration in drain A. This concentration does not differ much from the concentration in December 1998. It is known from field observation that an oil floating layer (Light Non-Aqueous Phase Liquid) is present at the upstream side of the fence, at least near filter 1. Without the values of drain A, the correlation coefficient (R^2) between simulated and measured values is 0.93.

Fence 3

At a first order degradation rate of about 0.6 d^{-1} (corresponding with a half life of 1.15 day), the measured concentration development can be simulated quite good, except for the concentrations in filters 1-4-7. The relatively high (average) concentration at the filters 1-4-7 can be partly explained by a new input of polluted groundwater in filter 7: from October to November the oil concentration increased from 1200 µg/l to 4200 µg/l. Furthermore, it seems biological degradation is not proceeding here, possibly because this region is not reached by the injected air. To a less extent, the same goes for the filters 3-6-9: the (limited) decrease of mineral oil concentration here is probably mainly due to biological degradation in the upstream zone between drain A and B. Without the values of filters 1-4-7, the correlation coefficient (R^2) between simulated and measured values is 0.76.

By using the results of the stop test (May - June 1999), it might be possible to calculate a maximum rate of biological degradation, assuming that all the vanishing oxygen is used by the biological degradation of mineral oil. Calculations for this were executed for drain 1A and drain 3A. The results are not presented here, because of the unreasonably high degradation rates calculated, which do not contribute to the understanding of the biological degradation of mineral oil. The measured disappearance of oxygen is caused by a lot of totally different mechanisms, of which biological degradation (not only of mineral oil), chemical oxidation and evaporation. The contribution of these various processes can not be estimated from the results of a stop test in the field.

Evaluation

From the modelling it is concluded that it is likely to suppose that biological degradation is proceeding in all fences. The first order degradation rate (k) has values between 0.5 d^{-1} (fence 2) and 0.65 d^{-1} (fence 1). The advantage of using a first order rate here, is that the value of it is concentration independent. Whereas the initial hydrocarbon concentrations in the fences differ strongly from one another, the rate of degradation is very similar. From this, we can conclude that the operation of the fences is very similar to one another. This is remarkable, because they differ in filling material and type of injection system.

In the column tests TNO-MEP determined a mineral oil (C6 - C16) removal of 55 % in 35 hours in the 0 % gravel column up to 98 % in 35 hours in the 90 % gravel column [Van Liere et al., 1998]. From this we can determine a first order degradation of 0.55 to 2.68 d^{-1}, by using the formula $C_t = C_0 \cdot e^{-kt}$ for first order decay. It is striking that the fitted values of the biological degradation rate in the fences are about the same as the ones determined in the laboratory for a 0 % gravel column. Moreover, in literature these values are quite common for (enhanced) aerobic decay of mineral oil. This confirms the reliability of the fitted values.

The measured and simulated rates of mineral oil degradation in the fences, make sure that we are dealing with aerobic biodegradation: anaerobic biodegradation of mineral oil can not be this fast. In this perspective, it is striking that we did not measure a structural increase of oxygen. In the world of water treatment this phenomena is known: aerobic degradation takes place whereas oxygen is not measured in concentrations above 1 mg/l.

A final indication for aerobic biological decay is contributed by the measurements of the soil air. Carbon dioxide, oxygen as well as total hydrocarbons are measured in the gas phase of the unsaturated zone. All the results are plotted in figure 4. From this figure it seems obvious that there is a moderate relation between total hydrocarbons and oxygen, and between total hydrocarbons and carbon dioxide on the other hand: the more hydrocarbons are present, the more carbon dioxide and the less oxygen is present.

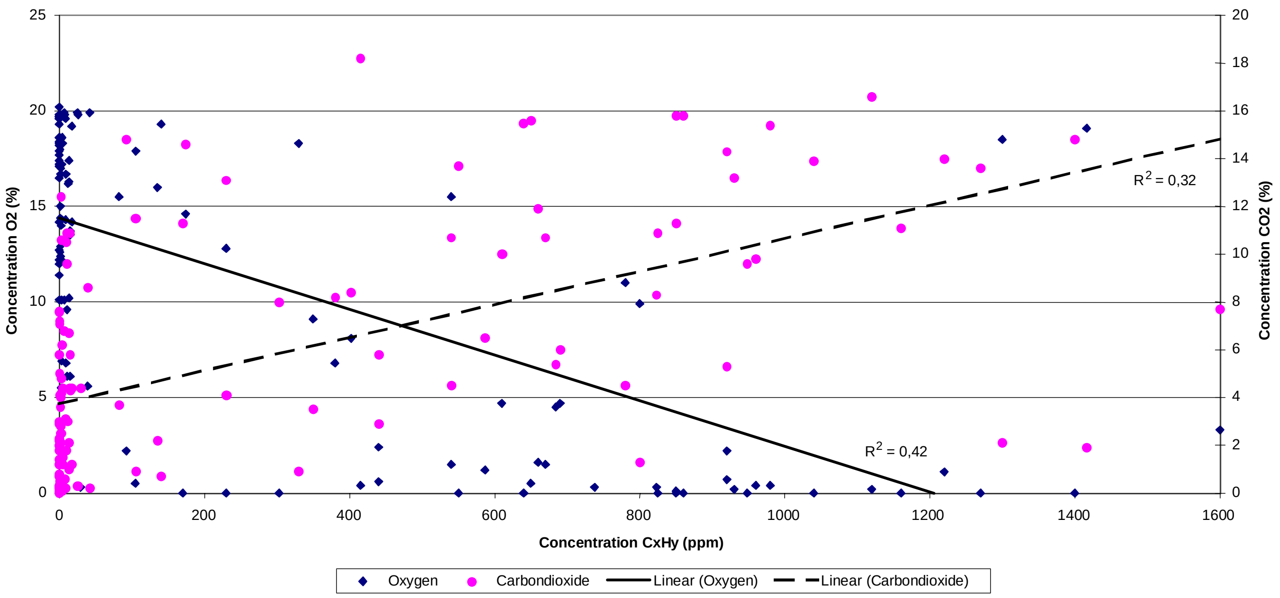


Fig. 4. Hydrocarbons, oxygen and carbon dioxide in unsaturated zone; biological fences 1, 2 and 3.

5.1.4 *Conclusion on performance*

From the modelling it is concluded that it is likely to suppose that biological degradation is proceeding in all fences. The rate of degradation has been determined and it seems to be a reasonable rate for aerobic degradation. The only question remaining, of the questions asked at the end of section 5.1.1, is whether the biological fences are capable of reducing pollutants to such levels that they cause no unacceptable levels of pollutants outside the site boundaries. This question can be answered by simulating the future development of the concentrations throughout the fences. For this, we used the models of section 5.1.3, starting at December 1998 until the effluent reaches hydrocarbon concentrations below the intervention criterion (I-value). The results are shown in figure 5, 6 and 7.

The time necessary for reaching the intervention criterion differs for each fence. It is dependent on the initial pollution present in the fences soil matrix as well as on the supply (input) of 'new' mineral oil. At pilot 3 it will take about 1 to 1½ years to get an effluent concentration below I-value. This fence was initially the least polluted one with lowest influent concentrations. At pilot 2 it will take about 3 years to get an effluent concentration below I-value. At the initially most polluted pilot 1, which also gets the highest input concentrations, it takes longest before an effluent concentration below I-value is reached: about 6 to 8 years.

So, as we now see it, the biological fences are capable of reducing pollutants to concentrations below the intervention criterion outside the site boundaries. When the local soil matrix where the fences are placed is non-polluted, a biological fence with the dimensions of the ones used in this study is 'operational' immediately. Because in this study the local soil matrix is initially polluted, it takes some time before the fence actually does what it was made for: transmitting groundwater with concentrations below acceptable levels.

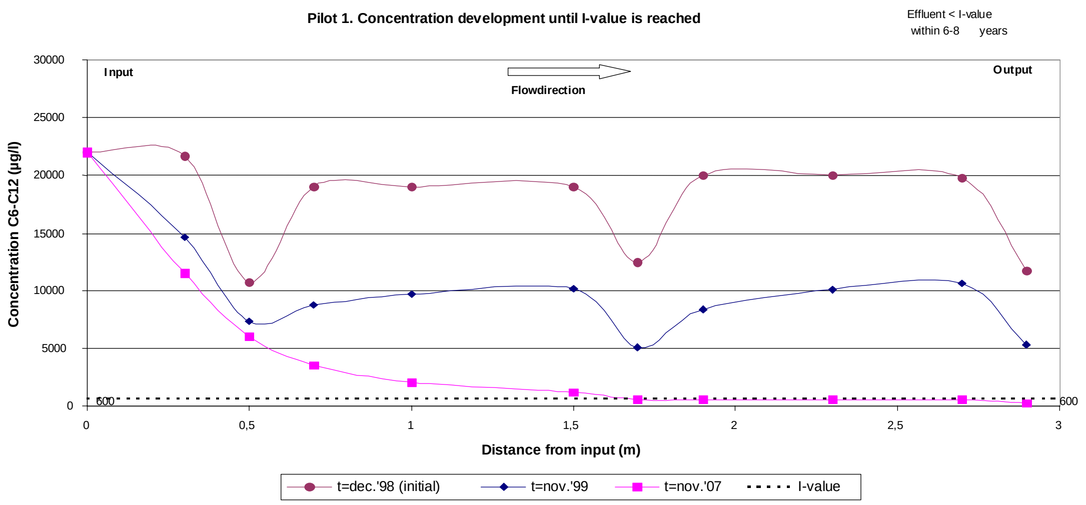
5.2 **Optimizing oxygen supply**

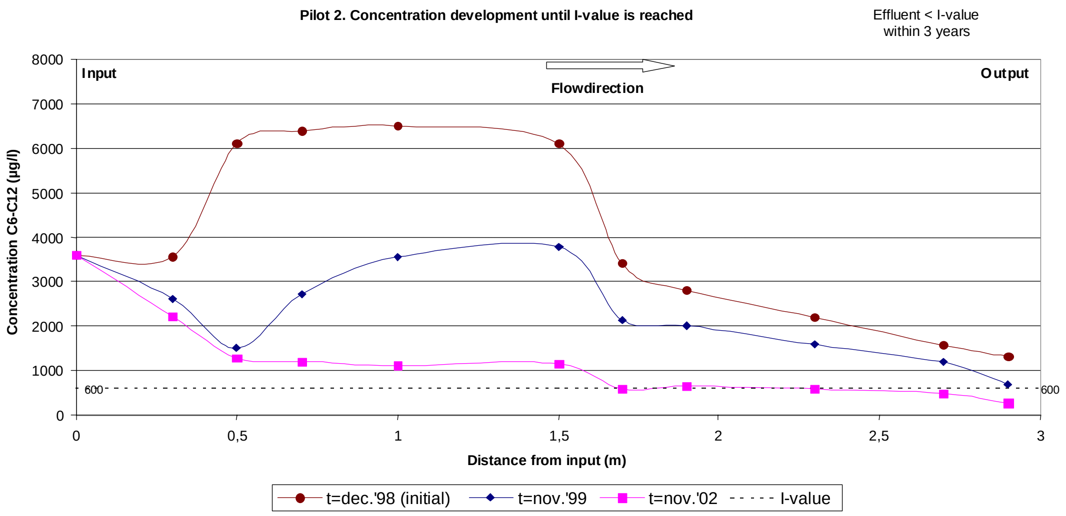
In appendix G and H an overview is given of oxygen and redox potential measurements from December 1998 until November 1999. The results at the final injection regime do not differ much from previous measurements at different injection regimes: the figures show a large variation in place and in time, even at a constant flow regime (August 1999 - November 1999). The oxygen levels in fence 2 and fence 3 are considered alike. The oxygen levels in the upstream measuring drain of fence 1 are considerably higher (> 5 mg/l).

Over time, the little oxygen measured seems to be distributed throughout the fences. However, from the zero measurements until now, no structural increase of oxygen level was measured. Although the redox potential has increased in general, it still indicates anoxic conditions in the soil matrix. Controlling the process needs knowledge of the process, so several possibilities for the apparent lack of oxygen were investigated. The results are summarized below.

Field investigation

Tests to check the equipment did not show any leakage or shortcut flows and it was concluded that the pumped air reached the soil (see appendix I). Tracer tests with helium indicated that at fence 2, 67 % of the injected air was transported vertically to the surface of the fence. At fence 1 and 3, respectively 4.1 % and 0.024 % of the injected air reached the fences surface and therefore by all appearances the air is transported horizontally to the harbour. For a spatial survey of the helium recovery we refer to appendix J. Near fence 3 at high tide air bubbles were seen on the harbour and near fence 1 at extremely high tide air bubbles were seen too. Supplementary soil profiles (see appendix L) indicated that small clay layers are present at the fences 1 and 3. These clay layers explain the horizontal transport of air at these fences.





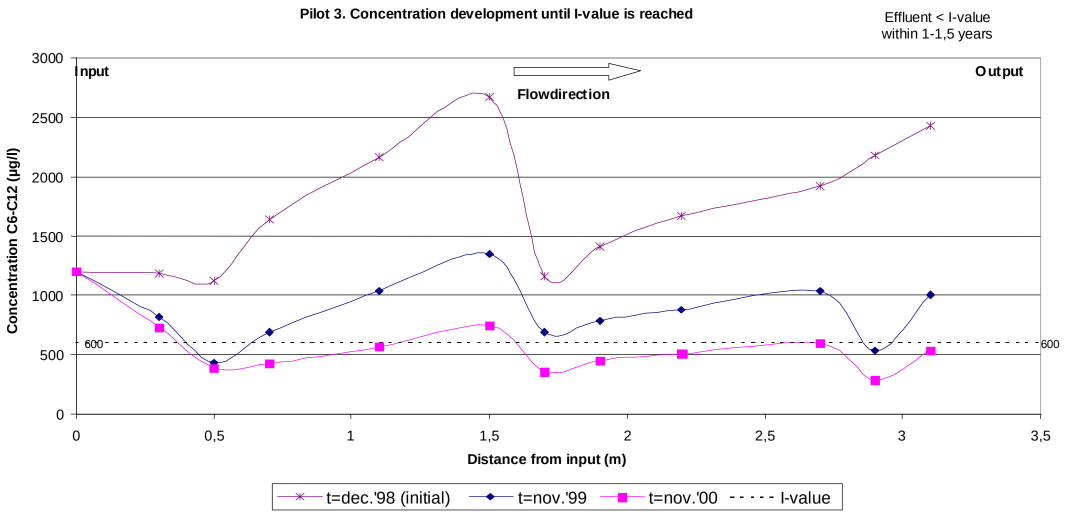


Fig. 5 - 7. Results model simulations of concentration development in all fences in the future until the intervention value is reached .

From research by Elder and Benson [1999] we also know that injected air in finer media of about 0.2 mm and smaller (like fine to medium sand), is transported nearly horizontally by way of air channels through the soil. Implicitly, this should mean that the exchange of oxygen from air to water is more efficient because of a longer residence time before emergence at the surface.

Probably at fence 2 clay layers were present as well, but these are disturbed during the construction of the fence with the drain machine and the replacement of the original soil by gravel. The gravel ditch at fence 2 can be considered as 'one preferential flow path'. According to research by Brooks et al. [1999] grain diameters of about 2 mm and above, shows air bubbles when sparged. In fence 2 gravel with a diameter of 4 - 16 mm is used. Air bubbles in gravel are moving relatively fast to the fences surface because of the upward pressure. From this it is not strange that at fence 2 the recovery at the tracer tests is higher.

Desk study on oxygen demand

Calculations on the reduction capacity of the soil were performed to check if the amount of injected air would reach the oxygen demand. The processes that were taken into account are:

- oxygen transfer efficiency from air to water;
- biological decay of mineral oil compounds in groundwater;
- biological/chemical decay of (dissolved) organic carbon in ground and groundwater (DOC);
- chemical oxidation of (reduced) minerals in ground and groundwater.

With the assumption that the oxygen demand of groundwater will be met first (relatively fast process) and only a surplus of oxygen in groundwater would be available for the demand of the soil (relatively slow process), it was concluded that enough oxygen (air) was injected. For further assumptions that were made and quantitative input and results of these calculations we refer to appendix K.

Evaluation

However, a structural raise of oxygen was not measured in the fences. Taking into consideration that at similar flow rates at the start tests an increase of oxygen level was measured, the most probable explanation for the apparent lack of oxygen in the groundwater of fences 1 and 3 is that the exchange of oxygen is transient and evolves in two phases:

1. after increasing air flow, the distribution of air through air channels initially is quite homogeneous and oxygen is measured throughout the fences;
2. after a while of constant injection, a few preferential flow paths are formed, air distribution has become heterogeneous and oxygen is measured at a few spots only.

So in steady state, during the operational phase of continuous air injection, only locally air and oxygen are present in the groundwater at fences 1 and 3. During the start/stop tests oxygen concentrations higher than 1 mg/l were measured in the measuring drains because the flow rates were varied relatively fast. The steady state is likely to be influenced by the pressure conditions in soil. When, for instance, hydrostatic pressure changes (showing a raise or fall of groundwater level), the location of the preferential flow paths might be changed, and with it the distribution of oxygen.

At fence 2 the cause for the apparent lack of oxygen is a different one. From the shape of bubbles we know that the surface is smallest in relation to the volume. So oxygen trapped in air bubbles is exchanged less efficient than oxygen in air channels. Moreover, because of a relatively short residence time of air before emergence at the surface of the fence, the transfer of oxygen to groundwater is even less efficient.

Resuming:

- In fine to medium sand (fences 1 and 3) injected air is nearly horizontally moved through air channels, which are heterogeneously present at a constant injection regime. Oxygen is transferred quite good from air to water, but is only locally present because of the wide spread air channels (preferential flow paths).
- In a coarse medium like gravel (fence 2) injected air is vertically moved by air bubbles quite homogeneously. The oxygen transfer is less efficient, because of the shape of air bubbles and the relatively short residence time before emergence at the fences surface.

Conclusions on optimizing the oxygen supply

The conclusions on optimizing the oxygen supply are:

- no structural increase of oxygen level in the groundwater was measured. Over time, the little oxygen which was measured seemed to be distributed throughout the fences;
- redox potential measurements showed that the soil matrix stayed anoxic, although less reduced than initially present: an increase of the redox potential is measured;
- air transport and oxygen transfer from air to groundwater are sensitive to soil heterogeneity and pore size distribution;
- it is still unknown to what extent oxygen transfer takes place from air to groundwater;
- it is not clear how much oxygen is available for biological decay of hydrocarbons.

5.3 Technical comparison of the three tested fences

5.3.1 *Construction*

Fence 1

The construction of the normal (dig) drain was not easy. It was difficult to remove the casing without pulling out the whole drain, due to the resistance of the soil. The difficulties will increase with the length of the drain. The construction of the no-dig drain was not attended by serious problems. This is also expected with an increasing length of the drain.

Fence 2

The construction of fence 2 involved serious problems. The presence of underground infrastructure (pipes, cables) was causing the problems during the installation of the drains. Furthermore the existing space at this location for the drainage machine was very limited. For this reason only a small drainage machine could be used. Both problems, underground infrastructure and limited space, are expected to be present at most existing industrial locations, which implies that the application of this type of construction of (biological) fences is limited to only a few existing locations, new and abandoned industrial areas.

Fence 3

The construction of fence 3 succeeded without any problems.

Two major advantages at the construction of a biological fence with vertical filters are:

- vertical filters can be placed at different depths;
- the intensity of the filters can vary in space.

Differences in depth and varying intensity in space are not possible with horizontal drains. Furthermore, the reliability of the installation (exact depth and place) of vertical filters is higher compared to the installation of horizontal drains.

5.3.2 *Maintenance*

During the test period of the pilot no maintenance problems occurred for the three different fences. The problems with the air injection of one of the horizontal drains of fence 1 (drain 1B) were caused by installation problems. For full-scale fences which have to operate for a long period of time, serious maintenance problems for the horizontal drains (fence 1 and 2) can arise if malfunctioning (e.g. clogging) occurs in the drains (the underground part of construction). Malfunctioning of the injection drain will have an impact on the whole fence. Malfunctioning of the vertical filters is in general much easier to solve and this will only have an impact on a small part of the fence.

Maintenance of the above ground air injection installation, which includes the injection unit (compressor), the piping and the valves, is expected to be more or less similar for the different fences. However, fence 3 contains more piping and valves to maintain.

5.3.3 *Performance of operation*

Based on the oxygen measurements and the helium tracer test it could be concluded that fence 1 and fence 2 were aerated over the whole length. On the other hand small variations in the depth of the installed drain can cause preferential flow of air by channelling. This problem will increase with an increase of the length of the drain.

Furthermore, it is unknown if it is possible to have an equal air distribution over the whole drain, when drains are used with a much larger length than the tested length of 30 m. Equal distribution of air with the use of vertical filters can be controlled, also with an increasing length of the fence.

To ensure an equal air distribution over the whole length of the drain, it is necessary always to have a high injection flow rate for fence 1 and 2 (relatively with respect to the oxygen demand). At lower air injection flow rates an equal distribution over the whole length of the drain, especially at the end of the drain, is not ensured. Lower air injection rates at fence 3, vertical filters, don't have an impact on the distribution of the injected air.

5.3.4 *Flexibility*

The flexibility of a fence with vertical filters is high. The aeration rate can be controlled at different levels at different places in the fence. This implies that extra air can be injected at different parts of the fence to overcome differences in air entrance in the soil and/or at parts with high oxygen consumption.

Furthermore, it is easy to extend the amount of injection points or to decrease (shut down) the amount of injection points during the time period of the performance of the fence.

The flexibility of fences with horizontal injection drains is low. Even a change of the injection flow rate can have an impact on the equal distribution of the injected air over the whole length of the drain.

5.3.5 *Conclusions*

The results of the technical comparison are summarized in table 5.

Table 5. Technical comparison.

type of fence	horizontal drain: drilling machine (fence 1)	horizontal drain: drainage machine (fence 2)	vertical filters (fence 3)
construction	+	--	+
maintenance	+\-	+\-	+
performance	+\-	+\-	+
flexibility	-	-	+

explanation:

--	badly
-	moderately
+\-	sufficient
+	good

Based on the technical comparison vertical filters (fence 3) are most preferable to construct and operate in a full-scale aerated biological fence.

CHAPTER 6

EVALUATION BIOLOGICAL FENCES VERSUS CONVENTIONAL FENCES

6.1 **Costs**

The actual length of the three test fences was 30 m. To compare the total investment and exploitation costs of the fences with conventional techniques, the total costs have been estimated for three 400 m long biological fences. The calculations are shown in appendix M. The results of the calculations for these three biological fences are present in table 6. The costs for fence 2 include the considerable costs for treating the contaminated soil from the trench. For Shell Pernis the cost estimates of three actual conventional systems were used. The costs for operation and replacement were estimated. The calculations for the conventional systems are shown in appendix M. The total costs of these systems are presented in table 7. Various assumptions had to be made to get to these estimations. We stress that the results should be used for this comparison only.

The costs consist of investment (initial), operational (annual) and replacement (once every ten years) costs. For both the biological fences and the conventional systems the assumption was made that the fences will be operated for a period of 50 years. The operational and replacement costs are capitalized at a interest rate of 4 % (capitalization factor of 21.48 and 1.798 respectively).

Table 6. Cost estimates of biological fences (in DFL, excl. VAT).

alternative	length	investment costs (per m)	exploitation costs (per m for 50 years)	replacement costs (per m for 50 years)	total costs (per m for 50 years)
pilot 1, drilling	400	800	2000	1400	4200
pilot 2, 90 % gravel drain machine	400	1400	2000	2500	5900
pilot 3, vertical filters	400	500	2000	1000	3500

For the conventional systems two situations are distinguished:

- a 'stand alone' Pump and Treat system (P&T SA) (see table 7);
- a groundwater extraction system (Pump) and treatment of the water at a central water treatment installation (P SA & CT), as is the situation at Shell Pernis (see table 8).

The costs for exploitation of the system (pumps, maintenance) are approximate estimates. The cost estimates for treatment of the water are based on the assumption that, for a pump and treat system with a length of approximately 400 m, about 2 m^3 per hour will be extracted.

Table 7. Costs of 'stand alone' Pump and Treat systems (in DFL, excl. VAT).

system	length	investment costs (per m)	exploitation costs (per m for 50 years)	replacement costs (per m for 50 years)	total costs (per m for 50 years)
1. horizontal drains, excavating	310	900	2800	1700	5400
2. horizontal drains, excavating	190	700	4600	1200	6500
3. vertical system	210	800	4100	1500	6400

Table 8. Costs of 'stand alone' Pump and Central Treatment systems (in DFL, excl. VAT).

system	length	investment costs	exploitation costs	replacement costs	total costs (per m for 1 year)
1. horizontal drains, excavating	310	900	1700	1700	4300
2. horizontal drains, excavating	190	700	2400	1200	4300
3. vertical system	210	800	2200	1500	4500

It can be seen that a biological fence is comparable to a conventional system (except for fence 2), like is done at the site of Shell Pernis. It is expected, that a proven design for a biological fence will be even cheaper than is estimated now. When all exploitation costs and treatment costs are taken into account, blowing of air into the system is expected to be considerably cheaper then pumping and treating water.

The costs for monitoring the various biological fences do not depend on the used system and are more or less the same for each fence. The costs of monitoring of conventional systems are not estimated. The costs are expected to be comparable.

6.2 **Environmental merits**

By comparing remedial options costs only is not the decisive factor. Beside costs the positive and negative environmental influences are important to decide which option will be the best to use. In this case a comparison between biological fences and conventional techniques in general is made. The inventory of environmental influences is made by using the designed REC-method. A comparison between the environmental influences of biological fences and conventional techniques are included in table 9.

Table 9. Environmental influences of biological fences and conventional techniques.

environmental influences		biological fences	conventional techniques
negative			
use of natural resources	use of (clean) groundwater	none	yes
energy	injection of air	yes	none
	pumping groundwater	none	yes
	water treatment		
use of space	water treatment installation	none	yes
chemicals	use of chemicals	none	none
	final residues	none	yes
positive			
soil and groundwater quality	prevention of future soil and groundwater contamination	improve	no improvement

By interpreting table 9 it can be concluded that in general the negative environmental influences by using a biological fence are less than by using conventional techniques. In case of using conventional techniques the use of clean groundwater, the pumping of groundwater, the implementation of a water treatment installation and the final residues are negative environmental influences. By using a biological fence these influences are not relevant. The negative environmental influence for this kind of remediation technique is the need of energy to inject air.

6.3 **Risks**

A failing conventional system will almost immediately cause a flow of contaminated groundwater over the borderline, because no buffering capacity of contamination in the soil is present. By using a biological fence the soil of the fence is cleaned and a buffering capacity for contamination is created. When this system is failing, the contamination will be buffered as long as there is a capacity to buffer present. Together with the residence time of groundwater in the fence, this gives time to restore the fences operation.

The biological process of degradation of hydrocarbon can in principle vary from place tot place along a fence. The effects are assessed by measuring concentration of process parameters or contaminants, which level can in principle vary again from place to place along a fence. This hampers the reliability of the fences. Conventional system are monitored by measurement of hydraulic head. Hydraulic head is influenced by its surrounding and therefore more representative for a larger part of the fence. This improves the reliability of the conventional fences.

CHAPTER 7

GUIDELINE FOR DESIGN AND MONITORING OF A BIOLOGICAL FENCE

7.1 **General**

Objective of a biological fence is to stimulate aerobic biodegradation of hydrocarbons in the saturated zone. Designing a fence is to make two ends meet: the oxygen demand at a specific site and oxygen supply by a specific installation.

With preliminary investigation information about site specific condition should be gathered. The total oxygen demand is ruled by biological and chemical processes in the saturated zone. Supply of oxygen is ruled by physical processes: air flow in saturated porous media and kinetics of oxygen transfer from air to groundwater. Designing a biological fence is combining general knowledge of the processes with the information of the conditions at the specific site.

In the next paragraph the design and the preliminary investigation are described.

7.2 **Design (including preliminary investigations)**

Objective of a fence is to prevent migration of hydrocarbons with groundwater by aerobic biological degradation.

This means that information is needed on the following subjects:

- the hydrocarbon content in ground and groundwater;
- the migration of hydrocarbons with groundwater in the saturated zone;
- the aerobic degradability of hydrocarbons;
- the initial redox conditions (and oxygen level) in the saturated zone.

The information needs to be combined. In the combining lies the designing. From this study follows that combined modelling of hydrology, solute transport and degradation, is a powerful instrument to define the boundary conditions for a design of a biological fence.

Hydrocarbon content
Hydrocarbons are a large group of components with each specific characteristics. It is necessary to know which components are present in ground and groundwater, upstream and in the fence and at which concentrations. This study shows that higher initial concentrations will result in a longer period before the output of the fence reaches acceptable levels. As a biological fence is aimed at controlling contaminant plume migration, the concentrations will be relatively low.

Migration of hydrocarbons
This is a result of the transport of dissolved hydrocarbons with groundwater flow at one hand and sorption of hydrocarbons at the other hand.

Groundwater flow
Groundwater flow can be determined by a tracer test. Tracer tests are time and money consuming. The conventional method is preferred: the groundwater flow is determined by the hydraulic conductivity, the hydraulic gradient and porosity. Various tests are available to obtain hydraulic conductivity and porosity. The hydraulic gradient can be determined by measurement of hydraulic head.

Sorption

Sorption is controlled by component characteristics and the fraction of organic matter in the saturated zone. The characteristics of specific components can be found in literature. Organic matter content is site specific and can be determined by sampling and analysis.

Degradability

It is generally accepted that hydrocarbons are degradable at aerobic metabolism(s). In literature the aerobic degradation rates in subsoils are given in a range of 0.01 - 2.5 d^{-1}. This study showed at three fences a degradation rate of 0.5 - 0.65 d^{-1}.

When the degradibility of a specific component is doubtful laboratory test are advised to asses this. Degradation rates achieved at laboratory are indicative and can not be translated simply to field conditions.

Conditions for aerobic degradation

At aerobic metabolisms oxygen is used as electron acceptor. To make aerobic degradation possible oxygen is needed. Other processes can use oxygen as well. Generally the redox conditions are oxic (positive redox potential) when at equilibrium oxygen is present. From this point of view it is necessary to have information about:

- the present redox conditions and oxygen demand (reduction capacity);
- the possibilities to meet this oxygen demand by oxygen supply.

However, this study among others show that it does not mean that aerobic degradation is impossible when no oxygen can be measured and redox conditions are anoxic. The supplied oxygen is probably used very quickly by aerobic degradation

Oxygen supply

At phase 1 [CUR/NOBIS, 1999] it was concluded that oxygen should be supplied by air sparging. Sparging of pure oxygen or supply of hydrogen peroxide needs complex systems, is more expensive, especially on the long run and might be harmful for micro-organisms.

Oxygen supply by injection of air will be influenced by air injection rate, the distribution of air flow through the saturated zone and by the efficiency of oxygen transfer from the air to groundwater.

Air flow and oxygen transfer are controlled by soil profile and soil texture. This study confirmed that transport of air is very sensitive to the (vertical) heterogeneity on micro scale of the soil. The sensitivity of air transport for (vertical) heterogeneity, shows that micro scale (mm) is at least as important as macro scale heterogeneity (dm). Heterogeneity causes preferential flow paths. Although heterogeneity can have a positive effect on the radius of influence of injection systems and residence time of air in the subsoil, air flow is uncontrollable and therefore heterogeneity is regarded as unfavourable.

Preliminary investigations should study soil profile on micro scale. As the Edelman device disturbs the soil on micro scale it needs an expert to interpret the soil profile. Drilling techniques that are able to take undisturbed soil samples are preferred. This study did not investigate the required density of soil drilling, but it is estimated that one drilling to at most every 5 - 10 m is needed.

At the choice of injection technique one can anticipate on soil heterogeneity. From the low recovery of helium at tracer test in this study, it can be concluded that the horizontal drilled drain and vertical injection filter leave the soil relatively undisturbed. At the horizontal drain installed with a drain machine the recovery of helium was much higher as the soil profile was homogenized. That this high recovery did not result in high(er) oxygen levels is thought to be caused by the texture of the gravel with which the trench was filled.

Soil texture determines whether air is transport in bubbles or through channels. The transfer of oxygen from air to groundwater is influenced by the surface of the groundwater to air interface. The interface is influenced by the form of air transport: literature indicates that at small pores (< 1 à 2 mm diameter) air channels develop and at large pore diameters (> 1 à 2 mm) the air is transported in bubbles. There are indications [Elder and Benson, 1999] that the size of air channels are again subject of pore diameter. The air to water interface again depends on the size of air channels and bubbles. So pore size (distribution) should be subject of preliminary investigation. A literature study on the latest insights on the relation between pore size and air transport and oxygen transfer is advised.

The results of this study correspond with literature on the apparent positive effect of intermittent injection on oxygen supply, as air flow is redistributed.

Oxygen demand

Biological oxygen demand

Biological oxygen demand is ruled by the amount of aerobic degradable components that is available in the fence or will supplied by groundwater flow. Aerobic degradable components are the hydrocarbons and organic matter. Every soil contains organic matter to a certain extent.

Only a part of this organic matter can be degraded under aerobic conditions, the rest will be inert. The amount of aerobic degradable organic matter is site specific.

Chemical oxygen demand

Chemical oxygen demand is ruled by the specific constituents (like reduced iron, humic acids etc.) of the soil and groundwater at the specific site.

As the total oxygen demand is very site specific, preliminary investigations should contain one or more tests to determine oxygen demand for groundwater and ground for the specific site. In phase 1 of this study TNO/MEP [Van Liere er al., 1998] performed micro-oxymax experiments to determine the total oxygen demand for the site of Shell Pernis.

In this study also calculations were performed to approximate biological and chemical oxygen demand of soil and groundwater. The calculation are shown in appendix K. These calculations show the potential total oxygen demand and do not consider kinetics of the various processes nor the interference of the processes. In this study it is assumed that the oxygen demand of groundwater will be met first (relatively fast process) and only a surplus of oxygen demand in groundwater would be available to meet the oxygen demand of the soil (relatively slow process).

Design

Combined modelling of groundwater flow, solute transport and degradation in the saturated zone, as is performed in phase 1 [CUR/NOBIS, 1999], is a powerful instrument to put up the conditions for the design of a biological fence. It brings together the information from the preliminary investigations about the various processes: Hydrocarbon content of groundwater and degradation rate lead to the required residence time. Residence time in combination with migration velocity will lead to the required with of the fence.

However, many existing models are based on the assumption that oxygen levels are not limiting degradation. Models that do consider oxygen levels would (probably) have a hard case accounting for the decrease of hydrocarbons by degradation as little oxygen was measured at these fences.

Modelling could be improved when the transfer of oxygen from air to groundwater is better accounted for. The implementation of biological fences for aerobic degradation of hydrocarbons would be improved when more knowledge is gained about transport of air and transfer of oxygen from air to groundwater.

7.3 Monitoring

The questions are what, where and how to monitor while operating a biological fence.

7.3.1 *What*

Objective of the monitoring is to get insight in what the performance of the fence is and how this performance can be controlled/optimized.

The performance of a fence is the reduction of the input hydrocarbon concentration to acceptable output concentrations. This reduction can be determined by measuring input and output hydrocarbon concentrations.

As the reduction of concentrations by biodegradation is a slow process, this monitoring can be considered as long-term monitoring. Efficient operation of the fence needs monitoring on a much shorter term.

The process of aerobic biodegradation of hydrocarbons uses oxygen. Oxygen level (and the corresponding positive redox potential) seems therefore a logical parameter to monitor. However, it has to be taken into account that, as this study shows, biodegradation can occur while hardly any oxygen can be measured and redox potential is negative and only slowly increasing. An alternative is to monitor the processes of oxygen supply by air injection, air transport and oxygen transfer.

Air injection

To make sure that air is pumped and reaches the subsoil.

- operation of pumps;
- air tightness of the installation.

Air transport oxygen transfer

A tracer test is useful to get insight in the distribution of air flow. In this study helium is used as tracer for air flow. Helium is inert and does not dissolve in groundwater (like nitrogen). Sulfur hexafluoride (SF6) is a tracer with similar properties to those of oxygen (except for consumption). As a consequence the distribution of SF6 tracer can be seen as an analogue for oxygen distribution.

7.3.2 *Where*

Again performance of the fence and controlling the process can be distinguished. As performance of a fence is determined by measuring input and output concentration of hydrocarbons, these measurements have to be carried out upstream and downstream of the fence. Controlling the process needs to be measured in or at the fence.

This study did not show any principle advantage of measuring by drain or by standpipes (see appendix N). Measurement by standpipes is more flexible as the are more easily installed.

7.3.3 *How*

Again performance of the fence and controlling the process can be distinguished. As biological degradation is slow process continues measurement is not very useful. Periodical measurement of hydrocarbon will suffice. Measurement of hydrocarbons can be done by sampling and analysis or on site with a sensor.

When oxygen level or redox potential are measured as process parameters continues measurements are preferable. This study showed that air transport through the saturated zone is dynamic, resulting in dynamic oxygen levels.

For measurements on air injection, air transport and oxygen transfer we refer to appendix I.

CHAPTER 8

CONCLUSIONS

8.1 Evaluation of the three tested biological fences

The three different fences, constructed and operated during the pilot test, are compared on hydrocarbon degradation, oxygen influx, optimal conditions, construction, maintenance, performance of operation, flexibility and costs.

The overall conclusion on the applicability of a biological fences is that when certain conditions are met (see chapter 7) a biological fence can prevent spreading of hydrocarbons.

Hydrocarbon degradation
Overall, all fences accomplish a decrease of hydrocarbon concentrations in groundwater. The decrease is seen for volitile and non-volatile components, indicating that volatilization is not the (major) explanation for the decrease. Whereas the initial hydrocarbon concentrations in the fences differ strongly from one another, the first order rate of degradation in the 3 fences is very similar and has values between 0.5 and 0.65 d^{-1}. From this, we can conclude that the operation of the fences is very similar to one another. This is remarkable, because the fences differ from one another in filling material and type of injection system.

The determined degradation rate in the fences is about the same as the biological degradation rate determined in the laboratory under aerobic conditions. The relatively high degradation rates of mineral oil in the fences, indicate that we are dealing with aerobic biodegradation: anaerobic biodegradation of mineral oil can not be this fast. A final indication for aerobic biological decay are the measurements of the soil air: the more hydrocarbons present, the more carbon dioxide and the less oxygen is present in soil air. From this all, we conclude that the decreasing concentrations are due to aerobic biological degradation.

In general, the acceptable levels at the harbour side of the fences are not reached yet. This is caused by the high levels of pollution initially present in the fences matrix: this initial amount of (mainly sorbed) hydrocarbons in the fences has to be dealt with first. That does not alter the conclusion that the degradation rate is that large, that in general a biological fence can prevent spreading of hydrocarbons.

Optimizing conditions
From the zero measurements until November 1999, no structural increase of oxygen level was measured. This was the case in all the fences. The oxygen content is probably influenced by:

a. the oxygen transfer from air to groundwater;
b. the transport of air (preferential flow paths);
c. the kinetics of oxygen demand;
d. the reduction capacity of the soil.

In all fences, a structural but slow increase of the redox potential is observed, but it still got negative values.

Based on the measured conditions (oxygen content and redox potential) it is not yet possible to compare the fences.

Construction, maintenance, performance of operation, flexibility and costs
The construction of fence 3, consisting of twenty vertical injection filters, installed by pulsing succeeded without any problems. The construction of fence 2, consisting of two horizontal injection drains in a gravel ditch, installed by the use of a drainage machine, went with most problems.

During the test period of the pilot no maintenance problems occurred for the three different fences. Nonetheless, malfunctioning of a long drain (fence 1 and 2) will have more impact than malfunctioning of a single vertical filter (fence 3).

To ensure an equal air distribution over the whole length of the drain, it is necessary always to have a high injection flow rate for fences 1 and 2. Equal distribution of air with the use vertical filters (fence 3) can be controlled, even at a low injection rate.

The flexibility of a fence with vertical filters is highest (fence 3): the injection filters can be controlled independently of each other.

For the tested site, the construction of fence 3 is the cheapest option, where the construction of fence 2 is the most expensive.

The conclusion is, that injection by vertical filters (fence 3) is the most cost-effective technique to construct and operate a full-scale aerated biological fence.

8.2 **Evaluation biological fences versus conventional fences**

The implementation and exploitation costs of a biological fence like pilot 3, is comparable to conventional systems (Pump and Treat). However, when no water treatment system is available on the site and the water treatment costs are taken into account, a biological fence based on air injection is expected to be considerably cheaper then a conventional pump and treat system.

Besides that, in general the negative environmental influences (energy, space, chemicals) by using a biological fence are less than by using conventional techniques.

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APPENDIX A

OVERVIEW LOCATION PILOTS AND PILOT SYSTEMS

Fig. A2. Cross-section of the pilots.

APPENDIX B

OVERVIEW MONITORING SYSTEMS

Fig. B1. Overview and codes measuring drains.

Fig. B2. Overview and codes monitoring wells.

Fig. B3. Overview and codes air measurements.

APPENDIX C

RESULTS FOC-SENSOR MEASUREMENTS

The hydrocarbon content was continuously monitored by Fibre Optic Sensors, in the measuring drains as well as in the monitoring filters. The sensors are coupled to two data loggers, from which data were downloaded at least once a month. During the monitoring some of the sensors were getting out of order. These sensors (1-A, 2-A, 2-B, 2-4) have been replaced at August 9th 1999 by IWACO and FCI.

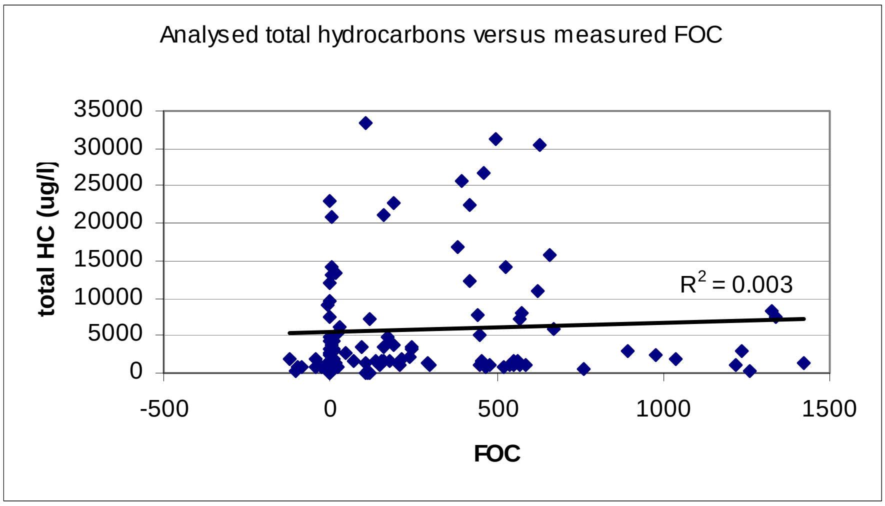
For the measurements of the FOC-sensors from December 1998 until November 1999 we refer to the added (coloured) graphs.

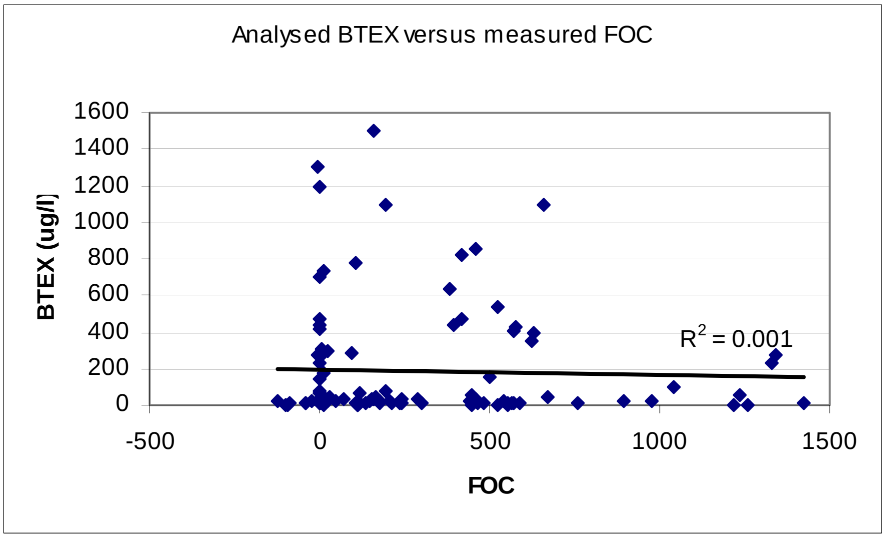
The picture we get from the sensor measurements is different from that of the chemical analysis. The three types of trends, increasing, decreasing and steady concentrations, are equally present. The points which really show consistent decrease (1-5, 1-11, 1-12, 3-6, 3-10 and 3-A), are also decreasing according to the chemical analysed groundwater samples in drains and filters. The rest of the detected concentrations (steady or increasing) are not corresponding with the results of the chemical analysis. The detected increase in concentration at point 1-B and 2-12 are remarkable. These ones are, without doubt, decreasing according to the chemical analysis. To a less extent, the same goes for the detected increase at points 3-2, 3-11 and 3-12.

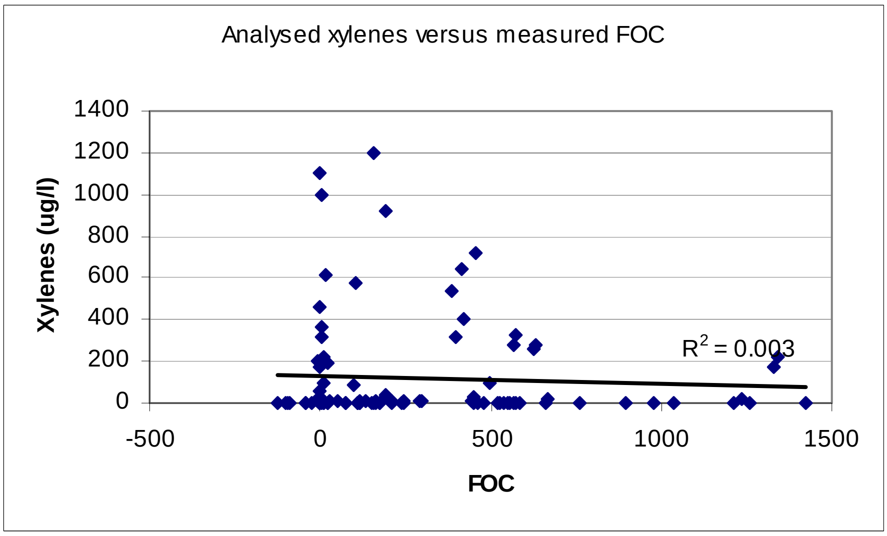
We tried to find a relation between the absolute values of the FOC-measurements and the chemically analysed concentrations. We refer to the added (XY scatter) figures. This relation is not present for total hydrocarbons, BTEX nor xylenes. If we first transpose (normalize) the absolute values to values relative to the values of December 1998, we find a very slight relationship between FOC-measurements and chemically analysed concentrations (see figures). The relationship is strongest for xylenes (correlation coefficient $R^2 = 0.33$).

The difference of trends between analysed and detected concentrations may be caused by the non-specific measurement by the sensors of total hydrocarbons. The sensors are calibrated for xylenes (that's why the discussed relationship of normalized values for this component is strongest). Possibly they are less sensitive for mineral oil compounds like alkanes. From field experience, we now know the sensors are also sensitive to frost or other physical disturbances and relatively high concentrations of mineral oil: pure product sticks to the sensor material and does not get of easily. For the time being, we therefore conclude the chemical analyses to be more reliable.

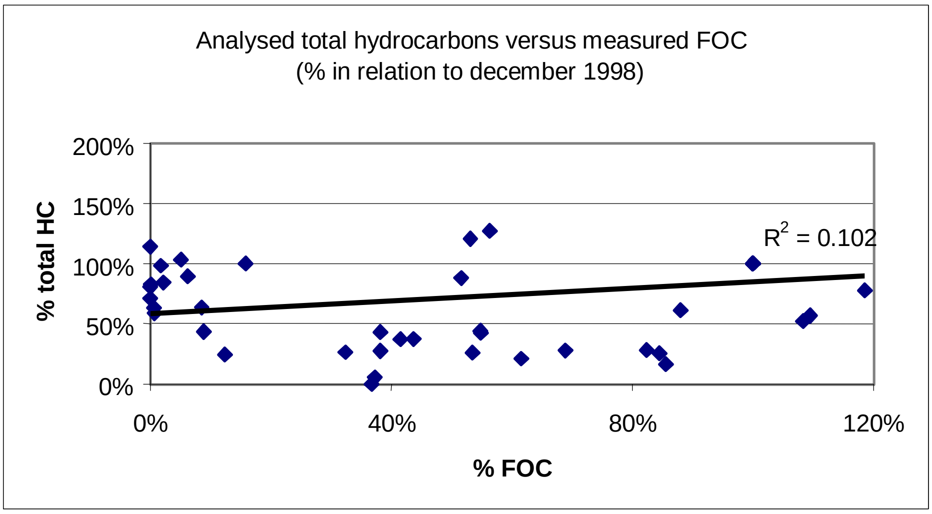
Corresponding pairs of absolute FOC-values versus analysed values

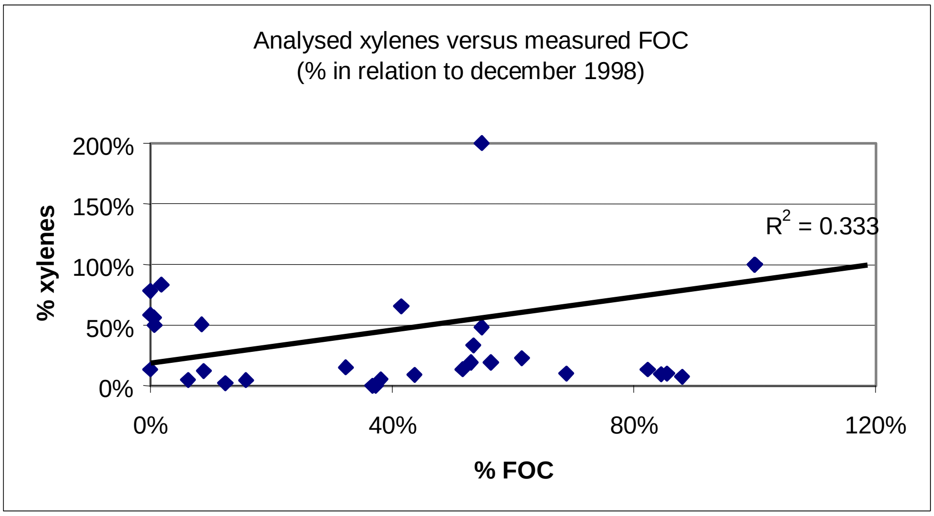


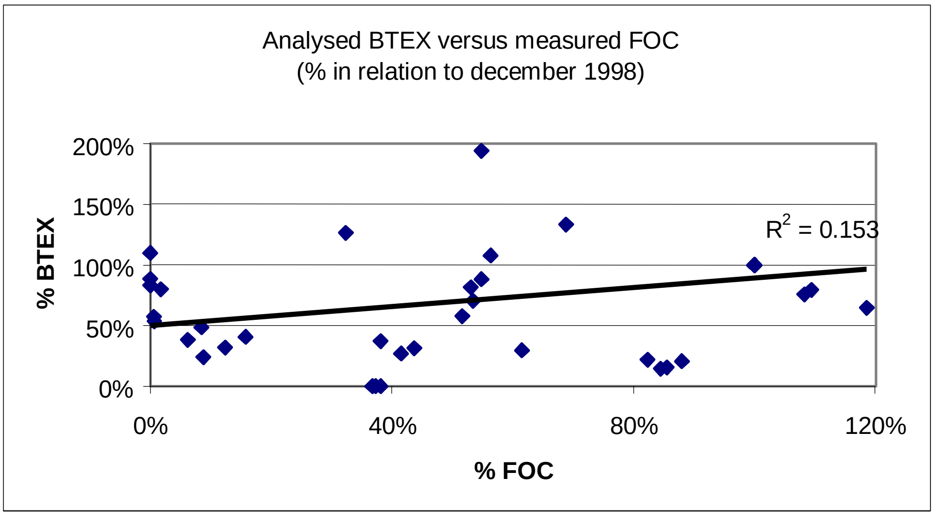




Corresponding pairs of normalized FOC-values versus analysed values







APPENDIX D

DEVELOPMENT OF CONCENTRATIONS HYDROCARBONS AND BTEX

APPENDIX E

MODEL INPUT

Modelling in SORWACO

To get an indication of the rate of biological decay in the fences 1, 2 and 3, the development of concentrations of the volatile fraction of mineral oil (C6 - C12) is simulated in a computer model for solute transport. Because the light fraction of mineral oil is the most dominant one in the field and because this fraction was observed by TNO during the column tests [Van Liere et al., 1998], we modelled solute transport for the fraction C6 - C12. The main advantage of the use of a model for solute transport is that processes like convection, dispersion, sorption and biological decay are integrated. On base of the knowledge of groundwater flow on this site, the organic carbon content of the soil and sorption of C6 - C12, biological decay can be fitted on the measured concentration development in the fences.

For the computer simulation we used the program SORWACO (version 0.03), developed by IWACO. SORWACO is a one dimensional, numerical solute transport model. One of the advantages of this model is the possibility of kinetic limited sorption of organic compounds, especially when groundwater flow is relatively fast. SORWACO divides the soil into a few cells, for which density, porosity and carbon content can be defined. So, in this case the flow path is split up into cells which consist of gravel (measuring drains) and cells which consist of (clayey) sand. Furthermore, SORWACO calculates with a first order decay.

Model input

In the tables E1, E2, E3a and E3b the used values for hydrology and physico-chemical parameters are reproduced.

Table E1. Hydrology parameters.

pilot number	k (m/d) *	ΔH (m) **	Δx (m)	v (flux) (m/d)	$\theta_{effective}$ (-) ***
fence 1	0.5	0.33	3.0	0.055	0.25
fence 2	0.5	0.33	3.0	0.055	0.25 - 0.50
fence 3	0.5	0,33	3.0	0.055	0.30

* field measurements indicated a resistance of about 0.65 m/d (IWACO, January 1998)
** field measurement IWACO, 1999
*** column test TNO, September 1998

Table E2. Physico-chemical parameters.

soil material	ρ (kg/m^3)	θ (-)	organic matter (% w/w)	K_{om} (l/kg) ***
clayey sand	1600	0.30	1.2 **	7320
gravel	1800 *	0.50	0.01	7320

* TNO, September 1998
** analytical measurement IWACO, January 1998
*** average distribution coefficient of C6 - C12, calculated on base of K_{ow} of octane and nonane

Table E3a. Distance-time relation of groundwater in pilot 1 en 2.

description of location	distance to drain C (m)	time (d)	soil material
output/drain C	0	0	gravel
filter 3-6-9	0.2	1.36	clayey sand
-	0.6	3.18	clayey sand
-	1.0	5.00	clayey sand
drain B	1.2	6.36	gravel
filter 2-5-8	1.4	7.73	clayey sand
-	1.9	10.0	clayey sand
-	2.2	11.4	clayey sand
drain A	2.4	12.7	gravel
filter1-4-7	2.6	14.1	clayey sand
input	2.9	15.5	clayey sand

v_{eff} in clayey sand = (0.5 · (0.33/3.0))/(0.25) = 0.22 m/d
v_{eff} in gravel = 0.22 · 0.25/0.5 = 0.11 m/d

Table E3b. Distance-time relation of groundwater in pilot 3.

description of location	distance to filter 3-6-9 (m)	time (d)	soil material
output/filter 3-6-9	0	0	clayey sand
drain C	0.2	1.36	gravel
-	0.4	2.73	clayey sand
-	0.9	5.00	clayey sand
-	1.2	6.36	clayey sand
drain B	1.4	7.73	gravel
filter 2-5-8	1.6	9.09	clayey sand
-	2.0	10.9	clayey sand
-	2.4	12.7	clayey
drain A	2.6	14.1	gravel
filter1-4-7	2.8	15.5	clayey sand
input	3.1	16.9	clayey sand

For the calculations, we assume t = 0 at December 1998. So the initial concentrations (C_0) are the concentrations which are measured at December 7th and 8th 1998. The end concentrations (C_t) are the concentrations measured at November 1st 1999 (t = 329 days). In the tables E4a, E4b and E4c the averages of the measured concentrations C6 - C12 is reproduced for monitoring drains and measuring filters.

Table E4a. Average concentrations C6 - C12 pilot 1 (µg/l).

drain number/filter numbers	December 1998	November 1999
1-4-7	21700	16000
1A	10700	415
2-5-8	19000	12530
1B	12500	3340
3-6-9	19800	9230
1C	11700	6430

Table E4b. Average concentrations C6 - C12 pilot 2 (µg/l).

drain number/filter numbers	December 1998	November 1999
1-4-7	3560	2080
2A	6100	6530
2-5-8	6100	3640
2B	3400	2300
3-6-9	1570	905
2C	1320	980

Table E4c. Average concentrations C6 - C12 pilot 3 (µg/l).

drain number/filter numbers	December 1998	November 1999
1-4-7	1180	2400
3A	1120	240
2-5-8	2670	1390
3B	1160	750
3C	-	-
3-6-9	2430	1610

We refer to the figures E1, E2 and E3 for an overview of the initial groundwater contamination used as input for the model, based on the averages of the concentrations measured in December 1998. From the initial situation it can be concluded that the present contamination in the fences is not homogeneously distributed. Partly this can be explained by the presence of the drains in a gravel ditch, which is initially non-polluted and where hardly any sorption of mineral oil takes place.

Calculations

Calculations were executed for the following scenarios:

1. upstream supply of mineral oil to the fences, no biological decay;
2. upstream supply of non-polluted water to the fences, no biological decay;
3. upstream supply of mineral oil to the fences, biological decay.

Ad 1.

This scenario is calculated to predict the situation in November 1999 if the upstream supply of mineral oil is continuous and biological decay of mineral oil does not occur.

Ad 2.

This scenario is calculated to predict the situation in November 1999 if there is no new supply of mineral oil and biological decay of mineral oil does not occur. Is the inflow of non-polluted water the cause of the decrease in mineral oil concentrations in the fences?

Ad 3.

This scenario is calculated to predict the situation in November 1999 if the upstream supply of mineral oil is continuous and biological decay of mineral oil occurs. Is it possible to predict the measured decrease in mineral oil concentrations in the fences by assumption of biological decay.

Fig. E1, E2 and E3. Overview of the initial groundwater contamination.

Results

The simulation results for November 1999 are presented in the figures 1, 2 and 3 (main text). In these figures the averages of the measured concentrations in November are presented as well. In the figures F1, F2 and F3 of appendix F the correlation coefficient (R^2) between modelled and measured concentrations on November 1999 is presented.

Overall, from the results it is concluded that the measured concentrations of mineral oil (C6 - C12) can not be simulated without the proceeding of biological decay (scenarios 1 and 2). Even if there is an inflow of non-polluted groundwater into the fences, the measured concentrations of November 1999 can not be explained (scenario 2). This is caused by the relatively short operational period of the fences (329 days) in combination with the relatively large retardation of the C6 - C12 fraction. By assuming biological decay, the measured concentrations and the shape of the spatial concentration development can be simulated.

Pilot 1

At a first order degradation rate of about 0.65 d^{-1} (corresponding with a half life of 1.07 day), the measured concentration development can be simulated quite good, except for the concentration in drain A. It seems that biological decay is locally higher here. This is a plausible explanation because pilot 1 has got just one operational injection drain (1A) left. The other injection drain 1B is obstructed since May 1999. Without the values of drain A, the correlation coefficient (R^2) between modelled and measured values is 0.89.

Pilot 2

At a first order degradation rate of about 0.5 d^{-1} (corresponding with a half life of 1.39 day), the measured concentration development can be simulated quite good, except for the concentration in drain A. This concentration does not differ much from the concentration in December 1998. It is known from field observation that an oil floating layer (Light Non-Aqueous Phase Liquid) is present at the upstream side of the fence, at least near filter 1. Without the values of drain A, the correlation coefficient (R^2) between modelled and measured values is 0.93.

Pilot 3

At a first order degradation rate of about 0.6 d^{-1} (corresponding with a half life of 1.15 day), the measured concentration development can be simulated quite good, except for the concentrations in filters 1-4-7. The relatively high (average) concentration at the filters 1-4-7 can be partly explained by a new input of polluted groundwater in filter 7: from October to November the oil concentration increased from 1200 µg/l to 4200 µg/l. Furthermore, it seems biological decay is not proceeding here, possibly because this region is not reached by the injected air. To a less extent, the same goes for the filters 3-6-9: the (limited) decrease of mineral oil concentration here is probably mainly due to biological decay in the upstream zone between drain A and B. Without the values of filters 1-4-7, the correlation coefficient (R^2) between modelled and measured values is 0.76.

Conclusion

From the modelling it is concluded that it is likely to suppose that biological decay is proceeding in all fences. The rate of decay between the fences is comparable, and has a value between 0.5 and 0.65 d^{-1}.

In the column tests TNO-MEP [Van Liere et al., 1998] determined a mineral oil (C6 - C16) removal of 55 % in 35 hours in the 0 % gravel column to 98 % in 35 hours in the 90 % gravel column. From this we can determine a first order decay of 0.55 to 2.68 d^{-1}, by using the following formula: $C_t = C_0 \cdot e^{-kt}$. It is striking that the modelled values of biological decay in the fences are about the same as the one determined in the laboratory for a 0 % gravel column. This confirms the reliability of the modelled values.

By using the results of the stop test (May - June 1999), it might be possible to calculate a maximum rate of biological decay, assuming that all the vanishing oxygen is used by the biological decay of mineral oil. Calculations for this were executed for drain 1A and drain 3A. The results are not presented here, because of the unreasonably high degradation rates calculated, which do not contribute to the understanding of the biological decay of mineral oil. The measured disappearance of oxygen is caused by a lot of totally different mechanisms, of which biological decay (not only of mineral oil), chemical oxidation and evaporation. These processes can not be separated easily.

APPENDIX F

MEASURED CONCENTRATIONS VERSUS SIMULATED CONCENTRATIONS

Fig. F1, F2 and F3. Correlation coefficient (R^2) between modelled and measured concentrations.

APPENDIX G

DEVELOPMENT OF OXYGEN CONTENT

APPENDIX H

DEVELOPMENT OF REDOX POTENTIAL

APPENDIX I

FIELD TESTS TO CHECK THE PILOTS OPERATION

The goal of the field measurements was to find an explanation for the apparent lack of oxygen in the groundwater. Three questions were defined:

1. Is the air really injected into the soil?
2. Where does the air go once it is injected?
3. Is the injected amount of air sufficient?

Ad 1.

The following possibilities were investigated:

- shortcut flow of air through bore hole of injection drains;
- shortcut flow of air through monitoring filters;
- leakage of air at the ends of the injection drains;
- influence of tide on the groundwater level. A difference in level can cause a unequal distribution of air over the injection drains.

Ad 2.

For this question a tracer test was performed.

Ad 3.

Per injection fence the injection rate was increased by steps (start tests) within a wider range than during the optimization period. After oxygen was measured at a significant rate stop tests were performed.

In addition a test was performed to exclude the possibility that a high entrance resistance of the measuring drains for aerated groundwater caused the apparent lack of oxygen.

The measurements have been carried out between May and September 1999. Some additional measurements have only been carried out for fence 2, because:

- until May 7th no oxygen raise was measured at all in this fence;
- the effect of changing the flow rate should be easy to measure here because a gravel ditch is present, so vertical preferential airflow from the injection drains to the measuring drains is expected.

Entrance resistance measuring drain

Method

The explanation for the low oxygen levels that are found in the measuring drain could be that aerated water outside the drains does not enter the drains because of a high entrance resistance. To exclude this possibility one of the measuring drains (2A) was flushed with a motor pump at a constant flow rate of 10 l/min for about 27 minutes. The drain was flushed with approximately 270 l of groundwater what exceeds the volume of the drain (which is about 200 l). In the meantime the oxygen level (mg O_2/l groundwater) and redox potential (Eh in mV) were measured in sampling tube 2A1. Before and during the measurements air was injected at a flow rate of about 15 m^3/hour. The results are reproduced in table I1.

Results

Table I1. Measurements during flushing drain 2A.

time (a.m.)	volume flushed (cumulative in litres)	O_2 (mg/l)	redoxpotential (mV)	pH	electrical conductivity (µS/cm)
10.08	0	0.22	-67	6.94	811
10.16	80	0.90	-78	6.99	779
10.25	170	1.11	-68	7.04	757
10.30	220	0.53	-71	7.02	726
10.35	270 (pump stop)	0.65	-72	7.12	699
10.45	-	0.50	-72	7.08	721

It seems that there is little influence on the oxygen level when the water in the measuring drain is refreshed. The oxygen level does not rise consequently: at first there is a raise to 1.11 mg/l after 170 l groundwater is pumped, but from there on, at an increasing pumped volume the oxygen level drops again to 0.5 mg/l. The latter value is regarded as the practical detection limit.

The results indicate that a measurement artifact, caused by a high entrance resistance of the measuring drain, is not the cause of the apparent lack of oxygen. Nevertheless, from August on the drains were flushed before measurement and sampling to avoid this potential artifact. No sudden raise of oxygen is seen from August on. This confirms that a measurement artifact can be excluded.

Shortcut flow

To exclude the possibility of shortcut flow of air through the layer (bore hole) around the injection or monitoring drain toward the surface near the end of the drains, some soap foam was added to the soil surface at the end of the drains. No bubbles were observed, and we conclude that no shortcut flow is occurring here.

To exclude the possibility of shortcut flow through the monitoring filters into the atmosphere, an instrument with a soap solution was used to make gas flow visible (soap bubbles). The resistance of the soap solution is considered negligible and the instrument should be able to show even very low flows. The instrument was placed on various monitoring filters. No reaction was observed at all.

Leakage of air at the end of the injection drains

The end of the injection drains is stuffed with PUR-foam. No escaping air was observed at the end of the drains.

Groundwater level

Until now it was regarded that the tide did not influence the groundwater level at the location. If the tide *does* influence the groundwater level, it is imaginable that at high tide the entrance resistance for the injected air is higher and the airflow will be influenced.

Method

The groundwater level in the monitoring wells was measured manually at high tide as well as at low tide for fence 2.

Results

The results are reproduced in table I2.

Table I2. Influence of tide on grondwater level and flow (m bgl) in monitoring tubes.

monitoring tube	TT in m at ref. *	surface level at ref.	groundwater level in m from TT		groundwater level in m to reference level		difference groundwater levels
			high tide, 9.30-9.45	**low tide**, 14.15-14.30	**high tide**, 9.30-9.45	**low tide**, 14.15-14.30	**high tide - low tide**
2-1	-1.35	-1.47	-1.57	-	-2.92	-	-
2-2	-1.32	-1.5	-1.73	-1.9	-3.05	-3.22	0.17
2-3	-1.28	-1.45	-1.91	-2.05	-3.19	-3.33	0.14
					slope 0.14	**slope 0.11**	**average high tide - low tide 0.16** *high permeability?*
2-4	-1.41	-1.56	-1.48	-1.51	-2.89	-2.92	0.03
2-5	-1.36	-1.52	-1.54	-1.54	-2.9	-2.9	0
2-6	-1.36	-1.49	-1.79	-1.9	-3.15	-3.26	0.11
					slope 0.26	**slope 0.34**	**average high tide - low tide 0.05**
2-7	-1.43	-1.56	-1.38	-1.4	-2.81	-2.83	0.02
2-8	-1.43	-1.56	-1.45	-1.43	-2.88	-2.86	-0.02
2-8	-1.43	-1.46	-1.86	-1.89	-3.18	-3.21	0.03
					slope 0.37	**slope 0.38**	**average high tide - low tide 0.01** *low permeability?*

TT in m at ref * top of tube at reference level

It is measured that perpendicular to the harbour at one side of the fence there is a difference of about 15cm between low and high tide, while the groundwater level at the other side is hardly influenced. This is probably due to differences in soil permeability. So there is a spatial variation of the reaction of groundwater level on the tide. This means that the distribution of air flow over the injection drains can be influenced: temporally one side of the fence gets more air than the other side because of a difference in entrance resistance. Still, this means that at all times at least one part of the fence should receive enough air to raise the oxygen level above 1.0 mg/l. As this was not measured we conclude that the small influence of the tide does not explain why hardly any raise of oxygen level could be measured.

Conclusions

From the additional measurements described here, one can conclude that the pumped air is not diverted by shortcut flow or leakage of the system, or could not be delivered because of influence of tide. The pumped air was really injected in the soil. This brings us to question 2: Where does the injected air go?

Tracer test

Method

To examine where the injected air and oxygen reach the surface, a gas tracer test was executed on July 12th and 13th. Helium, an inert gas, was added to the air in doses varying from 1.5 l/min (fence 3) to 3.0 l/min (fence 1 and 2). The helium was injected in the air just before that entered the compressors. After injection the helium was traced by a mobile helium analyser near the surface (10 cm bgl) on 24 locations at each fence. The measurements were performed in duplo, at 0.5 m from one another. So at each fence 48 measurements were performed.

Results

The measurements, an interpolation to the entire fences and an interpretation are attached in appendix J. The interpretation shows that:

- at fence 1 and 3 little helium was recovered at the fences surface, respectively 4.1 % and 0.024 %;
- at fence 2 about 67 % of the helium was recovered at the fences surface.

The high recovery at fence 2 is expected because the gravel ditch causes preferential flow paths straight to the surface. However, the measurements show low recovery at a part of the gravel ditch, in the middle of the fence. There are two possible explanations:

- A collapse of the trench during the construction caused a condensed layer, which diverts the air flow horizontally and keeps it from reaching the surface. This explains the low recovery at this part of the fence. The missing part of the air flow is probably transported in the direction of the harbour.
- The injection drain is blocked and does not give air and only at the north and south side the fence is sparged. There is no way to check this possibility without demolishing the fence.

The small recovery of helium at the subsurface of fences 1 and 3 might be explained by the existence of one or more poorly conducting soil layers between soil surface and depth of injection which divert the air flow. It is our experience that air flow is very sensitive to such dense layers and it will prefer to follow the most permeable layers. As bubbles were already observed in the harbour at fence 1 (extreme high tide) and at fence 3 (at high tide), probably most of the air comes to the surface in the harbour.

The results of the tracer test show that at fence 2 a considerable amount of the injected air is transported vertically and reaches the surface at the fences plot. At the other fences (1 and 3) the test indicates the transport of air is mainly horizontal (and probably pointed to the harbour).

This does not mean that the air escapes without exchanging its oxygen to the groundwater, and no oxygen becomes available for biological decay of pollution. This depends on the depth of the dense layers. When for example the dense layer(s) is (are) present in the unsaturated zone, exchange can take place between air and water during the travelling time between injection point and harbour, and the saturated zone in which the pollution is transported, will be aerated. After these measurements, soil profiles were examined for the presence and depth of dense layers.

Soil profiles at the fences

The results of the helium tracer test suggest that at the fences 1 and 3 a dense soil layer is present: a low recovery of helium at the soil surface and air bubbles in the harbour. To prove the existence of one or more dense layers and to see at what depth these are present, existing and additional soil profiles were examined on sight. The results are shown in appendix L. A cross-section of fences 1 and 3 is given in appendix L also.

Fence 3

The bore logs with the numbers OBP1 (south), OBP2 (north) and 3-10 (middle) belong to fence 3 (see location plan of appendix L). The bore logs sometimes show clay layers and often the presence of clay lumps in sandy layers is mentioned. As clay is not likely to be deposited as lumps but as (thin) layers, these lumps are caused by the sampling technique and indicate the presence of thin clay layers. From the bore logs than one can conclude that at a depth of 1.6 - 1.7 m bgl, 2.45 - 2.5 m bgl (locally at north side) and below 3.8 m bgl clay layers are present. The injection filters are placed on the deepest clay layer at a depth of 3.5 - 4.0 m bgl. The monitoring filters and measuring drains are placed at a depth of respectively 2.5 - 3.5 m bgl and 2.5 m bgl. The groundwater level is about 1.7 m bgl. So the monitoring filters and the measuring drains should be reached by the injected air, but the injected air does not reach the surface because of the clay layer at 1.6 - 1.7 m bgl. Locally the saturated zone between 1.7 and 2.5 m bgl is probably not reached by the injected air.

Fence 1

The bore logs with the numbers OBP3 (south), OBP4 (north) and 1-10 (middle) belong to fence 1 (see location plan of appendix L). From these bore logs one can conclude that at a depth of 0.6 - 1.0 m bgl (locally in the middle), 1.6 - 1.7 m bgl (locally at south side), 3.4 - 3.7 m bgl (locally in the middle) and below 4.0 m bgl clay layers are present. The injection drains are placed on the deepest clay layer at a depth of 4.0 m bgl. The monitoring filters and measuring drains are placed at a depth of respectively 2.5 - 3.5 m bgl and 2.5 m bgl. The groundwater level is about 1.6 m bgl. So the monitoring filters and the measuring drains should be reached by the injected air, except locally in the middle of the fence because of the layer between 3.4 and 3.7 m bgl. The injected air does not reach the surface at the south side and the middle of the fence because of the clay layers at 1.6 - 1.7 m bgl and 0.6 - 1.0 m bgl. This explains the low recovery at these places. The relatively higher recovery of helium at the north side can be explained by the absence of a clay layer over here (see bore log OBP4 in appendix L).

Start/stop tests from May 7th until June 24th

Method

The influence of the flow rate on the oxygen content of the groundwater is examined by varying the air flow rate of the injection drains. This is done during a so-called start test. For the start tests the total flow capacity of the system (45 - 50 m^3/h) was available for the fence on which the start test had to be carried out. Before the start tests were performed no air injection was operational. The flow rate was increased by steps. Every flow rate was continued for at least 45 minutes. At one of the flow rates the oxygen level seemed to be highest. At this flow rate the fence was operated for at least one week, after which oxygen levels in the monitoring drains of the fence were measured again. After these measurements the injection was stopped to execute a stop test. During the stop test the influence in time of shutting down the air injection on the oxygen level of the groundwater was examined.

Result start test

Fence 1

The difference between the oxygen level at sampling tube 1A1 (about 2.0 mg/l), with sampling point 1A3 (about 6.5 mg/l) is an indication of a strong variety of oxygen level through the fence that seems to be caused by preferential flow paths to a part of the measuring drain.

A summary of the results is shown in table I3 and figure I1.

Table I3. General relation between air flow and oxygen concentration in groundwater for measuring drain 1A.

flow rate (m^3/h)	oxygen level (mg/l)
1	0.6 - 2.8
15	1.4 - 6.6
20	1.6 - 2.4

The results show that oxygen levels rises when the flow rate is increased to 15 m^3/h, but falls when the flow rate is further increased to 20 m^3/h , indicating that there is no linear relationship between oxygen level and injection rate.

Fence 2

At 40 m^3/h the oxygen concentration in the drains 2A and 2B is about 1.5 and 2.0 mg/l, while the oxygen concentration in downstream drain 2C is about 0.4 mg/l. As expected, this means that the injected oxygen is *vertically transported* to the surface through the gravel trenches in which the monitoring drains 2A and 2B lie on top of the injection drains 2A and 2B. Measuring drain 2C is located downstream of the injection drains outside the gravel trenches with injection drains.

At 30 m^3/h sampling tube 2B1 shows a oxygen level of 3.1 mg/l, while at sampling tube 2B2 oxygen level is still below 1 mg/l. At an airflow rate of 40 m^3/h, 2B2 shows a significant rise of oxygen to about 2 mg/l. This indicates that higher flow rates are needed to provide the whole length of the drain with air (instead of water).

A summery of the results is shown in table I4 and figure I1.

Table I4. General relation between air flow and oxygen concentration for measuring drain 2A.

flow rate (m^3/h)	oxygen level (mg/l)
15	0.2 - 0.5
20	0.5 - 1.4
30	1.4 - 2.5
40	0.8 - 1.6

Fig. I1. Start test pilots 1, 2 and 3.

The results show that oxygen levels rises when the flow rate is increased to 30 m^3/h, but falls when the flow rate is further increased to 40 m^3/h, indicating that there is no linear relationship between oxygen level and injection rate.

Fence 3

A summery of the results is shown in table I5 and figure I1. At an injection rate of 15 m^3/h sampling tube 3A shows a large variation of the oxygen level, from 0.1 to 4.9 mg/l, along the drain. The results show that the oxygen level falls when the flow rate is increased to 32 m^3/h, indicating that there is no linear relationship between oxygen level and injection rate.

Table I5. General relation between air flow and oxygen concentration for measuring drain 3A.

flow rate (m^3/h)	oxygen level (mg/l)
15	0.1 - 4.9
32	0.0 - 0.2

Discussion on the start test

The relationship between oxygen concentration and injection rate is not linear. This is supposed to be caused by a physical change in air flow at different flow rates. The consequence is that it is not useful to maximise the injection rate. There seems to be an optimum flow rate at which the most oxygen is exchanged from injected air to groundwater. We can not say that by this an *optimum* flow rate is determined. The used range of flow rates is apparently sufficient to raise the oxygen level (at two of the three fences).

The three fences react differently:

- at fence 1: a maximum O_2 concentration of 7 mg/l at a flow rate of 15 m^3/h per drain;
- at fence 2: 2.5 mg/l at a flow rate of 15 m^3/h per drain;
- at fence 3: 5 mg/l at a flow rate of about 4 m^3/h per injection line.

These differences are supposed to be caused by the sensitivity of a system for undesirable flow patterns. In fence 2 the coarse and rather homogeneous medium might result in larger bubbles what might be less favourable for the transfer of O_2 from air to the water phase. With vertical injection systems (fence 3) the total filter length is smaller, thus resulting in higher flow velocities over the filters at the same injection rate. Table I6 shows a comparison of the injection rates and flow velocities of fence 1 and 3.

Table I6. Comparison of flow velocities of fence 1 and 3.

fence	Q_{inject} (m^3/hour)	filter length (m)	flow velocity (m^3/hour.m)
1	20	2·30	0.33
3	8	20·0.5	0.8

Besides that, in the fences 1 and 3 dense layers are present just above the monitoring filters and measuring drains, so the travel time of air in the soil is longer and the exchange of oxygen from air to water is more efficient.

The time to maximixe the oxygen levels at a certain injection regime, is not easy interpretative from the start test. From fence 2 and 3 one might conclude that about 15 minutes of air injection is sufficient to maximixe the oxygen levels. However as the flow rates were varied fast, this conclusion might be premature.

Results stop test

From the stop test results of biofences 1 and 3 (see fig. I2) it is concluded that within about 30 minutes the injected oxygen level is decreased to less than 1 mg/l. This decrease is very high in reference to our experiences. The oxygen is either consumed by biological or chemical processes.

Nevertheless, this result, together with the result of the start test that it takes about 15 minutes to maximize the oxygen levels, can be used as indication on how to operate an intermittent injection regime.

For biofence 2 we did not succeed to execute a stop test, because an oxygen level higher than 1.0 mg/l could not longer be reached after the start test, as this was tried several times.

Overall conclusions additional measurements

It can be excluded that a measurement artifact, caused by a high entrance resistance of the measuring drain, is the cause of the apparent lack of oxygen.

It is *excluded* that a substantial part of the injected air is escaping by shortcut flow through bore hole or injection drain or by leakage of air at the end of the injection drains and all the pumped air is injected into the soil.

At fences 1 and 3 little helium is recovered at the subsurface during the tracer test. It is proven by bore logs that some dense clay layers are present between the point of injection and the surface. This means that at (a part of) these fences the exchange of oxygen from air to water is more efficient in the saturated zone because of a longer travel time of air.

From the results of the start test it is concluded that the relationship between oxygen concentration and injection rate is not linear. There seems to be a certain injection range at which there is an maximum achievable oxygen concentration. Higher flow rates possibly cause a different physical form of air (bigger bubbles or channels) from which exchange of oxygen is less efficient. The results of the stop test show that within about 30 minutes a raised oxygen level (5 - 7 mg/l) is reduced to the initial level (< 1 mg/l).

On base of these additional measurements and the calculations (see appendix K) the injection regime was permanently adjusted on August 17th. A continuous injection system was chosen:

1. in view of technical purposes;
2. to avoid deficiency of oxygen at any time.

Since August 17th the following flow rates have been injected:

- fence 1: 10.3 m^3/hour (1 drain);
- fence 2: 24.7 m^3/hour (12.35 m^3/hour each drain, 2 drains);
- fence 3: 8.2 m^3/hour (about 0.4 m^3/hour each filter, 20 filters).

Fig. I2. Stop test pilots 1 and 2.

APPENDIX J

RESULTS TRACER TEST WITH HELIUM (FIGURES)

Table J1. Interpretation tracer test.

	pilot 1	pilot 2	pilot 3
diameter sd modus	6 mm	6 mm	6 mm
surface sd modus	2.8274E-05 m^2/sd modus	2.83E-05 m^2/sd modus	2.83E-05 m^2/sd modus
number sd modi	48	48	48
surface modi total	0.00135717 m^2 sd/plot	0.001357 m^2 sd/plot	0.001357 m^2 sd/plot
width plot	30 m	3.5 m	30 m
length plot	3.5 m	30 m	3.5 m
surface plot	105 m^2	105 m^2	105 m^2
surface modi/surface plot	1.2925E-05 (-)	1.29E-05 (-)	1.29E-05 (-)
average of measurements	2.63E-05 ml/s surface sd modus	4.39E-04 ml/s surface sd modus	7.86E-08 ml/s surface sd modus
average flux plot	2.0 ml/s	33.9 ml/s	6.1E-03 ml/s
input helium flux	3 l/min	3 l/min	1.5 l/min
input helium flux	0.05 l/s	0.05 l/s	0.025 l/s
input helium flux	50 ml/s	50 ml/s	25 ml/s
% recovery	4.1 %	67.9 %	2.4E-02 %

Fig. J1. Tracer test pilot 1.

Fig. J2. Tracer test pilot 2.

Fig. J3. Tracer test pilot 3.

APPENDIX K

CALCULATIONS ON OXYGEN DEMAND

Method

Because there was doubt within the consortium that the amount of injected air is sufficient to optimize conditions for aerobic decay of hydrocarbons, (re)calculations of oxygen demand for biological decay and chemical oxidation of ground and groundwater were executed. The most important processes to take into account are:

- oxygen transfer efficiency from air to water;
- biological decay of mineral oil compounds in groundwater;
- chemical oxidation of (reduced) minerals in ground and groundwater;
- biological/chemical decay of (dissolved) organic carbon in ground and groundwater (DOC).

For the calculation the following assumptions were made:

- the oxygen transfer from air to soil (= groundwater and ground) is 10 %;
- the oxygen consumption by biology is based on experience and literature;
- the chemical oxygen demand is mainly contributed by $Fe(II)OH_2$ (reaction 1, see the last page of this appendix) or by FeS (reaction 2, see the last page of this appendix). The latter should result in an increasing sulphate concentration and a lowering of pH. Measurements point out that the pH is more or less stable. Nevertheless, this is a reaction which should be accounted for, because of its potential huge influence on oxygen demand;
- the chemical oxidation reactions are instantaneous;
- about 80 % of the reduced minerals present in the solid phase are attainable/susceptible.

Results

The indicative calculations are attached in this appendix. The results are summarized in table K1.

Table K1. Calculated oxygen demand of each fence.

process	oxygen demand
chemical oxidation of minerals in ground (assumed a finite process)	125 - 1120 kg O_2
chemical oxidation of minerals in groundwater chemical/biological oxidation of DOC in groundwater biological oxidation of mineral oil comp. in groundwater	18.7 - 146.8 g O_2/d 79.8 g O_2/d 4710 g O_2/d
total oxygen demand of groundwater	4810 - 4940 g O_2/d (≈ 7.8 - 8.0 m^3 air/hour)

To avoid underestimation, some of these data have been double-checked by comparing them with other (field)sources:

1. The calculated amount of oxygen that is needed for the decay of DOC can be checked by comparing it to the BOD mentioned in the CUR/NOBIS report 'Characterization and design of a biological fence' [CUR/NOBIS, 1999]. The BOD_{15} is about 11 mg/l ≈ 41.3 g O_2/d. So the calculated oxygen need of about 79,8 g O_2/d is probably over estimated.
2. The calculated total oxygen demand of groundwater can be checked by comparing it to the results of micro-oxymax experiments on groundwater by TNO-MEP [Van Liere et al., 1998]. From these tests it was concluded that the reduction capacity of the groundwater is 43 mg O_2/l/d, with a total reduction capacity of 431 mg O_2/l. This means that every day about 28.7 mg O_2/l is needed for a complete degradation within the fence (calculation not shown

here). In total 1615 g O_2/d for the whole fence is needed then. So the calculated total oxygen demand of groundwater of about 4810 g O_2/d is probably over estimated.

3. At another micro-oxymax experiment TNO-MEP determined the reduction capacity of ground. From these tests it was concluded that the reduction capacity (total oxygen demand) of the ground is 138 mg O_2/l/d (41.4 kg O_2/d for each fence). TNO-MEP concludes that this capacity is not as high as expected when compared with the capacity of groundwater, but is expected to last longer. How much longer is not clear as the total reduction capacity of ground was not determined in the test.

The difficulty with the calculated oxygen demands is that we do not know the kinetics of the various processes. The assumption is that, as oxygen is supplemented to groundwater, the oxygen demands in the groundwater will be met first, and once the groundwater is aerobic (and an oxygen concentration gradient between ground and groundwater is present) the ground will be oxidized.

At the lowest airflow rate of 8.2 m^3/h in biofence 3, about 5080 g O_2/d is injected. This exceeds the amount of the calculated oxygen demand for groundwater (4810 - 4940 g O_2/d), which is regarded as an overestimation. As yet, from this it is concluded that the amount of injected air (and oxygen) should be sufficient.

APPENDIX L

SOIL PROFILES AND LOCATION OF SOIL PROFILES

APPENDIX M

COST CALCULATION BIOLOGICAL FENCE AND CONVENTIONAL P&T SYSTEM

APPENDIX N

EVALUATION OF MONITORING

Monitoring of process parameters

This study shows the inefficiency of monitoring of oxygen levels to asses the operation of biological fences. At this study a sometimes remarkable decrease of hydrocarbons was seen, which is most likely an effect of degradation, although low oxygen levels were measured and measurements of redox potential confirmed these anoxic conditions. Possibly tracer tests are a good alternative for a quick assessment of the distribution of injected air and perhaps as well for an indication of extend of transfer from air to groundwater.

Measuring drain versus vertical stand pipes

Hypotheses

The concentration of mineral oil along a profile should decrease with depth. However, the concentration of oxygen in a soil profile will behave otherwise. Because of the injection of air in the subsoil, the concentration of oxygen should show a profile with an increasing concentration of oxygen with depth. In the three fences this hypotheses will be checked, for both the fences, as well as within the fences each.

Oxygen

Fence 1

In row A an increase of the concentration of oxygen has been measured in the measuring drains while in the standpipes no oxygen was found. At line B of the measuring drain higher oxygen were measured than in the standpipes In the standpipes in line C (downstream) a clear increase of oxygen was measured (this is also seen in standpipe 1-1). In the measuring drains in this same line no oxygen was measured.

Fence 2

In the standpipes of fence 2 low concentrations of oxygen were measured. In standpipes 2-2, 2-10 en 2-12 relevant concentrations of oxygen were measured. In the drains more oxygen was measured, with an increase in concentration in line B.

Fence 3

For fence 3 standpipe 3-10 (the deepest filter) is the only standpipe where oxygen has been measured in a relevant amount. In the measuring drains of line A and B oxygen was also measured but there was no obvious increase or decrease of the total oxygen. In the measuring drains more oxygen was measured than in the standpipes.

Total HC

Fence 1

In the standpipes of fence 1 the concentration of hydrocarbons is higher than in the measuring drains. At all points the concentration decrease in time or do not change at all.

Fence 2

In the standpipes in line A the concentrations of hydrocarbons increased in time. In the measuring drains no obvious decrease or increase of the hydrocarbons were measured. In lines B and C there is a clear decrease in the measured concentration. In general, the hydrocarbons in the standpipes were present in higher concentrations than in the measuring drains.

Fence 3

At most of the sampling points, in the standpipes as well as in the measuring drains, the total amount of HC is decreasing in time. Especially in the deepest filters HC is decreasing quit fast. The measured concentrations of hydrocarbons in the standpipes are higher than the concentrations measured in the measuring drains.

FOC-sensor versus lab analysis

For the measurements of the FOC-sensors from December 1998 until November 1999 we refer to the report of phase 3, of December 13th, 1999 [Heijnen and Praamstra, 1999].

In phase 2 [Heijnen and Vis, 1999] is was concluded that the information from the sensor measurements differ from that of the chemical analysis.

In this phase the FOC-measurements were compared with the concentrations from laboratory analysis. At first the absolute measurements were compared, after that the relative concentrations (september 1999 = 100 %) The comparison was done with the total hydrocarbons concentration, the BTEX concentration and xylene concentration (see the figures in appendix C). In case of comparing the absolute data of the parameters no relation is observed. By comparing the relative increase or decrease in relation to September 1999 also no relation is observed.

The difference of trends between analysed and detected concentrations may be caused by the non-specific measurement by the sensors of total hydrocarbons. The sensors are calibrated for xylenes. Possibly they are less sensitive for mineral oil compounds like alkanes.

From field experience, we now know the sensors are sensitive to frost or other physical disturbances and relatively high concentrations of mineral oil: pure product sticks to the sensor material and does not get of easily. For the time being, we therefor conclude the chemical analyses to be more reliable.