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# Environmental assessment of contaminated site remediation in a life cycle perspective


Gitte Lemming

DTU Environment
Department of Environmental Engineering

PhD Thesis
September 2010

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The thesis will be available as a pdf-file for downloading from the homepage of the department: www.env.dtu.dk

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# Preface

The work reported in this PhD thesis, entitled 'Environmental assessment of contaminated site remediation in a life cycle perspective', was conducted at the Department of Environmental Engineering at the Technical University of Denmark with Professor Poul L. Bjerg as main supervisor and Professor Michael Z. Hauschild as co-supervisor. The PhD project ran from October 2006 to June 2010 and was funded by the Technical University of Denmark.

The research was primarily carried out at DTU, but included a 3-months external research stay (Sep-Dec 2009) at the Interuniversity Research Centre for the Life Cycle of Products, Processes and Services (CIRAIG) at École Polytechnique in Montreal, Canada.

The content of the PhD thesis is based on three scientific journal papers of which two are published. In the thesis, these papers are referred to by author names and Roman numerals (e.g. Lemming et al., I) and are attached as appendices to the thesis.

**I** Lemming, G., Hauschild, M.Z. and Bjerg, P.L. Life cycle assessment of soil and groundwater remediation technologies: literature review. International Journal of Life Cycle Assessment 2010, 15 (1), 115-127.

**II** Lemming G., Friis-Hansen P. and Bjerg P.L. Risk-based economic decision analysis of remediation options at a PCE-contaminated site. Journal of Environmental Management 2010, 91 (5), 1169-1182.

**III** Lemming G., Hauschild, M.Z., Chambon, J., Binning, P.J., Bulle, C., Margni, M. and Bjerg P.L. Life cycle assessment as a decision support tool for evaluation of contaminated site remediation alternatives. Submitted manuscript.

The papers are not included in this web-version, but can be obtained from the library at DTU Environment. Contact library@env.dtu.dk or Department of Environmental Engineering, Technical University of Denmark, Miljoevej, Building 113, DK-2800 Kgs. Lyngby, Denmark.

Additionally, the following reports and publications, related to the topic of the thesis, have been co-authored during the PhD-study, and will also be referred to in the thesis:

Troldborg M., Lemming G., Binning P.J., Tuxen N. & Bjerg P.L. Risk assessment and prioritisation of contaminated sites on the catchment scale. Journal of Contaminant Hydrology 2008, 101 (1-4), 14-28.

Weber, K., Lemming, G., Wodschouw, N., Munch-Andersen, C.Z., Jensen, C.B., Kiilerich, O., Sørensen, K. & Terkelsen, M. Remediation Strategy for Soil and Groundwater Pollution – RemS – A Decision Support Tool. In conference proceedings of GreenRemediation. Incorporating Sustainable Approaches in Site Remediation, Copenhagen, Denmark, 9-10 November 2009.

Chambon, J., Lemming, G., Broholm, M.M., Binning, P.J., and Bjerg, P.L. Model assessment of reductive dechlorination as a remediation technology for contaminant sources in fractured clay: Case studies Delrapport III. Environmental Project 1296, 2009. Danish Environmental Protection Agency. Copenhagen, Denmark.

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Gitte Lemming

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First of all, I would like to thank my supervisor, Professor Poul L. Bjerg, for his consistent support and advice throughout the project and for his enthusiasm about my project. I also wish to thank Professor Michael Hauschild, who became my co-supervisor during the last part of the study and was a great support due to his knowledge and experience with life cycle assessment.

I am also thankful to all the co-authors who contributed to my work: Peter Friis-Hansen, Manuele Margni, Cécille Bulle, Julie Chambon and Philip J. Binning. It was a pleasure to work with you!

Furthermore, I wish to extend a warm thanks to all staff and students at CIRAIG, Montreal, for making my research stay there both professionally and personally rewarding.

The Capital Region of Copenhagen, especially Carsten Bagge Jensen, is acknowledged for providing me with data about the case studies that were used in the project. The Technical University of Denmark is gratefully acknowledged for financing the PhD stipend and Otto Mønsted Foundation is acknowledged for financial support for conference and research travels. The Postgraduate School of Industrial Ecology (PSIE) is greatly acknowledged for organising and funding my participation in a very interesting PhD course on life cycle assessment in 2007.

I also wish to thank to my friends and colleagues at DTU Environment, especially Mads Troldborg, my office mate, for good company during the last 3 years and for his comments on my thesis. Thanks to Anne Harsting for her help with paperwork and to Torben Dolin and Lisbet Brusendorff for their help with illustrations.

Lastly, I wish to thank my family and friends outside the university, for helping me think of other things than my project.

# Summary

Many contaminated sites worldwide constitute a hazard to their surroundings and must undergo remediation. Chloroethenes such as trichloroethene (TCE) and perchloroethene (PCE) are among the most frequently encountered contaminants in the subsurface due to their widespread use as solvents in dry-cleaning and industries. Chloroethenes are dense non-aqueous phase liquids (DNAPLs) with high density and viscosity and low solubility in water. These characteristics allow a spill to migrate deep into the subsurface, where it can act as long-term source of dissolved-phase groundwater contamination.

Due to the longevity of chloroethene source zones, conventional pump-and-treat technologies are inefficient and may require operation for centuries. Excavation of the contaminated soil and subsequent treatment and disposal of the soil is another ex situ option, however most suitable for contaminant source zones located close to the surface. As an alternative to these ex situ remediation methods, in situ remediation methods for chloroethenes have been developed to target the contaminants in their subsurface location. These technologies cover chemical, biological and physical methods of which the latter can be enhanced by heating the subsurface.

This PhD project investigated the applicability of life cycle assessment as a tool for environmental assessment of remediation of contaminated sites. This was done focusing specifically on chloroethene-contaminated sites and remediation technologies relevant for this type of contaminant. LCA is an environmental assessment tool that compiles a very wide array of environmental exchanges (emissions to air, water, and soil, and resource consumption) associated with the life cycle of a product or service .and translates them to impacts (global warming, acidification, human toxicity, ecotoxicity, etc.).

A literature survey showed that although a number of studies of LCA and remediation had been published during the recent 11-year period only two of them included assessment of chloroethene remediation. However, these studies focused on ex situ remediation or groundwater plume remediation using a reactive barrier. Thus, the majority of innovative in situ remediation methods for chloroethene source zone remediation were not covered in the literature.

Within the project, life cycle assessments of remediation alternatives for source zone remediation of two chloroethene-contaminated sites were performed. These studies covered the assessment of in situ techniques soil vapor extraction

(SVE), in situ thermal desorption (ISTD) and enhanced reductive dechlorination (ERD) and the ex situ technique of excavation followed by off-site treatment.

The results from the first case study, which compared SVE, ISTD and excavation with off-site treatment, showed that SVE had the lowest environmental impacts when a timeframe of 30 years was used, but became less preferable than ISTD and excavation if a more realistic timeframe of 100 years was used. In the other case study, ERD, ISTD and excavation with off-site treatment were compared. The study showed that ERD is a promising low-impact technology for this type of site as it had significantly lower impacts than ISTD and excavation in all impact categories and performed only slightly worse than the no action scenario, where only monitoring was carried out. ISTD had the highest potential impact on global warming due to the large electricity use, but for the remaining impact categories excavation had comparable or larger impact scores than ISTD.

The above mentioned results cannot be seen as to apply universally. LCAs of contaminated site remediation are inherently site-specific as many inputs to the LCA depend on the location of the site, e.g. transportation distances for excavated soil and clean refill and the country-specific electricity production. The depth, water content and contaminant levels of the remediated soil volume are other sources of variation between sites. In addition, system and time boundaries and the type of LCA conducted (attributional or consequential) has an impact on the final results.

Life cycle assessments aim to compare environmental burdens associated with different ways of obtaining the same function or service denoted *the functional unit*. Most studies define the functional unit as the volume of contaminated soil or groundwater to be treated and combine it with a remedial target for the contaminant concentration. However, although two remediation methods reach the same remedial target with time, their timeframes can be substantially different. This quality difference can be included in the LCA by assessing the so-called *primary impacts*. Primary impacts are local toxic impacts related to the contamination at the site as opposed to the *secondary impacts* stemming from the remedial actions.

Primary impacts have typically been assessed using *site-generic* characterization models representing a continental scale and excluding the groundwater compartment. Soil contaminants have therefore generally been assigned as emissions to surface soil or surface water compartments. However, such site-generic assessments poorly reflect the fate of chloroethenes at

contaminated sites as they exclude the groundwater compartment and assume that the main part escapes to the atmosphere.

In the two case studies, the primary impacts were assessed using *site-dependent* procedures, where the contaminant emissions to groundwater over time were estimated based on site-specific contaminant fate and transport models. This made it possible to account for important processes, such as the formation of chlorinated degradation products and to include the site-specific exposure of humans via ingestion of groundwater used for drinking water. The inclusion of primary impacts in the environmental assessment of remediation alternatives gave a more complete basis for comparison of technologies with substantially different timeframes and efficiencies.

LCA was concluded to be a useful tool for environmental assessment of remediation of contaminated sites although unresolved issues remain. Among the obstacles identified for the use of LCA as decision support for remedy selection is the fact that conducting an LCA is very data and time consuming. Furthermore, the multi-indicator result may be difficult to interpret especially given the higher uncertainty of the toxicity-related impact categories. Thus, improvements of characterization methods for toxic impacts as well as expansion of remediation-relevant LCI databases were among issues identified for future attention in order to enhance the applicability of LCA. Moreover, further development of methods for monetization of life cycle impacts may enhance the use of LCA within this field as it makes it easier to integrate the result of the environmental assessment with other decision criteria such as remediation cost.

# Dansk sammenfatning

Mange forurenede grunde over hele verden udgør en trussel for deres omgivelser og må oprenses. Klorethener, som eksempelvis triklorethen og perklorethen, er blandt de foreningstyper, der oftest findes i undergrunden på grund af deres udbredte anvendelse som opløsningsmidler i kemisk tøjrensning og i industrien. Klorethener er dense non-aquos phase liquids (DNAPLs) med høj densitet og viskositet og lav vandopløselighed. Disse egenskaber gør, at et spild vil kunne bevæge dybt ned i undergrunden, hvor det kan fungere som en langvarig kilde til grundvandsforurening.

På grund den lange levetid af kildeområder forurenet med klorethener, er konventionel afværgepumpning og vandbehandling ineffektiv og kan kræve behandlingstider på flere hundrede år. Afgravning af den forurenede jord og efterfølgende rensning og deponering er en anden ex situ metode, som dog er mest anvendelig for overfladenære forureninger. Som et alternativ til ex situ oprensning er der udviklet metoder til in situ oprensning af klorethener rettet imod forureningens placering i undergrunden. Disse teknikker omfatter både kemiske, biologiske og fysiske metoder, hvoraf de sidste også omfatter opvarmning af jorden.

Dette ph.d.-projekt har undersøgt anvendelsen af livscyklusvurdering (LCA) som et værktøj til at foretage miljøvurderinger af oprensning af forurenede grunde. Dette blev gjort med et specifikt fokus på klorethen-forureninger og relevante afværgemetoder for denne type forurening. LCA er et miljøvurderingsværktøj som samler et bredt spektrum af udvekslinger med miljøet (emissioner til luft, vand og jord samt ressourceforbrug) relateret til hele livscyklussen af et produkt eller en service og omsætter dem til miljøpåvirkninger (global opvarmning, forsuring, human- og økotoksicitet etc.).

Et litteraturstudie viste, at selvom der er publiceret et antal studer omhandlende LCA og oprensning, så inkluderer kun to af disse vurderinger af oprensning af klorethen-forurening. Disse to studier har fokuseret på ex situ oprensning eller oprensning af en grundvandsfane ved brug af en reaktiv væg. Størstedelen af de innovative afværgemetoder til in situ oprensning af kildeområder forurenet med klorethener er derfor ikke dækket i litteraturen.

I dette projekt blev der udført livscyklusvurderinger for oprensnings-alternativer for to grunde forurenet med klorethener. Disse studier inkluderede vurderinger af in situ teknikkerne jordventilering (SVE), in situ termisk

desorption (ISTD) og stimuleret reduktiv deklorering (SRD) samt ex situ oprensning ved brug af afgravning og efterfølgende jordrensning.

Resultaterne fra det første case-studium, der sammenlignede SVE, ISTD og afgravning med ekstern rensning, viste at SVE havde de laveste miljøeffekter, hvis en tidshorisont på 30 år blev anvendt, men at SVE blev mindre favorabel end ISTD og afgravning, hvis en mere sandsynlig tidshorisont på 100 år blev anvendt. I det andet case-studium blev oprensning med SRD, ISTD og afgravning med ekstern rensning sammenlignet. Studiet viste, at SRD er lovende som miljøvenlig afværgeteknologi for denne type forurening, da den havde signifikant lavere miljøpåvirkninger end ISTD og afgravning, og kun i begrænset omfang var dårlige end no-action-scenariet, der kun inkluderede monitering. ISTD havde den største potentielle effekt på global opvarmning grundet det høje elforbrug, mens afgravning for de øvrige påvirkningskategorier havde højere eller sammenlignelige påvirkninger i forhold til ISTD.

De ovenstående resultater kan ikke anses for at være universelt gældende. LCA'er af oprensning af forurenede grunde er gennemgående lokalitets-specifikke, da mange inputs til vurderingen afhænger af grundens geografiske placering, f.eks. transportdistancer for forurenet jord og rent opfyldningsjord samt elproduktion, som afhænger af hvilket land man befinder sig i. Dybde og vandindhold samt forureningsniveauet i det oprensede jordvolumen er andre kilder til variation mellem forurenede grunde. Dertil kommer, at system- og tidsafgrænsninger samt typen af LCA (traditionel LCA eller konsekvens-LCA) har en betydning for det endelige resultat.

Livscyklusvurderinger har til formål at sammenligne miljøpåvirkninger relateret til forskellige metoder til at opnå den samme funktion eller service kaldet *den funktionelle enhed*. De fleste studier definerer den funktionelle enhed som et volumen af jord eller grundvand, der ønskes oprenset og kombinerer dette med et oprensningsmål givet ved en forureningskoncentration. Selvom to oprensnings-metoder når det samme oprensningsmål med tiden, kan deres oprensningstid være markant forskellig. Denne kvalitetsforskel kan inddrages i LCA'en ved at inkludere vurdering af de *primære miljøeffekter*. Primære miljøeffekter er lokale toksiske effekter relateret til forureningen på grunden, i modsætning til de *sekundære miljøeffekter*, som stammer fra selve oprensningsaktiviteterne.

Primære effekter er typisk blevet vurderet ved brug af *steds-uafhængige* karakteriseringsmodeller, der repræsenterer en kontinental skala og ekskluderer grundvandsmiljøet. Jordforureninger er derfor typisk blevet karakteriseret som

emissioner til overfladejord eller overfladevand. Sådanne steds-uafhængige vurderinger giver imidlertid en mangelfuld beskrivelse af kloretheners skæbne på forurenede grunde, da de ekskluderer transport til grundvandsmiljøet og antager at størstedelen af forureningen afdamper til atmosfæren.

I de to case-studier blev de primære miljøeffekter vurderet ved brug af *steds-afhængige* metoder, hvor emissionerne af forurening til grundvandet over tid blev estimeret ved hjælp af site-specifikke modeller af forureningsstoffernes transport og skæbne. Dette gjorde det muligt at tage højde for vigtige processer såsom dannelse af klorerede nedbrydningsprodukter, og at inkludere lokalitets-specifik human eksponering til forureningen via grundvandet, der anvendes som drikkevand. Ved på denne måde at inkludere primære effekter i miljøvurderingen af de forskellige alternativer til oprensning opnåedes et bedre grundlag for at sammenligne teknologier med markant forskellige oprensningstider og effektiviteter.

LCA konkluderes at være et brugbart værktøj til miljøvurdering af oprensning af forurenede grunde, selvom uløste problemstillinger fortsat findes. Blandt forhindringerne for anvendelsen af LCA som beslutningsstøtte til teknologivalg er det faktum, at metoden er meget data- og tidskrævende. Derudover kan det være svært at tolke multi-indikator-resultatet af en LCA, især i betragtning af at toksiske miljøeffekter generelt har højere usikkerhed end non-toksiske miljøeffekter. Forbedringer i karakteriseringsmetoder for toksiske effekter, samt udvidelse af LCI databaser for afværge-relaterede processer er derfor blandt de områder, der bør fokuseres på i fremtiden. Videre udvikling af metoder til værdisætning af livscykluseffekter kan desuden medvirke til at øge brugen af LCA inden for dette område, da det vil gøre det lettere at integrere resultatet af miljøvurderingen med andre beslutningsparametre som eksempelvis oprensningsomkostningerne.

# List of contents

# 1 Introduction and background

Management of contaminated soil and groundwater is a current environmental issue due to the large number of contaminated sites existing all over the world. Many of these sites pose a risk to human health and the surrounding environments and ecosystems and therefore require cleanup. In the US, US EPA (2004) estimates that a total of 300,000 sites will need cleanup during the next 35 years and on the European scale, the European Environment Agency (EEA, 2007) estimates that out of nearly 3 million sites with potentially polluting activities approximately 250,000 sites in the member countries require cleanup.

Chloroethenes, such as trichloroethene (TCE) and perchloroethene (PCE) are organic solvents that have been extensively used in all parts of the world for dry-cleaning and metal degreasing purposes. As a consequence of their widespread use and their persistency in subsurface environments, chloroethenes are among the most commonly encountered soil and groundwater contaminants. In Denmark, contamination with chloroethenes has been mapped at 19% of all sites with groundwater contamination (Danish EPA, 2006). On the US National Priority list (US EPA, 2004) chloroethenes were by far the most common group of organic contaminants at sites prioritized for remediation. TCE and PCE are rated as class 2A carcinogens by IARC (International Agency for Research on Cancer) meaning that they are probable carcinogens and vinyl chloride (VC), which is a possible degradation product from PCE and TCE, is a proven carcinogen (IARC, 2010). Consequently, allowable concentrations in drinking water are very low, e.g. 0.2-1 µg/L in Denmark (Danish EPA, 2009).

Applicable technologies for remediation of chloroethenes depend on the geological setting of the zone to be remediated. Remediation may target highly contaminated source zones in low- or high-permeability deposits or it may target a more diluted contaminant plume in groundwater. A wide range of technologies exist for both source zone and groundwater plume remediation (see Chapter 2). Examples of remediation technologies for chloroethene contamination are in situ methods such as thermally enhanced remediation, chemical oxidation methods and enhanced bioremediation methods. These methods target the contaminant in its subsurface location. Alternatively, ex situ methods imply that contaminated media (soil or groundwater) is excavated/pumped to the surface and treated on or of off-site.

## 1.1 Environmental assessment of remediation

A remediation technology removes a local contamination, but at the same time the remediation activities contribute to environmental impacts on the local, regional and global scale as they use energy, chemicals and raw materials and generate emissions and waste. Such impacts from remediation may be termed *secondary impacts* to environment as opposed to the *primary impact*s to environment related to the on-site contamination (e.g. Volkwein et al., 1999; Toffoletto et al., 2005; Cadotte et al., 2007).

Life cycle assessment (LCA) is a widely used decision support tool for environmental assessments. It is a quantitative method aimed at comparing environmental impacts related to fulfilling a defined function or service. LCA aggregates impacts occurring at all stages in the life cycle of the compared service, from raw material extraction, to production, use and final disposal (ISO, 2006a). Because remediation may result in problem-shifting, LCA can be seen as an appropriate tool for environmental assessment due to its broad scope and systems perspective (Godin et al., 2004).

LCA has been applied for environmental assessment of remediation in a number of studies reported in the literature (Lemming et al., I; Morais and Delerue-Matos, 2010; Suèr et al., 2004), but these have so far focused mostly on ex situ remediation and soil pollutants such as hydrocarbons, polycyclic aromatic hydrocarbons (PAHs) and metals. Environmental assessments of in situ remediation techniques for chloroethenes have only been dealt with sparsely in the published literature (Lemming et al., I). In addition, studies that included primary impacts in their LCA focused on impacts in either surface water (Cadotte et al., 2007; Toffoletto et al., 2005; Godin et al., 2004) or soil (Lesage et al., 2007b; Ribbenhed et al., 2002).

Chloroethenes are frequent groundwater contaminants and a spill may serve as a long-term source to groundwater contamination. However, although relevant for soil remediation, primary impacts due to groundwater contamination has not to date been included in LCA of site remediation (Lemming et al., I). This may be due to the fact that deeper soil layers and groundwater has traditionally been neglected in fate models used for characterization of toxic emissions in LCA.

Currently, there are no legislative or regulatory incentives to incorporate environmental assessments or sustainability assessments into the remediation selection process. However, an increased focus on holistic decision-making regarding remedy selection for contaminated sites remediation has been observed both in Europe and the US, where forums and networks for *sustainable* or *green*

remediation have been established. Examples of such initiatives are Sustainable Remediation Forum U.S., (SURF) Sustainable Remediation Forum UK (SuRF UK) and Green Remediation by US EPA (2008).

## 1.2 Decision support criteria for remedy selection

After remediation of a contaminated site has been decided on, a screening of applicable technologies for remediating the site is typically conducted to evaluate the options and select the most appropriate one. In this selection process, a variety of aspects encompassing technical as well as environmental and economic considerations may be included. Thus, environmental assessment as is the focus of this PhD study does not stand alone as decision support regarding contaminated site remedy selection. Cost considerations and assessments of technical applicability and performance are other obvious decision criteria as presented in Table 1. Moreover, other issues may be of high importance such as the disruption experienced by people living at or near the site. Excavation work may lead to disruptions such as noise, dust and vibrations, whereas in situ remediation may cause a smaller level of disruption, but during a longer time period.

**Table 1.** Overview of key decision parameters for contaminated site remedy selection

| Technical considerations | Environmental considerations | Economic considerations | Other considerations |
|---|---|---|---|
| • Applicability and accessibility of technology<br>• Remediation efficiency<br>• Remediation time | • Local toxic impacts at site (*primary impacts*)<br>• Environmental impacts stemming from remediation (*secondary impacts*) | • Cost of remediation | • Disruption of site residents/neighbors |

Environmental assessment of contaminated site remediation is, however, very closely linked with the technical assessments of site cleanup. The assessments of technical performance and dimensioning of a remedial action provide important inputs to the environmental assessment on technology design and dimensioning, expected remediation time and remediation efficiency. In addition, technical considerations provide inputs to the remediation cost estimates. Consequently, good-quality technical assessments are very important prerequisites for decisions regarding remedy selection.

## 1.3 Aim of the PhD project

The aim of the PhD project has been to investigate the use of life cycle assessment (LCA) for environmental assessment of contaminated site remediation. The environmental assessment is aimed to be used for decision support at the stage where a remedial action for the site has already been decided on. More specifically, focus is given to sites posing a risk for groundwater contamination with chloroethenes as these contaminants are highly relevant due to their frequent occurrence in the environment and the fact that they have received only limited attention in published literature. Two case studies were carried out in the scope of the PhD project, both of which deal with in situ remediation of chloroethene-contaminated clay tills as this type of technologies were not previously assessed in the literature. Specific objectives of the project have been to:

1. Provide an overview of existing experience and studies to date with LCA as a tool for environmental assessment of remediation (Lemming et al., I).
2. Use LCA for comparative assessments of secondary environmental impacts of remediation scenarios for a chloroethene-contaminated site including both ex situ and in situ remediation (Lemming et al., II; III).
3. Integrate assessment of local toxic impacts (primary impacts) associated with the contaminated site in the LCA by the use of site-specific models for estimation of the leaching of contaminants to groundwater under different remediation scenarios (Lemming et al., II; III).
4. Identify challenges and recommendations related to LCA used for supporting the decisions on remedy selection at chloroethene-contaminated sites threatening groundwater.

In relation to assessing local toxic impacts (primary impacts), these were narrowed down to looking at the potential human toxic impact associated with leaching of contaminants to groundwater used for drinking water purposes.

The aim of this thesis is to give an introduction to the subject of LCA and remediation and to provide an overview of the existing literature on the subject including the results of the papers attached as appendices. Based on this, challenges and recommendations for using LCA in the context of remediation of chloroethene-contaminated sites are made.

# 2 Chloroethenes and remediation options

## 2.1 Chloroethenes

Perchloroethene (PCE) and trichloroethene (TCE) are chlorinated ethenes (chloroethenes) that have been widely used, e.g. as cleaning agents for textiles and as metal degreasers in industries. Uncontrolled storage and disposal in the past has caused the contaminants to leak into the subsurface and be a threat to groundwater quality as well as indoor air quality due to their volatility.

PCE and TCE belong to the group of dense non-aqueous phase liquids (DNAPLs). DNAPLs are characterized as liquids being denser than water, immiscible with water and having a high viscosity. These characteristics enable a spill to migrate as a separate liquid phase through the subsurface leaving behind a trace of residual phase contamination trapped in the soil pores. A separate phase will due to the high density be able to migrate into deep aquifers and form pools of contamination, when an impermeable layer is reached (Stroo et al., 2003). A spill of PCE or TCE can also with time diffuse into zones of low permeability from where remediation is more complicated (Christiansen, 2010) and from where they can serve as a long-term source to groundwater contamination. Figure 1 illustrates a chloroethene spill, which has resulted in both a trace of residual phase contamination, mobile pools, diffusion into a clay matrix and formation of dissolved groundwater plumes in an upper and a lower aquifer.

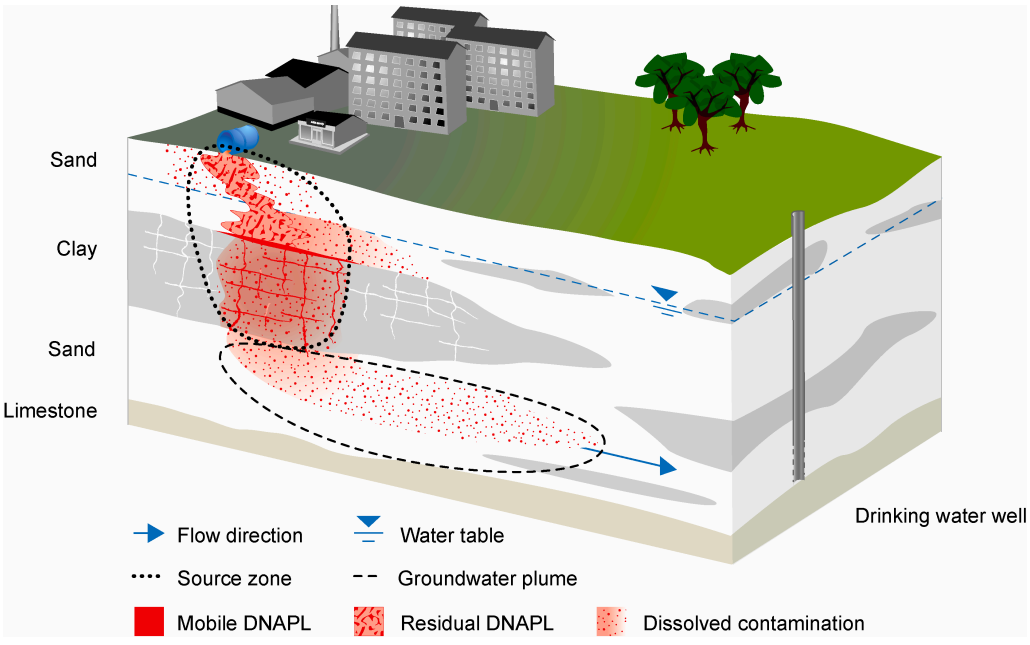


**Figure 1.** Conceptual sketch of a chloroethene spill spreading to fractured clay and forming a dissolved groundwater plume.

### 2.1.1 Degradation of chloroethenes by reductive dechlorination

PCE and TCE are fairly persistent in most natural soil and groundwater environments as they do not undergo direct oxidation under aerobic or anaerobic conditions. Under strict anaerobic conditions, they can be biodegraded via the reductive dechlorination pathway where the parent product (TCE or PCE) is dechlorinated sequentially to ethene, which is non-toxic and easily mineralized (see Figure 2). The metabolic process requires the presence of an electron donor (usually hydrogen) to reduce the chloroethenes (Aulenta et al., 2006; Christ et al., 2005; Bjerg et al., 2006).

Whereas a broad range of organisms can degrade PCE and TCE to dichloroethene (DCE) only bacteria of the genus *Dehalococcoides* has been shown to be able to degrade DCE and vinyl chloride (VC) completely to ethene by reductive dechlorination (Scheutz et al., 2008). Thus, under natural conditions, the degradation is often incomplete leading to accumulation of cis-DCE and VC (Christ et al., 2005; Bjerg et al., 2006) of which the latter is highly unwanted due to its carcinogenic properties.

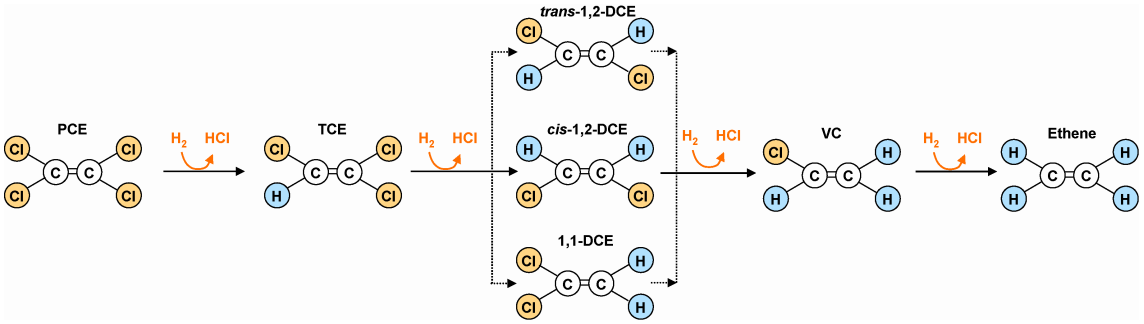


**Figure 2.** The anaerobic reductive dechlorination pathway for PCE/TCE via dichloroethene (DCE) and vinyl chloride (VC) to ethene. Cis-1,2-DCE is the predominant dichloroethene generated during this microbial process. Figure from Bjerg et al. (2006).

### 2.1.2 Toxic and carcinogenic properties

Chlorinated solvents pose a risk to humans and ecosystems due to their toxic and carcinogenic properties. The International Agency for Research on Cancer (IARC, 2010) has classified PCE and TCE as group 2A carcinogens (probably carcinogenic) and vinyl chloride, a possible metabolite, is classified in Group 1 (carcinogenic). A compilation of human toxicity data for PCE, TCE, DCE (3 isomers) and VC is presented in Table 2. The overview includes reference doses (RfD) for oral ingestion and reference concentrations (RfC) for inhalation expressing threshold values for chronic exposures. Furthermore it includes

cancer potency factors that express the lifetime likelihood of a cancer response per unit dose ingested or inhaled.

It should be noted that both PCE and TCE are currently under reassessment in US EPA's Integrated Risk Information System program (US EPA, 2010) and that the cancer potency data from US EPA are marked as provisional. The California EPA toxicity criteria database (CalEPA, 2010) also provides such data.

**Table 2.** Compilation of human toxicity data for chloroethenes from different sources

| | CAS number | Reference dose/reference concentration | | Cancer potency factor | |
|---|---|---|---|---|---|
| | | Ingestion (RfD) ($mg\ kg^{-1}\ d^{-1}$) | Inhalation (RfC) ($mg\ m^{-3}$) | Ingestion ($kg\ d\ mg^{-1}$) | Inhalation ($kg\ d\ mg^{-1}$) |
| **PCE** | 127-18-4 | 0.01 [1)] | 0.27 [3)] | 0.051*; 0.54 [2)] | 0.021 [2)] |
| **TCE** | 79-01-6 | No data | 0.6 [2)] | 0.011*; 0.013 [2)] | 0.007 [2)] |
| **1,2-cis-DCE** | 156-59-2 | 0.01 [1)] | No data | No data | No data |
| **1,2-trans-DCE** | 156-60-5 | 0.02 [1)] | 0.06 [3)] | No data | No data |
| **1,1-DCE** | 75-35-4 | 0.05 [1)] | 0.02 [1)] | No data | No data |
| **VC** | 75-01-4 | 0.003 [1)] | 0.1 [1)] | 0.75 [1,a)]; 1.5 [1,b)]; 0.27 [2)] | 0.27 [2)] |

* Provisional value (US EPA, 2000a; 200b)

[1)] Integrated Risk Information System (IRIS) (US EPA, 2010)

[2)] California EPA toxicity criteria database (CalEPA, 2010)

[3)] PPRTV: Provisional Peer Reviewed Toxicity Values, from Risk Assessment Information System (RAIS, 2010)

[a)] Continuous lifetime exposure during adulthood

[b)] Continuous lifetime exposure from birth

## 2.2 Remediation options for chloroethenes

A remediation application can be defined as a technical solution that seeks to reduce the risk associated with a contaminated site (Vik et al., 2001). Risk may in this case be defined as the probability for adverse impacts on human health and ecosystems. In addition, specific receptors such as groundwater are important drivers for remediation projects in many countries (Vik et al., 2001).

A typical site contaminated with chloroethene is illustrated in Figure 1. It consists of a subsurface source zone containing most of the contaminant mass and a groundwater plume zone with less contaminant mass. Remediation technologies can therefore either be directed at source zone remediation or groundwater plume zone remediation or a combination of these.

Remediation of contaminated soil or groundwater can either take place *in situ* or *ex situ* depending on the physical location of the treatment of the contaminated media (see Figure 3). Ex situ remediation involves excavation of contaminated soil and/or pumping of contaminated groundwater. Contaminated groundwater is typically treated on site, whereas soil is typically transported for off-site treatment and disposal. In situ remediation targets the contaminants in their actual location in the subsurface either by mass removal or mass transfer methods. Mass removal methods are true in situ methods as they destroy the contaminant directly in the subsurface, whereas mass transfer methods needs an on-site treatment system for extracted chlorinated vapors. Table 3 presents an overview of remediation techniques applicable for source zone and/or groundwater plume remediation of chloroethenes. The list of technologies in Table 3 focuses on in situ remediation methods and is not exhaustive, but serves to give an overview of technology types available for remediation of this type of subsurface contaminants. Excluded from the overview in Table 3, are the so-called containment methods, which reduce the risk associated with a contaminated site by removing the pathway between the contaminant source and receptors e.g. by capping the surface and/or installing a sheet pile wall around the contaminated soil . These methods reduce the risk of human exposure on the site, but not always the risk of groundwater contamination.

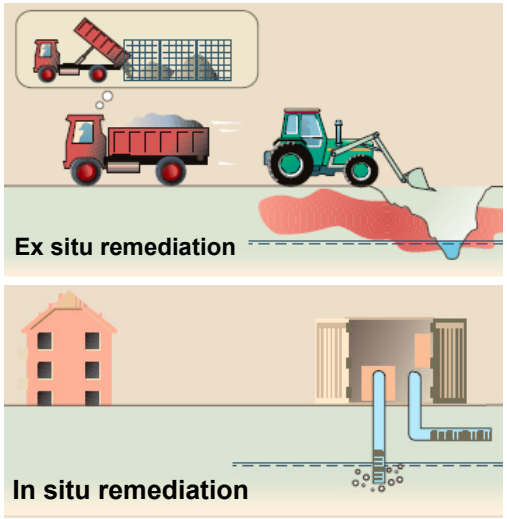


**Figure 3.** Illustration of ex situ and in situ remediation. From ScanRail Consult et al. (2000).

**Table 3.** Overview of remediation technologies for remediation of chloroethene source zones and groundwater plumes respectively.

| Remediation technology (acronym) | Relevant zone | | Source zone restrictions |
|---|---|---|---|
| | Source | Plume | |
| *Ex situ* | | | |
| Excavation | x | | Most feasible close to ground surface |
| Pump-and-treat (P&T) | | x | |
| *In situ mass removal* | | | |
| Monitored natural attenuation (MNA) | x | x | Specific degraders present |
| Enhanced reductive dechlorination (ERD) | x | x | Faster in high permeability deposits |
| In situ chemical oxidation (ISCO) | x | x | Faster in high permeability deposits |
| Soil mixing with zero-valent iron (ZVI-Clay) | x | | |
| Permeable reactive barrier (PRB) | | x | |
| *In situ mass transfer* | | | |
| Soil vapor extraction (SVE) | x | | High permeability and unsaturated |
| Multiphase extraction (MPE) | x | | Fine sand to silty sediments optimal |
| Airsparging and soil vapor extraction (AS/SVE) | | x | |
| Steam enhanced extraction (SEE) | x | | High permeability |
| In situ thermal desorption (ISTD) | x | | Faster in low permeability deposits |
| Electrical resistance heating (ERH) | x | | |

### *2.2.1 Ex situ remediation*

Excavation can in many cases be impractical, e.g. at large depth or in urban areas where lack of free space and the high level of site disruption is problematic. Pump-and-treat was earlier often used as alternative to excavation at such sites. However, due to the low water solubility and the diffusion-controlled release of contaminants sitting in low permeability matrices, chloroethene source zones can sustain a groundwater plume for decades to centuries (McGuire et al., 2006; Stroo et al., 2003).

### *2.2.2 In situ remediation*

In situ mass transfer methods include soil vapor extraction methods (SVE, e.g Hutzler et al., 1991), where soil vapor is extracted under vacuum to enhance the volatilization of chloroethenes. Airsparging (AS, e.g. Johnson, 1998) involves air injection to the saturated zone to enhance the stripping of chloroethenes to the vapor phase and is combined with SVE in the unsaturated zone to collect the vapors. Multiphase extraction (MPE, e.g. US EPA, 1999) is a mass transfer method for simultaneous extraction of soil vapor and groundwater. Thermal remediation methods have been developed to accelerate this in situ volatilization

of chloroethenes. These involve heating of the soil with either steam injection (SEE, e.g. Heron et al., 2005), conduction heating (ISTD, e.g. Heron et al., 2009) or electrical resistance heating (e.g. Beyke and Fleming, 2005).

Available in situ mass removal methods cover both chemical and biological removal processes. Chemical methods can either be in situ chemical oxidation methods (ISCO), which involve adding an oxidant such as permanganate or persulfate to the subsurface (Siegrist et al., 2001; Tsitonaki et al., 2010) or it can be chemical reduction with zero-valent iron (ZVI, Cundy et al., 2008). Enhanced reductive dechlorination (ERD, e.g Aulenta et al. 2006; Scheutz et al., 2008) is a biological remediation method for chloroethenes aimed at enhancing the anaerobic degradation pathway depicted in Figure 2 by biostimulation and most often also bioaugmentation i.e. addition of specific microorganisms able to degrade the chloroethenes. The chemical and biological removal options mentioned above are applicable both for source zone and plume remediation.

Chemical reduction with ZVI in source zones involves mixing of the soil with a clay containing ZVI and bentonite (ZVI-Clay, e.g. Wadley et al., 2005; Fjordbøge and Kjeldsen, 2010). Reduction using ZVI can also be designed as a plume treatment technology, where the ZVI is built into a permeable reactive barrier (PRB, e.g. Henderson and Demond, 2007) that captures the plume, for example by the use of a funnel construction.

In low permeability source zones deposits where the distribution of remedial amendments (e.g. oxidants for ISCO or substrate and bacteria for ERD) is more complicated, the remediation can be combined with specialized delivery methods (e.g. direct push injection or fracturing methods) for optimizing distribution and reducing diffusion time for the contaminants (Christiansen, 2010).

### *2.2.3 Remediation objectives and cleanup targets*

Remediation objectives for a contaminated site can be divided into absolute and functional objectives (ITRC, 2008). Absolute objectives describe the overall goals of remediation and represent social values such as protection of human- or ecosystem health and groundwater resources. The absolute objectives need to be accompanied by a number of functional objectives expressing how the absolute objective is achieved. In the case of groundwater protection, which is the focus here, the functional objectives define the cleanup target associated with a certain zone (source zone/groundwater plume) in quantifiable and readily measurable metrics such as contaminant concentrations (ITRC, 2008).

The need for remediation is usually decided on based on a risk assessment where the risk of contamination spreading to the groundwater at the site is assessed. In many countries the risk assessment is based on comparison of measured or model-predicted groundwater concentrations at a "point of compliance" with generic standards of maximum concentration levels (MCLs) allowed in the groundwater. The point of compliance may be located at or below the water table just below the source zone or at some specified downstream distance from the source. The exact location differs from country to country and may also be site-specific (Troldborg, 2010). The Danish guideline for risk assessment operates with a point of compliance located at a downstream distance corresponding to one year of groundwater transport or maximally 100 meters from the source (Danish EPA, 1998).

For remediation actions directed at a contaminant source zone, which is not located in the groundwater, it is beneficial to define a "short term" cleanup target that can be used as a stop criterion for termination of the remedial action in the source zone. This short term cleanup target should be defined as to ensure that the residual contaminant mass will not cause a violation of the "long-term" remedial target of complying with the groundwater MCL at the point of compliance. Figure 4 illustrates possible location of compliance points/zones for the long-term and short-term cleanup targets.

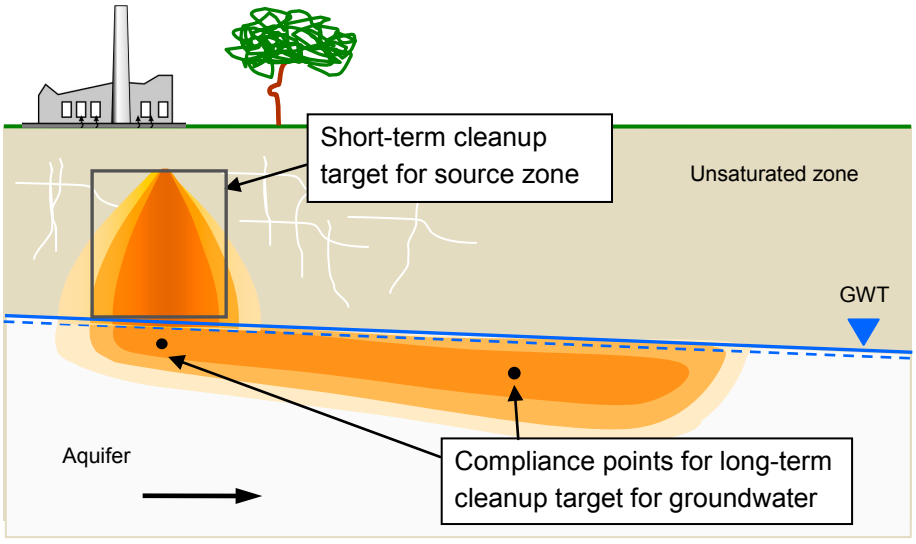


**Figure 4.** Conceptual illustration of possible locations of compliance points for cleanup targets for source zone and groundwater. Modified after Troldborg (2010).

Groundwater risk assessment usually follows a tiered approach, where the applied models depend on the knowledge level for the site. Thus, models are initially simple screening models, but with increasing site data available more detailed analytical or numerical reactive transport models taking more processes into account can be used (Troldborg, 2010).

Such detailed numerical models (e.g. Chambon et al., 2009; 2010) are also useful for establishing short-term cleanup targets for the source zone by "back-calculating" from the long-term groundwater cleanup target. Furthermore, they are essential for estimating timeframes for remediation technolgies such as ERD and ISCO in low permeability source zones, where long-term field data does not exist. This issue is further discussed in Section 5.3.

## 2.3 Management of contaminated sites in Denmark

In Denmark, groundwater is the predominant source of drinking water and therefore constitutes an essential and valuable resource. The water policy in Denmark seeks to ensure groundwater as a clean source of drinking water, which needs only a simple aeration and sand filtration before being distributed to the consumers. The aim is therefore to remediate contamination at the source to prevent deterioration of the groundwater resource. More extensive cleaning such as activated carbon filtration is only used as a last resort if no other possibilities exist for providing clean groundwater (Danish EPA, 2004). Thus groundwater protection is a main concern when remedial efforts are prioritized. Other important causes for remediation of contaminated sites are the risk of exposure to contamination via indoor air or direct soil contact.

Presently 24,000 sites in Denmark are mapped as contaminated or potentially contaminated (Danish Regions, 2010) and an additional 55,000 sites are estimated to follow within the next 40 years (Kiilerich, 2006). The total cost associated with managing these sites is estimated to approximately 2 billion euro of which around 70% is expected to be publicly financed (Kiilerich, 2006). The publicly financed remediation efforts in Denmark are managed by the five Danish regions which are responsible for mapping, investigation, prioritization and remediation of the sites. Annually, approximately 1000 site investigations are carried out and 60-100 publicly financed remedial actions are initiated in Denmark (Danish Regions, 2010). In addition to this, 400-650 sites per year are remediated based on private means (Danish EPA, 2006).

Although the annual number of publicly financed remediation projects is low relative to the total number of sites, the remediation actions consumes about

70% of the total budget spent on contaminated site management in the Danish regions (Jensen, 2010). It is therefore crucial to prioritize the remedial efforts well and ensure that the chosen remediation methods maximize the overall environmental benefit.

The management of contaminated sites currently follows a stepwise approach in Denmark as illustrated in Figure 5. After a site has been mapped as contaminated and entered the pool of contaminated sites, it will be subject to a risk assessment and prioritization phase aimed at identifying which sites that constitute a risk in terms of groundwater contamination or area use (exposure via soil contact and/or indoor air). Based on this phase, which usually requires additional site investigations, it will be decided whether the specific contaminated site needs to be remediated. If a remedial action is necessary for the site, the next step is to decide on the remediation method to use.

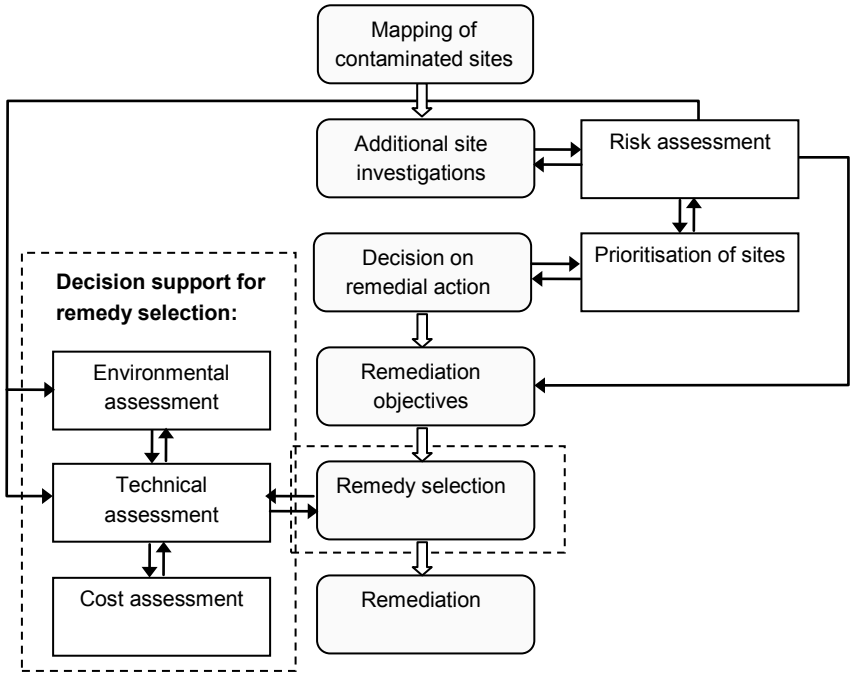


**Figure 5.** Illustration of stepwise approach for management of contaminated sites in Denmark. Environmental assessment is part of decision support for remedy selection as marked with the dotted line.

Environmental assessments as part of decision support tools as dealt with in this thesis are aimed at assessing the environmental performance of applicable remediation methods after the site has already been prioritized for a remedial action. Thus, the aim is not to decide on whether a site should be remediated or not as this question is already answered based on the risk assessment.

Currently, environmental assessments are not routinely carried out for decision support on remedy selection in Denmark. An excel-based tool for environmental assessment of remediation was developed in 2000 by ScanRail Consult et al. and was applied on a few test cases. The tool has not been updated in terms of the technologies covered and the underlying database since it was issued. Recently, another Danish screening tool for environmental assessment of remediation has been developed (RemS, Weber et al., 2009) on initiative from the Capital Region of Copenhagen, who wished to strengthen this part of their selection process.

# 3 Remediation and life cycle assessment

## 3.1 Life cycle assessment

Life cycle assessment (LCA) was chosen as the method for environmental assessment because it is a comprehensive tool covering a wide range of environmental impacts. Moreover, it takes a life cycle perspective which enables the estimation of direct as well as indirect environmental impacts related to the remediation activities being investigated. In this way environmental exchanges related to the entire lifecycle (from "cradle to grave") of each component in the assessed system are included. Furthermore, LCA is an internationally standardized method (ISO, 2006a; 2006b) and is commonly used for environmental assessments of products and services. According to the ISO standard, LCA is composed of four phases as illustrated in Figure 6: *Goal and scope definition, inventory analysis, impact assessment*, and *interpretation*. A short description of the phases is given in Figure 6.

Despite the ISO standardization of LCA, conducting a life cycle assessment is not always straightforward and the analyst is faced with a number of methodological choices such as the type of LCA to be conducted (attributional versus consequential LCA – see Section 3.2.2) and choice of impact assessment model as a wide range of models with different sophistication level exist.

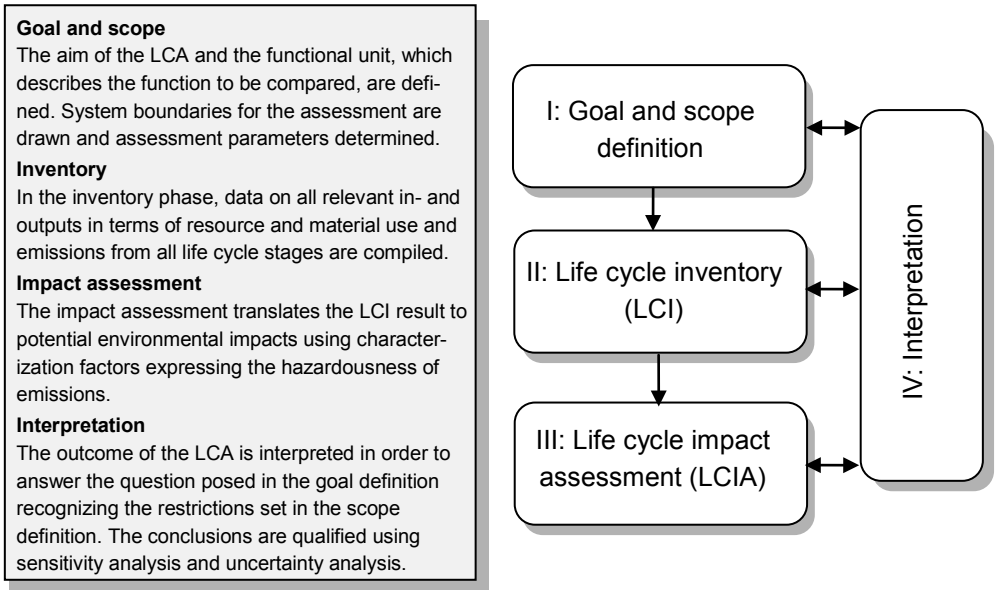


**Figure 6.** The four stages of life cycle assessment as specified in ISO 14040 (2006a) and a short description of each phase.

### *3.1.1 Conceptual framework in relation to remediation services*

A remediation system is composed of a number of direct remediation activities on- or off-site that falls into the different phases of the remediation project: installation, operation, monitoring and dismantling phase. Each of these phases requires inputs from upstream activities in terms of materials and energy and generates waste for downstream treatment as illustrated in Figure 7. The direct remediation is linked to the upstream and downstream processes via transportation processes. Furthermore, transportation processes are also included in the direct remediation activities, e.g. for monitoring or soil transportation. The waste treatment may produce energy or raw material that links back to the upstream production processes or to other life cycles that utilize these commodities.

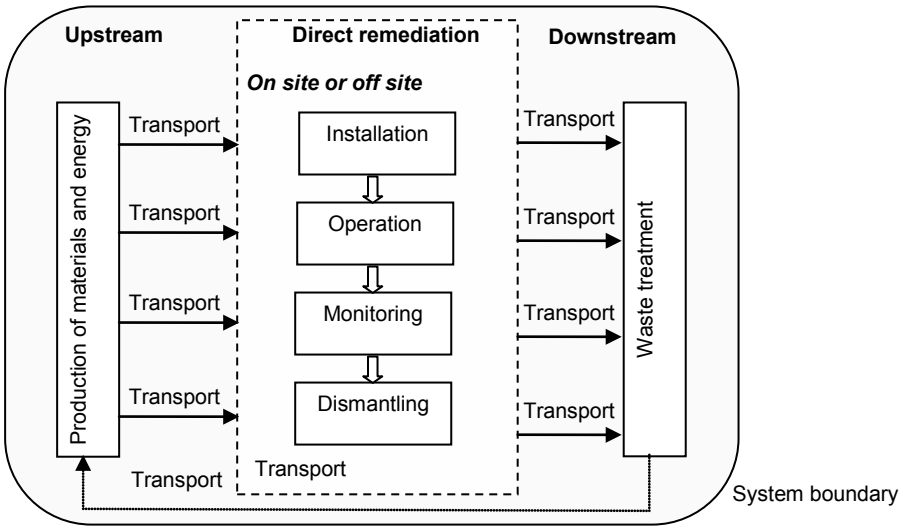


**Figure 7.** Direct and indirect activities associated with contaminated site remediation

### *3.1.2 Primary, secondary and tertiary impacts related to remediation*

A characteristic of remediation services is that they reduce the risk associated with a local contamination problem either by source removal or pathway cut off. In the literature of LCA and remediation, environmental impacts have earlier been divided into *primary* and *secondary impacts* (e.g. Volkwein et al., 1999; Toffoletto et al., 2005; Cadotte et al., 2007).

Secondary impacts are environmental impacts created due to the direct remediation processes as well as the upstream and downstream processes as illustrated in Figure 7. These impacts are distributed on numerous geographical locations both locally, regionally and globally.

Primary impacts, on the other hand, are local toxic impacts related to the on-site pollution due to its spreading to air, water and soil and subsequent

exposure of humans and ecosystems. The primary impact is accordingly highly site-specific and is the reason that remediation is carried out. In addition, land use impacts due to the land occupation associated with remediation has been included as a primary impact (Page et al., 1999) and Diamond et al. (1999) suggested inclusion of impacts on non-chemical soil quality parameters such as soil moisture and aquifer recharge.

Lesage et al. (2007a; 2007b) introduced the term "tertiary impacts" which are related to the post-remediation phase and the subsequent use of the site. The site under assessment in their study was an urban brownfield site, i.e. an idle site that is contaminated and cannot be reused for industrial or residential purposes before it is remediated. The tertiary impacts quantified covered the environmental impacts associated with developing and settling on new suburban land.

### *3.1.3 Overview of LCA studies of remediation*

Diamond et al. (1999) were the first to define a framework for using life cycle assessment and life cycle management in the environmental assessment of remediation technologies. Table 4 presents an overview of LCA case studies of contaminated site remediation published within the recent 11-year period. All 14 studies included assessments of the secondary impacts of remediation and 10 of these also included assessments of primary impacts, however with a main focus on soil and surface water impacts. Only one study included tertiary impacts.

## 3.2 Goal and scope definition

The goal and scope definition is an important phase in any LCA as it describes the aim of the LCA and defines the functional unit, which is the service or function to be compared in the study. Finally, it sets the boundaries for the assessment, which include:

- A boundary between the *technosphere* (the technological system) and the *ecosphere* (the environment), i.e. where does an environmental flow cross this boundary and becomes and emission?
- A *system boundary* defining the studied system and the included and excluded processes
- A *temporal boundary* defining the timeframe for the inventory of environmental flows

Furthermore, a choice is made about the type of LCA to be conducted (attributional or consequential, see Section 3.2.2).

**Table 4.** Overview of LCA case studies of remediation services and the impacts included, Pri: Primary impacts, Sec: Secondary impacts and Ter: Tertiary impacts. (S): Source zone remediation, (G): Groundwater plume remediation. The table is expanded from Lemming et al. I.

| Reference | Technologies included | | Contaminants | Impacts included | | |
|---|---|---|---|---|---|---|
| | | | | Pri | Sec | Ter |
| Page et al. (1999) | *Ex situ* | Excavation and disposal (S) | Lead | x | x | |
| Volkwein et al. (1999) | *Ex situ* | Excavation and on-site secured disposal (S)<br>Excavation and decontamination (S) | PAHs, mineral oil, chromium | | x | |
| | Containment | Surface sealing with asphalt (S) | | | | |
| ScanRail Consult et al. (2000) | *Ex situ* | Excavation and external biological treatment (S) | Chloroethenes, hydrocarbons | x | x | |
| | *In situ* | Biosparging (G)<br>Bioventilation (S)<br>Permeable reactive barrier (G)<br>Biological barrier (G) | | | | |
| Vignes (2001) | *Ex situ* | Pump-and-treat (on-site vacuum steam stripping) (G)<br>Pump-and-treat (on-site activated carbon treatment) (G)<br>Excavation and thermal treatment (S) | TCP and total xylenes (G)<br>Mix of organic contaminants (S) | x | x | |
| | *In situ* | Aerobic bioremediation (S)<br>Anaerobic bioremediation (S) | | | | |
| | Containment | Cap and contain (S) | | | | |
| Ribbenhed et al. (2002) | *Ex situ* | Thermal treatment (S)<br>Bioslurry (S)<br>Soil washing (S) | PAHs, mercury, cadmium | x | x | |
| Blanc et al. (2004) | *Ex situ* | Excavation and off-site landfilling (S)<br>Excavation and on-site containment (S)<br>Excavation and liming stabilization (S)<br>Excavation and bio-leaching (S) | Sulfur | | x | |
| Godin et al. (2004) | *Ex situ* | Excavation and on-site secured disposal (S)<br>Excavation and treatment (S)<br>Excavation and incineration (S) | Spent potlining landfill | x | x | |
| Toffoletto et al. (2005) | *Ex situ* | Excavation with on-site biopiles (S) | Diesel oil | x | x | |
| Bayer and Finkel (2006) | *Ex situ* | Pump-and- treat (G) | PAHs, Tar | | x | |
| | *In situ* | Permeable barrier (G) | | | | |
| Cadotte et al. (2007) | *Ex situ* | Pump-and-treat (G)<br>Excavation with on-site biopiles (S) | Diesel oil | x | x | |
| | *In situ* | Natural attenuation (S)<br>Bioventing (S)<br>Chemical oxidation (G)<br>Biosparging (G)<br>Oil removal (NAPL)<br>Bioslurping (NAPL) | | | | |
| Lesage et al. (2007a; 2007b) | *Ex situ* | Excavation and off-site disposal (S) | Metals, PAHs, hydrocarbons | x | x | x |
| | Containment | Covering with 30 cm of clean soil | | | | |
| Higgins and Olson (2009) | *Ex situ* | Pump-and-treat (G) | Chloroethenes, chloroethanes | | x | |
| | *In situ* | Permeable reactive barrier with zero-valent iron (G) | | | | |
| Lemming et al. (II) | *Ex situ* | Excavation, off-site aeration and disposal (S) | Chloroethenes | x | x | |
| | *In situ* | Soil vapor extraction (S)<br>In situ thermal desorption (S) | | | | |
| Lemming et al. (III) | *Ex situ* | Excavation, off-site treatment and disposal (S) | Chloroethenes | x | x | |
| | *In situ* | Natural attenuation (S)<br>Enhanced reductive dechlorination (S)<br>In situ thermal desorption (S) | | | | |

PAHs: Polyaromatic hydrocarbons. TCP: 1,2,3-Trichloropentane

### 3.2.1 Functional unit and time boundary

In the reviewed LCA studies of contaminated site remediation, the functional unit is typically defined as the treatment of a certain subsurface volume or mass (See Table 5). Most studies set the reference flow equal to the size of the contaminated site to be treated, whereas Lesage et al. (2007b) used a reference flow of 1 ha of brownfield. In some of the studies, the functional unit is combined with a remedial target for the site. This target is either described as an acceptable risk level (Volkwein et al., 1999; Blanc et al., 2004), a concentration-based quality criterion (Bayer and Finkel, 2006; Toffoletto et al., 2005; Cadotte et al. 2007) or a contaminant mass reduction target for the source zone (Lemming et al., III).

**Table 5.** Functional unit applied in LCA studies reported in the literature. Studies that do not define a functional unit were left out of the table. The table is expanded from Lemming et al. (I)

| Reference | Functional unit | Time boundary for LCI |
|---|---|---|
| Diamond et al. (1999), Page et al. (1999) | Production of an equivalent amount of treated soil and groundwater (mass/volume) | 25 years |
| Volkwein et al. (1999) | The ensemble of activities to achieve a certain risk level | Not specified |
| Ribbenhed et al. (2002) | 1000 kg of dry sediment into treatment | Not specified |
| Blanc et al. (2004) | A treatment of the site that allows environmental risks to be reduced to an acceptable level over the short term | Short term |
| Godin et al. (2004) | The management of 460,000 $m^3$ of wastemix and 200,000 $m^3$ of contaminated soil for a period of 50 years | 50 years |
| Toffoletto et al. (2005) | Remediation during 2-year period of 8,000 $m^3$ of diesel contaminated soil to the Quebec B criterion | 2 years |
| Bayer and Finkel (2006) | Control of a certain contaminated aquifer zone by complying with a certain concentration level. | 30 years |
| Cadotte et al. (2007) | Remediation of a 375 $m^3$ diesel-contaminated site to the Quebec B criterion in soil (700 mg $kg^{-1}$) and to the detectable limit of $C_{10}$–$C_{50}$ for potable groundwater and surface water (0.1 mg $L^{-1}$) | 2 years - 300 years depending on technology |
| Lesage et al. (2007b) | Legal and appropriate intervention on 1 ha of the tracked brownfield | 4 years [1)]<br>44 years [2)] |
| Higgins and Olson (2009) | The system-specific requirements (energy, materials) needed to provide effective capture of the contaminant plume and treatment for 30 years. | 30 years |
| Lemming et al. (II) | Treatment of the 7,500 $m^3$ of contaminated soil within a 30 year timeframe | 30 years/100 years |
| Lemming et al. (III) | Treatment of 700 $m^3$ of contaminated soil resulting in a 98% removal of the contaminant mass within this volume | Infinite/100 years |

[1)] Attributional LCA
[2)] Consequential LCA

As seen in Table 5, the time boundary varies greatly between the studies and ranges from a few years (Blanc et al., 2004; Toffoletto et al., 2005) to decades (Diamond et al., 1999; Page et al., 1999; Godin et al., 2004; Bayer and Finkel 2006; Lemming et al., II) and centuries/infinite (Lemming et al., III). In Cadotte et al. (2007) the time boundary varies from 2 to 300 years depending on the technology in question. In Lesage et al. (2007b) the timeframe of the consequential LCA was longer than for the attributional LCA in order to include tertiary impacts related to greenfield occupation during 40 years.

### *3.2.2 Attributional and consequential LCA*

A distinction can be made between two types of LCA as mentioned previously: Attributional LCA (ALCA) and consequential LCA (CLCA). An attributional LCA seeks to model the average environmental exchanges related to production of one unit of a product or good (e.g. 1 kWh of electricity). As a general rule, the modeled production technology therefore represents the specific applied technology for the direct remediation processes (e.g. the treatment activities) and the relevant average technology for the up- and downstream processes (e.g. generation of electricity or production of materials). On the contrary, consequential LCA aims to model the environmental exchanges associated with a change in demand of a product. Thus, instead of using average production technology, the *marginal* technology is used as this is the one which is predicted to respond to an increased demand of a certain good (Rebitzer et al., 2004; Ekvall and Weidema, 2004).

Some authors have suggested that CLCA is the most useful approach when the result is to be used for decision-making, whereas ALCA is suited for descriptive purposes such as accounting (Lundie et al., 2007; Tillman, 2000). However, if the uncertainties associated with the consequential modeling outweigh the benefits gained, then attributional LCA is recommended (Lundie et al., 2007). In a recently published guide to LCA (JRC, 2010a) consequential LCA is recommended for decision support on the meso or macro level related to the strategic level e.g. raw material strategies, technology scenarios and policy options. Attributional LCA is recommended for micro-level decision support related to specific products and for accounting purposes i.e. descriptive documentation of a system's (product, sector, country etc.) life cycle impacts.

Attributional LCA was used in all studies on site remediation reviewed except for Lesage et al. (2007a; 2007b), who used both an attributional and a consequential approach for the assessment of brownfield rehabilitation. In the

CLCA, Lesage et al. (2007a; 2007b) defined the alternative to rehabilitating an urban brownfield as the development of new land ("greenfield") and included the avoided impacts from greenfield development and longer transport distances to the new suburban greenfield. Lemming et al. (III) studied the effect of changing from average to marginal electricity production for a thermal remediation scenario requiring high amounts of electricity (see Section 4.3.1).

## 3.3 Life cycle inventory

The life cycle inventory (LCI) compiles all elementary flows to and from the assessed systems and is often the most time consuming part of an LCA. Generic LCI databases have been constructed in order to aid this data collection and contain data on basic products and services such as raw materials, electricity generation and transport processes, which are needed in every LCA. These databases were initially developed with a national focus only, such as the Danish EDIP database (Frees and Pedersen, 1996) and the US LCI database (NREL et al., 2004). Examples of databases with a European scope are the Swiss ecoinvent database (Frischknecht et al., 2007) and the European reference life cycle database, ELCD (EC, 2009). The majority of data in LCI databases represent average production conditions, thus representing an attributional inventory modeling approach (Finnveden et al., 2009).

Examples of databases used in the reviewed studies are EDIP (Lemming et al., II), ecoinvent (Cadotte et al., 2007; Lesage et al., 2007b; Lemming et al., III) and the US LCI database (Cadotte et al., 2007; Toffoletto et al., 2005). Despite the development of LCI databases, LCA analysts still face challenges when system-specific data are lacking in general databases. This is also the case for specific materials and services used in remediation projects. Production of activated carbon used for on-site vapor or water treatment in many remediation applications, as well as special soil amendments used for in situ remediation (microbial cultures, zero-valent iron, microbial substrates, chemical oxidants etc.) and laboratory services for soil and groundwater analyses are examples of important processes lacking in the available inventory databases. The lack of relevant processes entails a risk of important processes being excluded from the LCA. In Lemming et al (III), this lack was accommodated by collection of specific data from microbial culture producers, laboratory practices and published LCA data in the literature (activated carbon production). Zero-valent iron is another example of a material used in remediation that is not included in general databases. Higgins and Olson (2009) therefore represented it as cast iron.

## 3.4 Life cycle impact assessment

In the life cycle impact assessment (LCIA) the inputs and outputs quantified in the inventory are translated to potential impacts. According to the ISO standard (ISO 2006a; 2006b), mandatory steps of LCIA are *classification* and *characterization* of these environmental flows. The classification assigns inventoried emissions to the impact categories they contribute to and the characterization determines how much the emission contributes to each impact. The contribution to an impact is expressed by a so-called characterization factor (*CF*) that linearly relates an emitted mass (*M*) into a compartment *i* to an impact score (*IS*). The impact scores are then summed across all environmental flows *x* contributing to the given impact *m*:

$$IS_m = \sum_x \sum_i M_{x,i} \cdot CF_{x,i,m} \qquad \text{(Eq. 1)}$$

Several LCIA methodologies exist and can be divided into midpoint or endpoint models. Midpoint impact indicators are located early in the cause-effect chain, whereas endpoint indicators describe damage to the so-called areas of protection, which are human health, natural environment and natural resources (Finnveden et al., 2009). In addition, the man-made environment may also be included as an endpoint (Udo de Haes et al., 1999). Figure 8 illustrates the relation between inventory, midpoint and endpoint indicators.

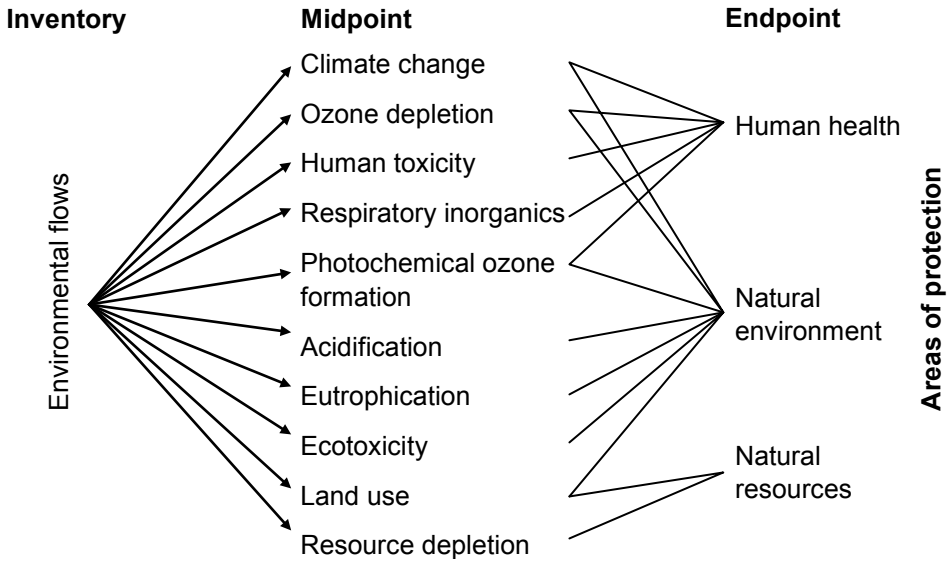


**Figure 8.** Illustration of the relation between inventory, midpoint and endpoint impact assessment, and areas of protection. Simplified after JRC (2010a).

Of the reviewed LCA studies of site remediation, all except Lesage et al. (2007b) applied midpoint characterization models. The most frequently applied midpoint model is the EDIP model, which was used in five studies (ScanRail Consult et al., 2000; Toffoletto et al., 2005; Godin et al., 2004; Lemming et al., II; Lemming et al., III). Whereas four of these studies used the EDIP97 model (Wenzel et al., 1997), Lemming et al. (III) used the updated 2003 version (Hauschild and Potting, 2005). Table 6 lists the impact categories of EDIP97 and EDIP2003 and their respective units. The impact categories covered by the two models are essentially the same, but the characterization models of EDIP2003 are updated for photochemical ozone formation, acidification and eutrophication to have a higher environmental relevance as they take the sensitivity of the environments receiving the emissions into consideration. Furthermore, some of the categories are broken into subcategories e.g. eutrophication which is divided into terrestrial and aquatic eutrophication (Hauschild and Potting, 2005).

**Table 6.** Impact categories included in EDIP97 and EDIP2003 and their respective units. Eq: equivalents

| Impact category | EDIP97 unit | EDIP2003 unit |
|---|---|---|
| Global warming | kg $CO_2$ eq | As EDIP97 |
| Ozone depletion | kg CFC11 eq | As EDIP97 |
| Photochemical ozone formation | kg $C_2H_4$ eq | Impact on human health: person·ppm·hours<br>Impact on vegetation: $m^2$·ppm·hours |
| Acidification | kg $SO_2$ eq | $m^2$ UES [a)] |
| Eutrophication | kg $NO_3^-$ | Terrestrial eutrophication: $m^2$ UES [a)]<br>Aquatic eutrophication (N): kg N eq<br>Aquatic eutrophication (P): kg P eq |
| Human toxicity air/soil/water | $m^3$ air/soil/water | As EDIP97 |
| Ecotoxicity soil/$water_{chronic}$/$water_{acute}$ | $m^3$ soil/water | As EDIP97 |
| Misc. non-renewable resources (fossil energy carriers, metals) | kg | |
| Misc. waste types | kg | |

[a)] $m^2$ UES: Area of unprotected ecosystem i.e. the area that is brought to exceed the critical load of acidification/ eutrophication.

The North American model, TRACI, was applied in two studies (Cadotte et al., 2007; Higgins and Olson, 2009). Some of the studies used more simplified and non-ISO-compliant LCIA models, for instance Vignes (2001) who excluded classification or Blanc et al. (2004) that terminated the assessment at the

inventory level. While most studies based their LCIA on one model only, Lemming et al. (III) combined the use of EDIP2003 with an LCIA model for respiratory impacts from particles (Humbert et al., 2009) and the toxicity model USEtox$^{TM}$ (Rosenbaum et al., 2008).

Lesage et al. (2007b) applied IMPACT2002+, which is an endpoint model, except for the impact climate change, which is only modeled at midpoint (in kg $CO_2$-eq.). The endpoints included are human health (in Disability adjusted life years, DALY), ecosystem quality (in PDF $m^2$ yr)[1] and resources (in MJ primary energy). Other examples of endpoint models are ecoindicator 99 (Goedkoep and Spriensma, 1999), which has been superseded by the recently launched ReCiPe model (Goedkoop et al., 2008), Lime (Itsubo and Inaba, 2003) and EPS (Steen, 1999). The latter two are examples of endpoint models including a monetary valuation step to result in a final cost in yen/euro.

Results from endpoint models can be seen as more relevant as they model the expected damage and are easier to interpret as they are restricted to fewer endpoints than the long list of impacts in midpoint models. However, at the same time, the extra steps included in the modeled cause-effect chain introduce higher uncertainty in the result as endpoint damage is not easily modeled (Hauschild, 2005).

### *3.4.1 Toxicity modeling in LCA*

Characterization models for toxic impacts of chemical emissions in LCA can roughly be divided into relatively simple fate and exposure models based on few chemical properties, e.g. EDIP97/EDIP2003 and more detailed multi-compartment, multi-pathway models that account for environmental fate, exposure and effects using a larger amount of data, e.g. IMPACT2002+ (Jolliet et al., 2003), USES-LCA (Huijbregts et al., 2000) and TRACI (Bare et al., 2003). Comparisons of toxicity characterization factors calculated with different models have shown variations up to 12 orders of magnitude for the same chemical (Rosenbaum et al., 2008). Variations stems both from difference in input data and model setup.

The EDIP model was the most frequently applied in the case studies of remediation. In the EDIP characterization model for toxicity, a simplified redistribution model calculates distribution factors $f_{x,i}$ of a chemical $x$ emitted a compartment $i$ (air, water or soil) based on the Henry's law constant and the

[1] PDF: Potentially disappeared fraction of species

atmospheric half life of the chemical. Combined with a transfer factor $T_i$, an intake factor $I_i$, a human toxicity factor *HTF,* and a biodegradation factor *BIO*, compartment-specific characterization factors for human toxicity are calculated as:

$$CF_{x,i} = f_{x,i} \cdot T_i \cdot I_i \cdot HTF_x \cdot BIO_{x,i} \qquad \text{(Eq. 2)}$$

The characterization factors represent the volume of the compartment impacted above threshold of effects per kg of chemical emitted. The human toxicity factor represents the inverse of the chemical-specific human reference dose or reference concentration (Wenzel et al., 1997).

USEtox™ (Rosenbaum et al., 2008) is a scientific consensus model, developed by a team of LCIA characterization model developers representing the modelers behind USES-LCA, EDIP, IMPACT 2002+ and CalTOX. The aim has been to enhance robustness and consistency in the calculation of toxicity characterization factors and to provide joint recommendations for characterization modeling and –factors for human and ecotoxic impacts in LCIA.

Compared to most of the other existing characterization models for toxic impacts, USEtox is simple, but compared to the EDIP model, USEtox has a larger complexity in terms of included data used as basis for the fate modeling, but also in the setup of the model world, which spans over two spatial scales (a continental scale and a global scale) each including five sub compartments (air, agricultural soil, natural soil, freshwater and marine water) that are linked via an inter-compartmental transfer model. Additionally, the continental scale includes a nested urban air box (Rosenbaum et al., 2008).

USEtox includes the modeling of "effects" based on chemical-specific dose-response data and the characterized results for human toxicity are given as cases of a cancer or a non-cancer effect respectively. For communication purposes the results are also referred to as comparative toxic units (CTU). The characterization model for human toxicity is composed of a fate factor (*FF*, in day), an exposure factor (*XF*, in $day^{-1}$) and an effect factor (*EF*, in cases $kg_{intake}^{-1}$) and is written in matrix form as (Rosenbaum et al., 2008):

$$\overline{CF} = \overline{FF} \times \overline{XF} \times \overline{EF} \qquad \text{(Eq. 3)}$$

The chemical-specific human health effect factors *(EF)* are calculated as $0.5/ED_{50}$, where $ED_{50}$ is the lifetime dose that causes an effect in 50% of the population (Rosenbaum et al., 2008).

Despite consensus-building, uncertainty of toxicity impacts is expected to remain higher than for other impacts due to the large number of chemicals involved and the complexity of the modeling e.g. in relation to synergistic chemical effects (Reap et al., 2008) and due to the fact that inventory data for chemical emissions are often incomplete (Finnveden, 2000). Furthermore, characterization factors for metal emissions often disregard important processes such as speciation and bioavailability and are therefore under further development and in USEtox marked as interim

Characterization of toxic impacts is especially important when modeling primary environmental impacts related to the local pollution on a site. The discussion on toxicity models will therefore be continued in Chapter 5 concerning primary impacts.

### *3.4.2 Normalization and weighting of impacts*

The characterized impacts on the midpoint level are given in a separate unit for each impact category, for instance kilogram of $CO_2$ equivalents (global warming) and kilogram of $C_2H_4$ equivalents (photochemical ozone formation). Normalization to person equivalents (PE) based on normalization references expressing the annual background impacts from an average person is a way to convert the different impacts to a common unit and enable a comparison of the magnitude across categories. To support comparison and aggregation across impact categories, the normalized impacts may also be multiplied with weighting factors that express the relative importance of the different impacts. Normalization and weighting are optional steps in LCA according to the ISO standards and the LCIA may be terminated at the characterized impacts.

In the EDIP methodology a weighting method for impacts based on political reduction targets for either Denmark or EU is available (Wenzel et al., 1997; Stranddorf et al., 2005). Weighting methods other than distance-to-target methods may be expert panel-defined weights or monetary weights. Weighting according to the EDIP method was applied in Godin et al. (2004), Toffoletto et al. (2005) and Lemming et al. (II).

## 3.5 Site dependency in LCIA and application to groundwater

LCA aggregates impacts occurring on multiple geographical locations. The receiving environments may differ considerably in their vulnerability to e.g. acidifying and eutrophying emissions depending on their background state and general characteristics. To account for such differences, *site-dependent* LCIA approaches have been developed through recent years in opposition to the traditional *site-generic* models. Site-dependent characterization factors are thus available on the country level for e.g. acidification and terrestrial eutrophication (Huijbregts et al., 2001; Hauschild and Potting, 2005). Characterization models for toxic impacts also include some site-dependency as they often operate with an urban and a rural box e.g. for air emissions (for example in USEtox and USES-LCA). However, the parameterization of such boxes still represents relatively large geographic scales.

LCA methodologies were initially developed for assessment of industrial products. This is reflected in the impact compartments included in LCIA models, which disregard migration of chemicals from soils to groundwater and thereby human toxic exposure through contaminated groundwater or discharge of groundwater to surface water. The development of site dependency in LCIA models has focused mostly on emissions to air, surface soil or surface water, whereas only limited focus has been given to soil-groundwater transport, which is clearly a very site-dependent matter (Hellweg et al., 2005). Hellweg et al. (2005) provided a method for a site-dependent fate assessment of heavy metal transport to groundwater from slag landfills, and Geisler et al. (2004) studied the site-dependent fate of pesticides.

## 3.6 Time horizons in LCA and long-term emissions

LCA is a methodology based on the principle of temporal justice meaning that impacts are equally important irrespective of when they occur in time. Thus, discounting (positive or negative) is typically not applied in LCA to impacts occurring in the future. However, the use of temporal cut-offs can be seen as a special type of discounting, where a discount rate of zero is applied for the time considered and a discount rate approaching infinity for the time thereafter (Hellweg et al., 2003).

Landfill emissions are examples of impacts that may be distributed over very extended time periods (>1000 years). Primary impacts from chloroethene-

contaminated sites can also have substantial timeframes associated with them, especially in relation to evaluation of no action scenarios, where no active remediation is carried out. Most of the LCA studies in the field of remediation have applied rather short timeframes (<50 years) in the assessments (see Table 5) and therefore neglect long-term emissions from landfills, e.g. from landfilling of excavated soil.

In Lemming et al. (III) an infinite timeframe was used in order to capture all impacts from landfilling as well as secondary and primary impacts of the long-term scenario "no action", which had an expected timeframe of 1200 years before leaching from the site was reduced to the remedial target. In a sensitivity scenario, the timeframe was reduced to 100 years. Figure 9 shows the normalized toxic impacts of the options "no action" and "excavation with off-site treatment and disposal", respectively, from Lemming et al. (III). The results show that a large part of the impacts occur after the initial 100-year period. For the no action option, the result is given as the sum of primary and secondary toxic impacts and the main part of both the primary and secondary human toxic impacts occurs after 100 years as this scenario has an estimated timeframe of 1200 years. For excavation, the toxic impacts are mainly related to leaching from steel slag landfills, which in the applied ecoinvent process is estimated for a 60,000 year timeframe. When long-term leaching (>100 years) is removed, the toxic impacts are therefore remarkably lower.

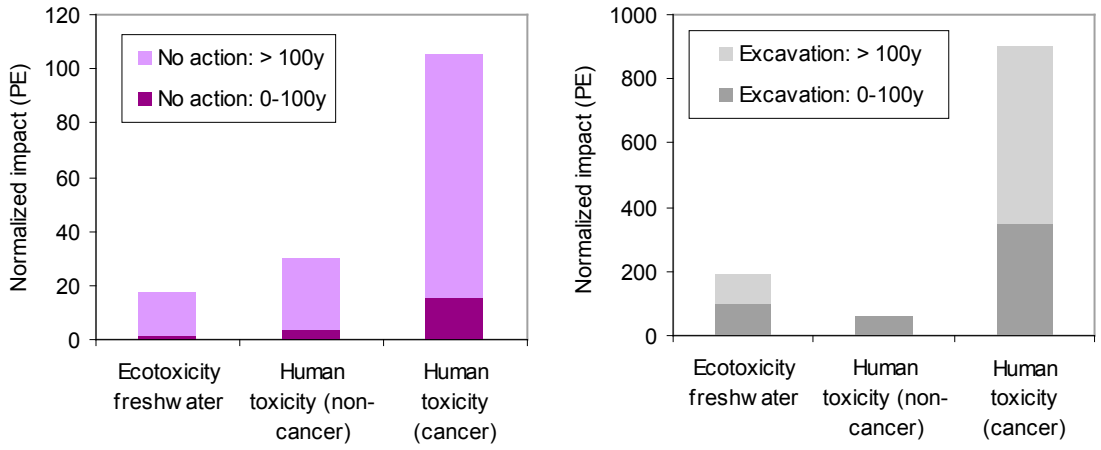


**Figure 9.** Normalized toxic impacts of the no action and the excavation scenario in Lemming et al. (III) divided into impacts occurring before and after 100 years. The result for the no action scenario is the sum of primary and secondary toxic impacts. Note the expanded scale on the figure to the right. PE: Person equivalents.

Instead of temporal cut-offs, which may lead to serious underestimation of impacts for landfills (e.g. Finnveden and Nielsen, 1999) or for long-term remediation scenarios (as seen in Figure 9), it has been suggested to make a distinction between emissions occurring in the near-term or the "foreseeable future" (< 100 years) and long-term emissions (> 100 years) (e.g. Hauschild et al., 2008; Pettersen and Hertwich, 2008). Hauschild et al. (2008) did this by introducing the new impact categories "stored human toxicity" and "stored ecotoxicity" covering toxic emissions from landfills occurring after 100 years. This procedure is also supported by the ecoinvent database, where impacts from landfills are divided into short-term emissions occurring in the first 100 years of disposal and long-term emissions occurring from 100 to 60,000 years after present, where 60,000 is an upper time boundary representing the occurrence of the next glacial period (Doka and Hischier, 2005).

## 3.7 Findings for LCA of site remediation

- A key aspect of LCA is that it compares potential environmental impacts of different options for providing the same service - the functional unit. In the reviewed studies, the functional unit is typically defined as remediation of a site to a certain contaminant level (based on concentration, mass or "risk").

- To account for differences in cleanup levels or timeframes of remediation, the assessment of secondary environmental impacts can be combined with assessment of the on-site environmental impacts from the contamination (the primary impacts) as done in several studies in the literature.

- Ideally, consequential LCA should be applied when the result is to be used for decision-making on remedy selection for contaminated sites. However, given the relatively small scale of most remediation projects and the difficulty and uncertainty in gathering marginal production data it can be recommended to investigate the sensitivity of the result by modeling only important processes such as electricity production using marginal technology.

- The inclusion of tertiary impacts in LCA of site remediation is beyond the scope of this thesis, but may be important for comparisons of remediation options, that do not ensure the same post-treatment use of the site, e.g. for residential purposes, and to evaluate whether remediation is environmentally

preferred to a no action scenario. As the prediction of such future impacts can be highly speculative it can be beneficial to involve a range of possible scenarios in the assessment of tertiary impacts.

- A variety of impact assessment models are available both for the midpoint and the endpoint evaluation level. Whereas midpoint indicators for non-toxic impacts are relatively equally modeled in-between models, modeling of toxic impacts varies greatly. USEtox$^{TM}$ is a newly developed model based on scientific consensus and is recommended in order to enhance robustness and comparison of results between studies. However, the model is still under development and some characterization factors are presently marked as interim e.g. for metals.

- Groundwater, which is an important environmental compartment especially for local emissions from contaminated sites, is not included in generic impact assessment models. This complicates primary impact assessment of chloroethene-contaminated sites leaching to groundwater.

- The timeframe of LCA of contaminated site remediation should be sufficiently long to cover both primary impacts (e.g. contaminant leaching) and secondary impacts (e.g. from landfills and long-term remediation options). A distinction between impacts occurring in the foreseeable future (< 100 years) and the far future (> 100 years) can be made, but a cut-off should be avoided as this favors options with timeframes over 100 years or options that require large amounts of landfilling.

# 4 Secondary impacts of remediation

## 4.1 Main contributing processes to secondary impacts

This chapter studies the existing LCA results concerning secondary impacts of remediation of chloroethenes. The main causes of secondary impacts from a remediation project depend on the technology applied. Table 7 gives an overview of main activities contributing to environmental impacts as found in LCA studies covering remediation of chloroethenes. Three studies dealing with other contaminants (hydrocarbons or PAHs) have been included as the covered technologies are also relevant for remediation of chloroethenes.

### *4.1.1 Ex situ remediation technologies*

Ex situ remediation involving excavation of contaminated soil and subsequent treatment was included in three studies (Lemming et al., II; III; ScanRail Consult et al., 2000). Two of these (Lemming et al., II; III) are presented in Figure 10 and Figure 11. It should be noted that the results on the figures are presented per site basis and that the soil volume treated in Lemming et al. (II) is approximately 10 times that of the case study in Lemming et al. (III) (cf. Table 5). Furthermore, the two studies use different LCIA models and databases.

Soil transportation, excavation and backfilling of soil on-site are among the main activities responsible for environmental impacts in all the mentioned ex situ studies. The ex situ soil treatment also caused significant impacts, depending on the treatment method. In Lemming et al. (II), the ex situ treatment, which involved vacuum ventilation of the soil with activated carbon treatment of extracted air, was the main contributor to many of the impact categories (see Figure 10). Due to the extensive soil cleaning in this study, the soil could subsequently be reused instead of disposed of in a landfill. In Lemming et al. (III), the soil was placed in piles and turned regularly followed by disposal in a landfill. As chloroethene is not readily biodegraded under such aerobic conditions, the removal of contaminants is only due to evaporation to air. This represents the most likely scenario for a chloroethene-contaminated soil in Denmark as no regulative thresholds exist for the reuse of it, but only for its disposal in a landfill (Hauge, 2008).

**Table 7.** Main contributors to secondary impacts of remediation of chloroethenes.

| Technology | Installation phase | | | | Operation phase | | | | | | | | | | References |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Sheet pile wall | Excavation and backfilling of soil | Wells | Funnel and gate | Electricity use on site | Diesel use on site | Remedial amendments | On-site treatment of air/water | Barrier medium (iron/activ. carbon) | Soil transportation | Off-sire soil treatment | Soil disposal | Monitoring | Dismantling | |
| **Source zone remediation** | | | | | | | | | | | | | | | |
| Excavation with ex situ aeration of soil | | x | | | | | | | | x | x | | | | Lemming et al. (II)** |
| Excavation with ex situ treatment in piles | | x | | | | | | | | x | x | | | | ScanRail Consult et al. (2000)* |
| Excavation with sheet pile wall, ex situ pile treatment and final disposal in landfill | x | x | | | | | | | | x | x | x | | | Lemming et al. (III) |
| Soil vapor extraction (SVE) | | | | | x | | | x | | | | | | | Lemming et al. (II)** |
| In situ thermal remediation (ISTD) | | | x[1)] | | x | | | x[2)] | | | | | | | Lemming et al. (II)**; Lemming et al. (III) |
| Enhanced reductive dechlorination (ERD) | | | x | | | x | x[3)] | | | | | | x | | Lemming et al. (III) |
| **Groundwater plume remediation** | | | | | | | | | | | | | | | |
| No action, only monitoring | | | | | | | | | | | | | x | | Lemming et al. (III) |
| In situ chemical oxidation with Fenton's reagent (ISCO) | | | | | | | x[4)] | | | | | | | | Cadotte et al. (2007)* |
| Permeable reactive barrier with ZVI/iron granulate | | | | x | | | | | x[5)] | | | | | | ScanRail Consult et al. (2000); Higgins and Olson (2009) |
| Permeable reactive barrier (PRB) with activated carbon | | | | x[5)] | | | | | x | | | | | | Bayer and Finkel (2006)* |
| Pump-and-treat (P&T) | | | x[7)] | | x | | | x | | | | | | | Higgins and Olson (2009); Bayer and Finkel (2006)* |

* The study does not assess remediation of chloroethenes, but has been included here as the technology is also applicable for chloroethenes. ** Results are based on background material from the study as presented partly in Figure 10.
[1)] Only in Lemming et al. (III); [2)] Only in Lemming et al. (II); [3)] Organic substrate and bioculture; [4)] Oxidant (Fentons reagent) was mentioned as most important contributor to impacts. Other contributions were not mentioned or shown; [5)] In both studies the reactive iron medium was represented as cast iron without additional processing; [6)] Both a steel funnel and mineral-based funnels were assessed; [7)] Only important in Bayer and Finkel (2006). The wells are made of PVC.

As seen in Figure 10 and 11, the ex situ treatment in piles in Lemming et al. (III) contributed less to the total impacts than the aeration applied in Lemming et al. (II), but necessitated landfilling of soil, which also generated impacts. Due to the relatively low contaminant mass held in the landfilled soil after treatment (1 mg/kg soil equivalent to approximately 1.3 kg of TCE), the landfill was modeled as an inert landfill with no emissions to groundwater. All impacts from the landfill thus stem from infrastructure and operation of the landfill (Lemming et al., III).

In one of the studies (Lemming et al., III) a sheet pile wall was necessary to support the excavation and it contributed remarkably to impacts in this study due to the large steel use, although it was assumed reused directly on two other projects. The steel production was found to be the main contributor to ecotoxic and human toxic impacts.

Three studies evaluated ex situ remediation of a groundwater plume by pump-and-treat (Higgins and Olson, 2009; Cadotte et al., 2007; Bayer and Finkel, 2006) and subsequent on-site water treatment by either activated carbon (Cadotte et al., 2007; Bayer and Finkel, 2006) or air stripping followed by catalytic oxidation of air emissions and a second water treatment step using activated carbon treatment (Higgins and Olson, 2009). In Higgins and Olson (2009) the electricity demand for pumping gave rise to the largest impacts followed by catalytic oxidation, whereas Bayer and Finkel (2006) found that the activated carbon was the major source of impacts followed by the pumping and the PVC wells. The contribution to impacts was not shown in Cadotte et al. (2007).

### *4.1.2 In situ remediation technologies*

In all studies evaluating PRB technologies for plume remediation (Higgins and Olson, 2009; ScanRail Consult et al., 2000; Bayer and Finkel, 2006), the reactive medium contained in the PRB (zero-valent iron, iron granulate or granular activated carbon) was the main cause or among the main causes of environmental impacts for the technology. Steel use for funnel and gate constructions for the PRBs was also mentioned as an important contributor to environmental impacts, especially in the study by Bayer and Finkel (2006). This study also concluded that replacing the steel-based funnel construction with a mineral-based one was favorable as it reduced the human health impacts of the PRB system significantly.

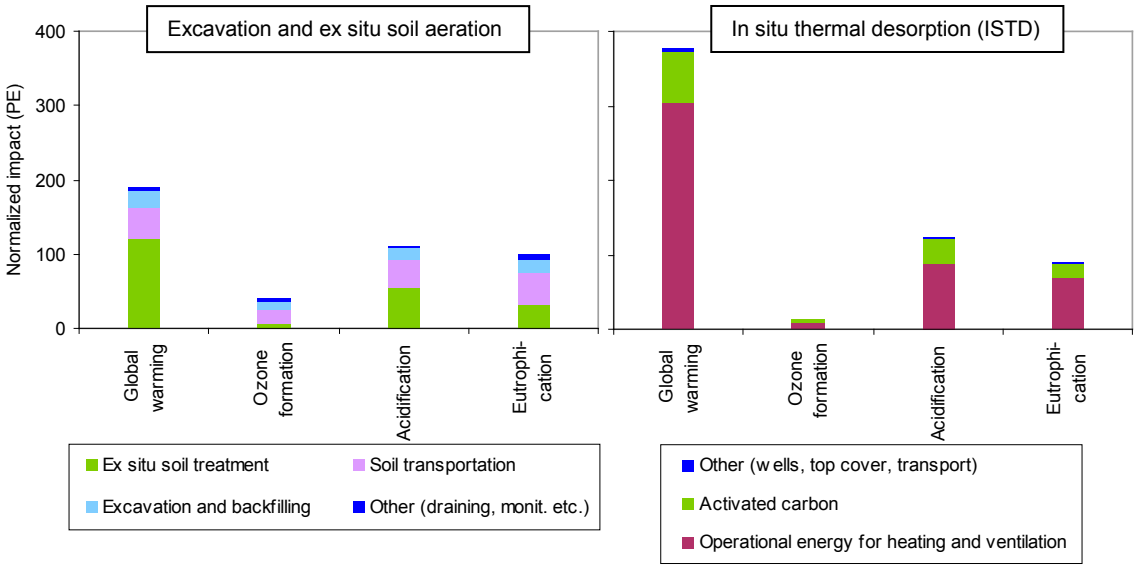


**Figure 10.** Normalized secondary impacts of excavation and ISTD option from Lemming et al. (II) (unpublished background data). Only non-toxic impacts are shown. The applied LCIA method is EDIP97 and the EDIP database was the main source of inventory data. Non-global and global impacts are normalized to person equivalents expressing the impacts from an average European (EU15, 1994) and world citizen (1994), respectively.

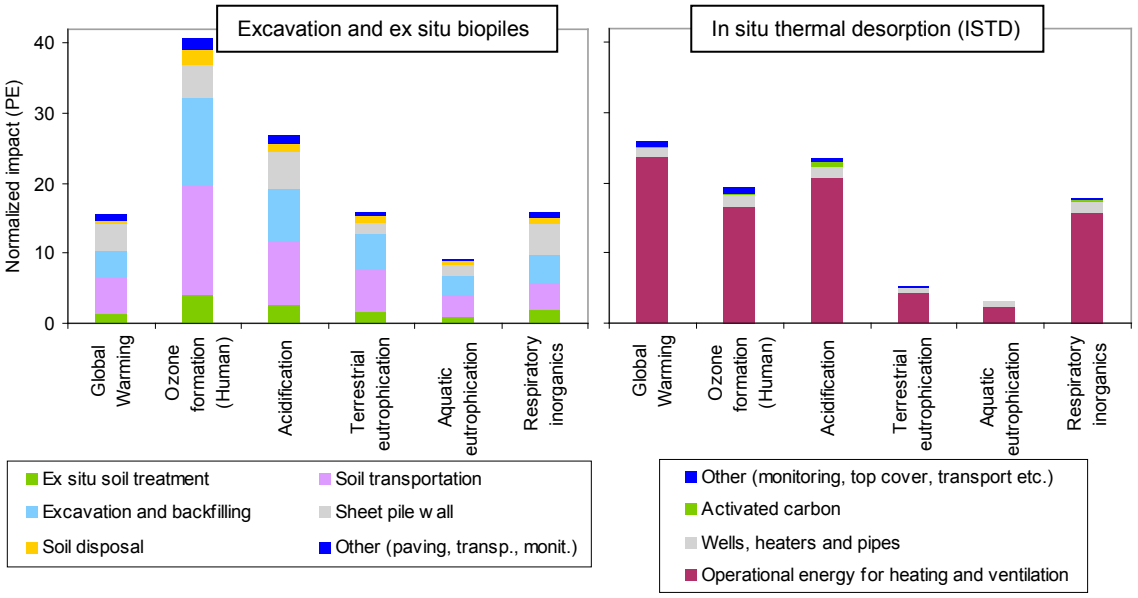


**Figure 11.** Normalized secondary impacts from the excavation and ISTD option in Lemming et al. (III). Only non-toxic impacts are shown. The applied LCIA method is EDIP2003 (Hauschild and Potting, 2005) except for respiratory inorganics, which is from Humbert et al. (2009). The ecoinvent database was the main source of inventory data. Non-global and global impacts are normalized to person equivalents expressing the impacts from an average European (EU27, 2004) and world citizen (2004), respectively.

On-site operational energy demand was the principal driver of impacts for soil vapor extraction (Lemming et al., II) and the thermally enhanced soil vapor extraction by ISTD (Lemming et al., II; III), where the soil is heated to accelerate the desorption of the chloroethenes. Figure 10 and Figure 11 show normalized impacts of the ISTD scenarios in Lemming et al. (II; III.) Besides heating, activated carbon for on-site vapor treatment also contributed significantly to impacts from SVE and ISTD in Lemming et al. (II), whereas in Lemming et al. (III), steel use for wells and heaters gave a significant contribution.

In situ bioremediation by enhanced reductive dechlorination has only been assessed in one study (Lemming et al., III) where a contaminated clay till source zone was to be remediated. The injected bioculture (6.7 tonnes of dilute solution) and organic substrate (5 tonnes of sugar cane molasses) to aid the microbial process were the main causes of impacts together with the energy demand for pumping and injection of these amendments to the subsurface. Potential methane production from anaerobic degradation of the substrate showed to be a main contributor to global warming and photochemical ozone formation (Lemming et al., III) (see Figure 12). Another finding was that monitoring activities (transport and laboratory analyses) were important due to the long time horizon associated with this technology (ca. 40 years). This was also the case for the no action scenario with a timeframe of 1200 years included in the same study by Lemming et al. (III) (see Figure 12). As mentioned by Lemming et al. (I), monitoring activities are most often neglected in LCA of remediation projects. The findings in Lemming et al. (III), however, underline that these should be included, when remediation scenarios with long timeframes are studied.

No LCA of in situ chemical oxidation (ISCO) for remediation of chloroethenes has yet been found in published literature. A study by Cadotte et al. (2007) assessed the use of ISCO using Fenton's reagent for remediation of a diesel-contaminated groundwater plume. As this technology is also applicable for chloroethene remediation, it was included in the overview in Table 7. The study concludes that the most important cause of impacts is the production and transport of the oxidant (Fenton's reagent), which was consumed in very large quantities (9,800 tonnes of $Fe_2SO_4$, 4,400 tonnes of $H_2O_2$ and 9,800 tonnes of distilled water).

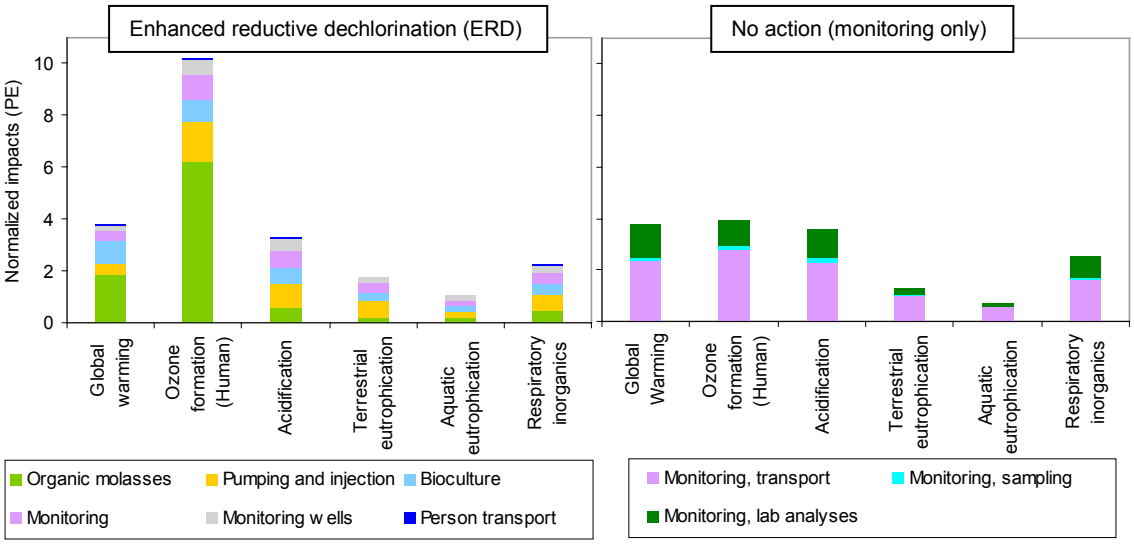


**Figure 12.** Normalized secondary impacts from the ERD and no action scenarios in Lemming et al. (III). Only non-toxic impacts are shown. The applied LCIA method is EDIP2003 (Hauschild and Potting, 2005) except for respiratory inorganics, which is from Humbert et al. (2009). The ecoinvent database was the main source of inventory data. Non-global and global impacts are normalized to person equivalents expressing the impacts from an average European (EU27, 2004) and world citizen (2004), respectively.

## 4.2 Technology rankings found in literature

Only three of the reviewed studies in the literature conducted comparative life cycle assessments of chloroethene remediation (Higgins and Olson 2009, Lemming et al., II; III). ScanRail Consult et al. (2000) also included assessment of chloroethene remediation, but did not conduct comparative assessments of several technologies for the same site. The relative ranking of the assessed technologies within each study is summarized in Table 8, which also includes a few studies that assessed technologies relevant for chloroethene remediation, but for other contaminants (Cadotte et al., 2007; Bayer and Finkel, 2006). The ranking is done by comparing results for each impact category, so that the best ranked option has the lowest impact score in the largest number of categories. Thus, all impacts are assumed of equal importance.

In Lemming et al. (II), the preference of technologies depended on the timeframe of the assessment. In the baseline assessment with a 30 year timeframe, SVE was favored over excavation and ISTD. However, as the SVE system in this case is installed in an unsaturated sand layer below the clay till source zone its timeframe will most likely be much larger as it does not remove the source only treats the amount that leaches out of the clay till. With a longer

timeframe, excavation and ISTD becomes more environmentally friendly alternatives than SVE.

**Table 8.** Relative ranking found in comparative LCA studies of technologies relevant for chloroethene remediation. For technology abbreviations, see Table 7.
A > B: A has a better environmental profile than B.
A, B: A and B have comparable environmental profiles.

| Study | Ranking | Comment |
|---|---|---|
| **Chloroethene remediation** | | |
| Lemming et al. (II) | SVE > Excavation, ISTD [1)] | Timeframe of 30 years. |
| | Excavation, ISTD [1)] > SVE | Timeframe of 100 years. |
| Lemming et al. (III) | NoA[2)], ERD > ISTD > Excavation | Infinite timeframe |
| | NoA[2)]>ERD > ISTD > Excavation | Timeframe of 100 years |
| Higgins and Olson (2009) | PRB > P&T | For treatment times of minimum 10 years |
| **Other contaminants** | | |
| Bayer and Finkel (2006) | P&T > PRB | For treatment times below 10-30 years [3)] |
| | PRB > P&T | For treatment times over 10-30 years [3)] |
| Cadotte et al. (2007) | Biosparging > P&T > ISCO | Quebec electricity mix with very low (< 2%) fossil energy content |

[1)] An internal ranking between excavation and ISTD not made as it is not straightforward as they both perform worst and best in a number of categories.
[2)] NoA (no action) was included as a reference scenario; however should not be considered as a management option due to long timeframe.
[3)] The break-even time depends on whether a mineral-based or steel-based funnel is used and which indicators that are compared.

The results from the comparative study in Lemming et al. (III) are shown in Figure 13 including sensitivity scenarios. In this study, the biological remediation method, ERD and the no action scenario, had the lowest environmental impacts. ISTD proved equal to or better than excavation for all impact categories except global warming. In the baseline scenario, a transport distance of 150 km was assumed for the excavated soil. A break-even distance was estimated at 450 km for the global warming potential. However, increasing the transportation distance would worsen the overall environmental burdens of excavation significantly compared to ISTD. Thus, from this study it can be concluded that the global warming potential cannot be universally applied as a single indicator of the overall secondary environmental impacts of a remediation technology.

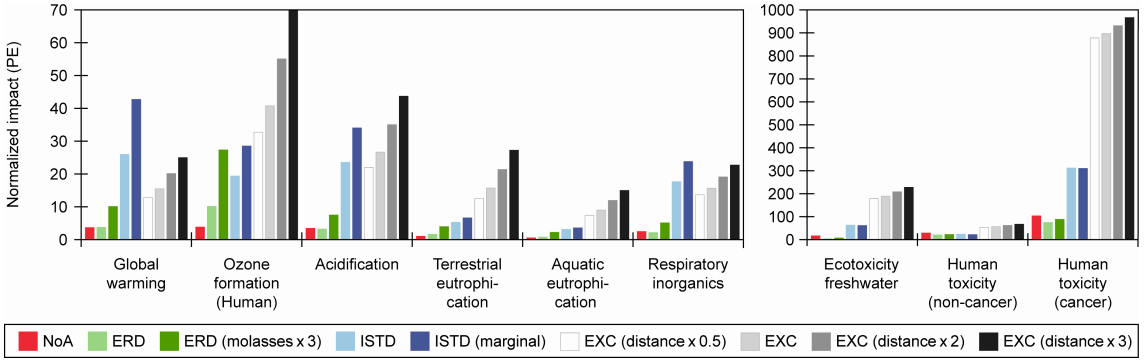


**Figure 13.** Normalized impacts of NoA (no action), ERD (enhanced reductive dechlorination), ISTD (In situ thermal desorption) and Excavation (EXC) in Lemming et al. (III). Additional sensitivity scenarios are included and show the effect of increasing the amount of organic substrate for ERD by a factor of 3, changing the electricity to marginal production (coal-based) for ISTD and changing the transportation distance of soil by a factor of 0.5, 2 and 3. It should be noted that the toxic impacts of NoA and ERD also includes primary impacts.

In a sensitivity scenario, a 100 year timeframe was used in place of the 1200 year timeframe used in the baseline scenario. This reduced the impacts of the monitored no action scenario as impacts related to the last 1100 years were cut off. Furthermore, it reduced the toxic impacts related to ISTD and excavation significantly, especially the cancer-related human toxicity and the ecotoxicity, which were strongly dependent on hexavalent chromium in leachate from steel slags landfills (illustrated for excavation in Figure 9). As mentioned in Section 3.6, ecoinvent uses a 60,000 year timeframe for landfill leaching, this allows for 25% of the chromium contained in the slags to be leached to the groundwater.

In both studies comparing groundwater plume remediation using either PRB technology or pump-and-treat, it was found that PRB was the preferred option in the long run. Break-even points were estimated at 10 years of operation (Higgins and Olson, 2009) and at 15-30 years of operation (Bayer and Finkel, 2006) depending on the type of funnel used and the impacts compared.

Remediation by ISCO was found to be the least preferred option of the groundwater plume technologies (compared to bioventing and pump-and-treat) in the study by Cadotte et al. (2007). It even had larger impacts than the scenario with 300 years of pump-and-treat. In conjunction with this it should be mentioned that the electricity for pumping in this study was represented by the average grid mix in Quebec, of which 95% is produced from hydropower and less than 2 % from fossil energy sources. Thus, if e.g. an average Danish grid mix (where approximately 70% is based on fossil energy sources) was used instead, the result might look very different. Drawing in the other direction, the

necessary amount of chemical oxidant in the study may be underestimated as it was estimated based on the stoichiometric requirement for complete mineralization of the contaminant. A significant part of oxidant will however be lost as it reacts with natural organic matter or carbonates in groundwater (ITRC, 2005). This was not accounted for in the study.

Another issue affecting the LCA of ISCO is the risk of metal mobilization in the aquifer due to changes in oxidation states or pH (ITRC, 2005). In cases where permanganate is used as oxidant, it may also contain impurities such as heavy metals (Crimi and Siegrist, 2003). Moreover, significant amounts of manganese dioxide ($MnO_2$) precipitates can form and influence the permeability of the aquifer/soil (ITRC, 2005).

Also for ERD, the application of organic substrates such as molasses or soy bean oil can cause release of metals and trace metals contained in the substrates. In Lemming et al. (III), the potential metal release from the injected sugar cane molasses was disregarded as the relatively small amounts only gave marginal contributions to the ecotoxic and human toxic effects if generic characterization factors were used. However, if site-dependent fate and exposure was taken into account, the impact may be larger. The uptake of metals during growth of the plant was furthermore disregarded in the study by Lemming et al. (III).

## 4.3 Critical issues when comparing secondary impacts

Precautions should be taken when comparing LCA studies conducted at different geographical locations, with different scopes, system and time boundaries and using different databases and impact assessment methods. Thus, the ranking of technologies presented in Table 8 can only be used as indications of the internal ranking of technologies, but cannot be assumed to apply for all sites. Some of the critical issues are elaborated below.

### *4.3.1 Site-specificity of data and results*

Firstly, LCAs of site remediation are highly site-specific. Important parameters such as transportation distances for ex situ treatments and disposal of excavated soil can vary greatly within and between countries as can transportation distances for clean backfill. As an example, in the excavation scenario in Lemming et al. (III) clean soil fill was transported 100 km to the site, while the remediated site was located right next to a gravel pit in Lemming et al. (II). Furthermore, the local conditions at a site control the setup of remediation e.g. the need of a sheet pile wall for excavation, which can increase the environmental impacts of exca-

vation significantly, especially for the toxicity-related impacts (Lemming et al., III). Furthermore, sites differ in geological and hydrogeological characteristics, which may also influence whether a sheet pile wall or extensive pumping is necessary for ISTD to reduce the flow of water to the heated zone or whether extensive pumping is necessary in conjunction with excavation or heating of a site.

Operational energy demand was an important parameter for technologies such as soil vapor extraction, thermal remediation methods and pump-and-treat as seen in Table 7. The environmental impacts from electricity production depend on the production technology (or mix of technologies) and fuel type chosen to represent the location where the electricity production takes place and varies considerably from country to country. The impact related to electricity is also dependent on whether average or marginal data is chosen to represent electricity production. In Lemming et al. (III), the effect of changing to marginal (coal-based) electricity instead of the average Danish consumption mix (38% coal, 21% natural gas, 19% renewables, 3% oil and 19% import from Sweden, Germany and Norway) was investigated (see Figure 13). The change to marginal electricity production caused an increase of 64% in global warming potential, 45% increase in ozone formation and 30% increase in respiratory effects from inorganics.

Finally, the size of the contaminated site and the amount of contaminant matters as remediation of larger sites can become more economic in terms of energy and equipment consumption per unit of soil volume treated. The area to depth relation also matters e.g. in terms of energy use for excavation or heating of the soil.

### *4.3.2 System and time delimitations*

A system delineation including a certain streamlining of the considered system is necessary to focus the analysis and the data collection. However, differences in the system delineations also influence the comparability of LCA results across studies. As previously mentioned, long-term emissions from landfills were typically neglected in the reviewed studies due to the short time boundary of the assessment. Some studies furthermore disregard the dismantling phase of the remediation technology or parts of it.

Recycling and direct reuse of materials and components are important decisions as this will often give important environmental improvements e.g. in the case of steel, which in many studies was found to be a very important

contributor to especially toxic impacts. Another type of crediting associated with the end-of-life phase was seen for activated carbon, where the energy from its incineration was credited (Lemming et al., II; III).

Another frequent simplification in the literature is disregarding the monitoring activities, due to missing data or assumed negligible impact. Results from Lemming et al. (III) show that this assumption holds for excavation and ISTD, but not for ERD and no action scenarios.

### *4.3.3 Inventory and impact assessment*

Other critical issues that cause discrepancies between LCA studies can be differences in the inventory data as well as the impact assessment method used. Discrepancies seen in results from Lemming et al. (II) and Lemming et al. (III) may for example be ascribed to the fact that they base their background data on different databases (EDIP and ecoinvent) and they use different LCIA models (EDIP97 and EDIP2003 combined with USEtox and respiratory inorganics). Thus, many impacts are modeled differently, e.g. ozone formation, acidification, eutrophication, ecotoxic and human toxic impacts. Furthermore, they use different normalization references. Lemming et al. (II) normalizes to an average European (EU15)/world citizen for 1994, whereas Lemming et al. (III) normalizes to an average European (EU27)/world citizen for 2004. It should be noted that the 2004 normalization references for EU27 are mostly lower than for the EU15 values. This is especially the case for the categories ozone formation and acidification.

### *4.3.4 Interpretation, ranking and uncertainties*

Interpretation of the LCA results and ranking of the compared technologies can be done in many ways, but in the end the ranking should be up to the decision maker, who may put weights on the individual impacts quantified in the LCA. The ranking presented in Table 8 was done simply by looking at the number of impact categories in which each technology performed the best judging from the characterized or normalized scores. This method is easy when the conclusion is clear i.e. one technology clearly has the lowest impact score in almost all categories, but less practical when the result is less clear, i.e. when one technology performs best in some categories and worst in other categories. This was the case for the comparison of excavation and ISTD in Lemming et al. (II), which therefore were difficult to rank internally using this procedure. The final

conclusion thus depends on the weights put on the different impacts by the decision maker.

When impact indicator scores are compared between remediation technologies, the uncertainties related to the results should be kept in mind. Uncertainties are related to the foreground data (site-specific data collected) as well as the background data (generic data from databases) in the inventory. Some databases (e.g. ecoinvent) include probability distributions for some inventory data given as a distribution type (e.g. lognormal) and a standard deviation. These probability distributions can then be used in a Monte Carlo analysis to estimate the uncertainty on the final result. Based on such analyses of parameter uncertainty for a case study on chemical production, Geisler et al. (2005) proposed, as a rule of thumb, that non-toxic impact scores should differ with a factor of 2 to be significantly different. No rule of thumb was proposed for toxic impacts, but the case study indicated that it may be up to two orders of magnitude. An analysis like this, however, also depends on the quality of the generic uncertainty factors in the inventory.

In addition to parameter uncertainty in the inventory phase, scenario uncertainty (choice of timeframe, boundaries etc.), model uncertainty and parameter uncertainty in LCIA are other sources of uncertainty (Huijbregts et al., 2003). Based on the inter-model variability in toxic characterization factors, Rosenbaum et al. (2008) found that the model uncertainty for human toxicity was 2 to 3 orders of magnitude and 1-2 orders of magnitude for ecotoxicity.

## 4.4 Findings for secondary impacts of remediation

- A number of LCA studies evaluating secondary impacts of remediation exist, however only few studies covered technologies relevant for chloroethene-contaminated sites. Enhanced reductive dechlorination (ERD) is promising as a low-impact technology for source zone remediation of clay till sites compared to in situ thermal remediation (ISTD) and excavation with off-site soil treatment (Lemming et al., III). Furthermore, existing studies show that permeable reactive barriers (PRBs) for groundwater plume remediation are preferred to pump-and- treat on the long term.

- As a common finding, the large environmental impact of steel was noted for technologies using it in large amounts for e.g. sheet pile walls, funnel and gate constructions in PRBs and well installations. Thus, recycling and direct reuse of steel components is highly recommended.

- Results of LCA of contaminated site remediation are very site-specific and dependent on the local conditions, the size and the contaminant levels of the site. Thus, results cannot directly be transferred from one site to another by simply scaling the inventory linearly based on the contaminated soil volume. Furthermore, system and time boundaries influence the result as do the geographical location e.g. in relation to transportation distances and electricity production technologies.

- Many technologies relevant for chloroethene remediation have not yet been assessed using LCA. Examples of these are in situ chemical oxidation (ISCO), soil mixing with zero-valent iron (ZVI-Clay) and thermal remediation using electrical resistance heating (ERH) or steam. Enhanced reductive dechlorination using other types of substrates and biocultures than in the case study by Lemming et al. (III) also has not been assessed. In order to carry out these assessments, inventories for production of several remedial amendments (particularly for ISCO, ERD and ZVI-Clay) are needed.

- The potential local impacts due to impurities such as trace metals contained in remedial amendments (e.g. substrate for ERD, oxidants for ISCO, and different zero-valent iron amendments) as well as mobilization of naturally occurring metals due to the subsurface changes is also still to be assessed with LCA.

# 5 Primary impacts of remediation

## 5.1 Conceptual model for primary impacts

As described in Section 3.1.2, in relation to environmental assessment of contaminated site remediation, the term *"primary impacts"* has been suggested to describe the potential environmental impacts related to the contamination residing at the site itself. It was noted (Lemming et al., I), that existing studies including primary impacts do this in very different manners. In the following chapter, these methods for assessing primary impacts are examined and recommendations in relation to primary impact assessment of chloroethene-contaminated sites are discussed.

In most studies, primary impacts were related solely to the impacts associated with the contamination due to exposure of humans and ecosystems. Lesage et al. (2007b) also included the negative impacts on biodiversity due to land transformation and occupation as primary impacts. These were, however, minor compared to the impacts from the residual contamination.

A conceptual model for potential primary impacts is shown in Figure 14. A contaminated site may cause human toxic impacts via direct and indirect exposures. The direct exposure routes are inhalation due to volatilization to ambient and indoor air, migration of contaminant to drinking water bodies (groundwater or surface water) and subsequent ingestion via drinking water. Other direct exposure routes are ingestion of soil, dermal contact with contaminated soil and water and inhalation of contaminants evaporating in showers. Indirect exposure routes include uptake via exposed food products (animal, vegetable and dairy products, fish etc.). Important exposure routes vary from site to site depending on the actual location and conditions of the site. For example, for urban sites without agriculture and livestock production indirect exposure via food is unimportant.

Primary ecotoxic impacts can take place in all environmental compartments where ecosystems are exposed to increased levels of toxic compounds. The compartments impacted by a contaminated site depend on the local environment surrounding the specific site. Ecotoxic impacts in groundwater have been excluded in studies considering this compartment (ScanRail Consult et al., 2000; Godin et al., 2004; Lemming et al., II; III) as the impact is expected to be minor. However, Godin et al. (2004) modeled the discharge of contaminants from groundwater to surface water and included the subsequent ecotoxic impact in surface water. As indicated in Figure 14, contaminant transfer between soil and

groundwater and between groundwater and surface water is not included in established LCIA models and needs to be modeled specifically to include them in LCA of site remediation. If a site is located within a groundwater catchment of a well field, discharge from groundwater to surface water can be disregarded as done in Lemming et al. (III) where the plume is expected to be fully captured by the groundwater extraction well.

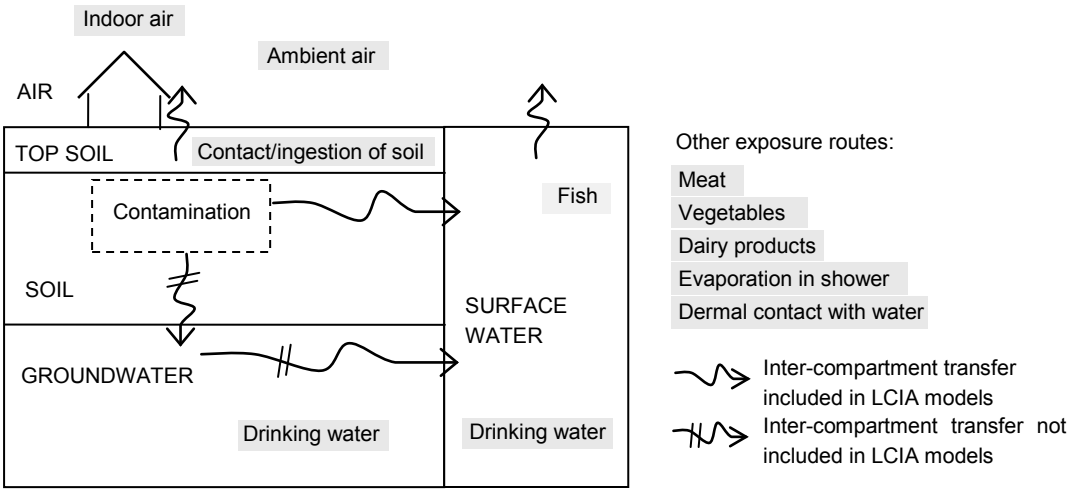


**Figure 14.** Conceptual sketch of inter-compartment transfer routes relevant for primary impacts from a contaminated site. Potential human exposure routes are marked in grey.

### *5.1.1 Technosphere and ecosphere*

As previously described in Chapter 3, environmental impacts in LCA result from the environmental flows (emissions, resource extraction) that cross the boundary between the technosphere (the technical system) and the ecosphere (the surrounding environment) (JRC, 2010a).

For primary impact assessment it is therefore important to define the border between the technosphere and the ecosphere. A common approach when evaluating primary impacts is to regard the contaminated soil volume (below a certain surface soil depth) as a part of the technosphere. This was done by Cadotte et al. (2007), Toffoletto et al. (2005), Godin et al. (2004), ScanRail Consult et al. (2000) and Lemming et al. (II, III). Thus, no impacts were accounted for within the contaminated soil volume, but only when the contaminant migrated to surface water, air or groundwater, from where there was a potential of exposure of humans and ecosystems.

If the contaminated site, however, is regarded to have a high intrinsic value e.g. due to the terrestrial ecosystem or the landscape it could be regarded as part

of the ecosphere (Payet, 2008). The ecosphere perspective was taken by Page et al. (1999), Ribbenhed et al. (2002) and Lesage et al. (2007b) who characterized the on-site contamination as an emission to soil.

## 5.2 Overview of studies assessing primary impacts

Table 9 summarizes the studies that included assessment of primary impacts in their LCA of site remediation. The studies are divided into two groups representing those that did a site-generic assessment and a site-dependent assessment of primary impacts, respectively. In addition to the studies previously mentioned in Table 4, a study by Ditor (2009) regarding sediment remediation was included here as it focuses on the comparison of site-generic and site-dependent assessments of primary impacts.

As seen in Table 9, most studies including evaluation of primary impacts used generic characterization factors to model the local impacts due to the residual contaminants at the site. Due to the lack of characterization factors for emissions to groundwater, Toffoletto et al. (2005) and Cadotte et al. (2007) used aquatic characterization factors as a proxy. Lesage et al. (2007b) and Ribbenhed et al. (2002) used terrestrial characterization factors, which did not take into consideration the possible redistribution to groundwater. Godin et al. (2004) used a groundwater transport model to estimate the discharge of contaminants to a nearby stream, but excluded impacts in the groundwater compartment as it was not used for human consumption.

### *5.2.1 Site-generic assessment of primary impacts*

Generic characterization factors are usually based on multi-compartment models parameterized to represent a very large scale (e.g. a continental scale and a global scale). The contaminant fate and exposure estimated using these models is therefore very generalized. Moreover, existing models differ widely in sophistication level and the number of compartments included. In addition to this, none of the available LCIA models include groundwater as a compartment. Thus, exemplified by the USEtox model (Rosenbaum et al., 2008), a release of TCE to the soil compartment (which has a vertical extent of only 0.1 meters) results in the majority of the mass evaporating to air (85%) and only small fractions ending up in soil and freshwater. If instead, emission to freshwater is used as a proxy for the groundwater compartment, the fraction evaporating to air is even higher (see Table 10). Thus, this distribution model (as well as other LCIA models) poorly represents the fate of a TCE at a contaminated site.

**Table 9.** Overview of studies including primary impacts and the methodologies used for assessing the contaminant fate and exposure. HT: human toxicity,; ET: ecotoxicity.

| Studies | Contaminants | Contaminant fate model for LCI [1)] | Contaminant fate and exposure models for LCIA [2)] | Primary impacts assessed |
|---|---|---|---|---|
| **Site-generic assessment** | | | | |
| Page et al. (1999) | As, Cd, Pb | Measured residual concentrations in soil | Generic – no redistribution | HT |
| Ribbenhed et al. (2002) | PAHs, Hg, Cd | Residual contaminant mass (measured) assigned as emission to soil | Generic soil CFs (USES-LCA) | HT; ET |
| Godin et al. (2004) | Spent potlining landfill (Fe, CN, Al, $F^-$) | Site-specific model of discharge to surface water | Generic aquatic CFs (EDIP97) | HT; ET |
| Toffoletto et al. (2005) | Diesel hydrocarbons | Residual contaminant mass in soil (measured) assigned as emission to surface water | Generic aquatic CFs (EDIP97) | HT; ET |
| Lesage et al. (2007a; 2007b) | Petroleum hydrocarbons | Residual contaminant mass (measured) assigned as emission to soil [4)] | Generic soil CFs (IMPACT2002+) | HT; ET |
| Cadotte et al. (2007) | Diesel hydrocarbons | Residual contaminant mass (modeled) in soil and groundwater assigned as emission to surface water | Generic aquatic CFs (TRACI) | ET |
| **Site-dependent assessment** | | | | |
| ScanRail Consult et al. (2000) [3)] | Hydrocarbons, chlorinated solvents | Contaminant mass (measured) assigned as emission to soil or groundwater | Simple steady state redistribution model including groundwater. No redistribution from groundwater. Generic exposure model (EDIP97) | HT; ET |
| Ditor (2009) | PAHs, PCB, metals | Site-specific model of emission to sediment, water and air | 1. Generic (IMPACT2002+)<br>2. IMPACT2002+ watershed model<br>3. Site-specific watershed model | HT; ET |
| Lemming et al. (II) | PCE | Site-specific model of leached mass to groundwater | No redistribution from groundwater [5)] as captured in water supply well; Site-specific exposure parameters | HT |
| Lemming et al. (III) | TCE | Site-specific model of leached mass to groundwater including sequential degradation | No redistribution from groundwater as captured in water supply well; Site-specific exposure parameters | HT |

[1)] In LCI, the contaminant fate from the technosphere to the ecosphere is modeled i.e. to the initial emission compartment.
[2)] In LCIA the contaminant fate and redistribution from the initial emission compartment is modeled.
[3)] The steady state redistribution model uses site-dependent data regarding groundwater recharge, distance to surface soil, contaminant concentrations in water and air phase and soil type.
[4)] The inventory for metal emissions was reduced to 1% of the total mass as a rough estimate for the fraction of metal ions, for which the IMPACT 2002+ CFs are valid.
[5)] 90% assumed removed to air by the aeration step in the water works

Firstly, the exclusion of the groundwater compartment as a separate compartment is a general shortcoming of LCIA models. Secondly, the large evaporation to air, may be the case just after a spill of TCE on the soil surface, but inadequately represents old sites, where the contaminant source has migrated downwards in the subsurface and maybe even diffused into areas of low permeability. Thus, for a typical chloroethene contaminated site, the fraction migrating to air seems clearly overestimated and will lead to underestimation of the primary impact as the atmospheric life time of TCE is short (half life of 4.5 days).

**Table 10.** Generic fate of TCE as modeled in USEtox.

| Final distribution of TCE in USEtox | Emission compartment | |
|---|---|---|
| | Continental natural soil | Continental freshwater |
| Air (continental) | 85% | 88% |
| Freshwater (continental) | 2% | 9% |
| Natural soil (continental) | 13% | - |
| Seawater (continental) | - | 3% |
| Remaining compartments* | - | - |

* Remaning compartments are: Air (urban), air (global), freshwater (global), natural soil (global), agricultural soil (continental), agricultural soil (global), ocean (global)

Contaminant fate in the subsurface is, however, difficult to describe using generic models as subsurface transport is very site-specific. Depth to groundwater, hydrogeological parameters and occurrence of macro pores (fractures) are examples of site-specific factors controlling the transport of contaminants. In addition to the transport-related factors, other factors such as degradation rates are important to model contaminant fate. The default degradation rates in USEtox are rather high for TCE in soil (half life of 75 days) and freshwater (half life of 38 days), which are not representative for conditions experienced at contaminated sites, where TCE is found to be very persistent especially under aerobic conditions (cf. Section 2.1.1). Under anaerobic conditions microbial degradation of TCE can occur via sequential dechlorination to ethene with DCE and VC as intermediate metabolites. Generation of these metabolites is not accounted for in USEtox or other LCIA models.

### *5.2.2 Site-dependent assessment of primary impacts*

The generic manner of modeling toxic impacts in LCA collides with the site-specific nature of primary impacts. This was acknowledged by ScanRail Consult

et al. (2000), Ditor (2009) and Lemming et al. (II; III) who included site-specific data in the modeling of contaminant fate. The latter three studies furthermore used modified exposure models to represent the local conditions at the site.

ScanRail Consult et al. (2000) introduced a relatively simple steady state model for the redistribution of contaminant from soil to groundwater and surface soil as an addition to the EDIP toxicity model. The model determines the redistribution factors based on the relative size of the upward diffusive flux of contaminant in air and the downward advective flux with infiltrating pore water, and includes site-specific data regarding contaminant concentrations in pore water and soil gas, the average distance to the surface soil, the soil type and the groundwater recharge rate. Degradation is disregarded in this model. As an example, distribution factors were calculated for the clay till site in Lemming et al. (III) contaminated by TCE. In this case, 68% of the contaminant will be redistributed to surface soil, from where it evaporates to air and only 32% will migrate to the groundwater. As this model does not include a detailed exposure and effect modeling step, site-specific data on exposed number of people, dilution etc. is not taken into account.

In Lemming et al. (II) and (III) site-dependent assessments of primary toxic impacts were carried out by including site-specific data in the contaminant fate modeling and by using local parameters to model exposure. In Lemming et al. (II) the mass discharge (g/year) of PCE to the aquifer over time was estimated from an analytical model by Troldborg et al. (2008) of the advective transport with infiltrating water assuming an equivalent porous medium and excluding biodegradation. It should be noted that the primary toxic impacts in this study were assessed differently to the secondary toxic impacts, which were evaluated using the EDIP method. The primary toxic impacts were estimated as the increased cancer probability due to ingestion of PCE-contaminated drinking water, which was then multiplied with the exposed number of people to obtain the expected number of cancer cases.

In Lemming et al. (III) a numerical contaminant transport model (Chambon et al., 2009; 2010) of the fractured clay till was used to estimate the mass discharge to the underlying aquifer. This model accounted for contaminant migration by diffusion from the clay matrix to the fractures combined with advective-dispersive vertical transport to the underlying groundwater aquifer via the fractures. Furthermore, it included microbial sequential degradation of TCE via the anaerobic reductive dechlorination pathway and accounted for formation of chlorinated degradation products. The mass discharge (g/year) of each chloro-

ethene to the aquifer was estimated using the numerical model and the subsequent exposure was estimated assuming that no further degradation took place in the groundwater during the transport to the well field. The contaminant plume was assumed fully captured in the water supply well and exposure concentrations in the abstracted drinking water were estimated by diluting the mass discharge into the annually abstracted groundwater volume. The exposure via drinking water was then estimated using human health effect factors from USEtox and using the actual number of people supplied with drinking water from the water supply.

Figure 15 displays three ways of calculating the primary impacts of the TCE-contaminated site in Lemming et al. (III): (A) a site-generic assessment where the entire contaminant mass is assumed emitted directly to the continental freshwater department in USEtox; (B) a site-generic assessment using the site-specific LCI result of the leached masses of TCE, cis-DCE and VC respectively, but applying USEtox generic characterization factors for an emission to freshwater; (C) a site-dependent assessment based on the site-specific LCI result of TCE, cis-DCE and VC emissions and using site-dependent parameters for dilution and fate of contaminants in the aquifer and for the exposed number of people. As previously mentioned, ecotoxic impacts were disregarded in the site-dependent assessment as the plume was assumed extracted in the downstream drinking water well. The results in Figure 15 clearly shows that the site-dependent assessment in this case gives much higher human toxic impacts (more than a factor of 100 higher) than found using the site-generic assessments.

The reason for this discrepancy between the site-generic and site-dependent results lies in the different fate models and in the difference in scale parameters (volume of water compartment/dilution and the exposed number of people). In USEtox, the degradation rates for TCE in soil and freshwater are much higher than those used in the site-dependent assessment. Thus, the residence time of TCE in soil and freshwater and thereby the exposure becomes low. Moreover, the generic fate model assumes that degradation removes TCE with no formation of degradation products. However, from Figure 15B and C it is evident, that actually the degradation product (VC) is responsible for the largest part of the human toxic impacts. Drinking water ingestion is in all cases assumed to be 1.4 L/day based on the default USEtox value and no further removal of contaminants is assumed for the drinking water. In the site-generic assessment, surface water is the source of drinking water and in the site-dependent assessment, groundwater is the source of drinking water.

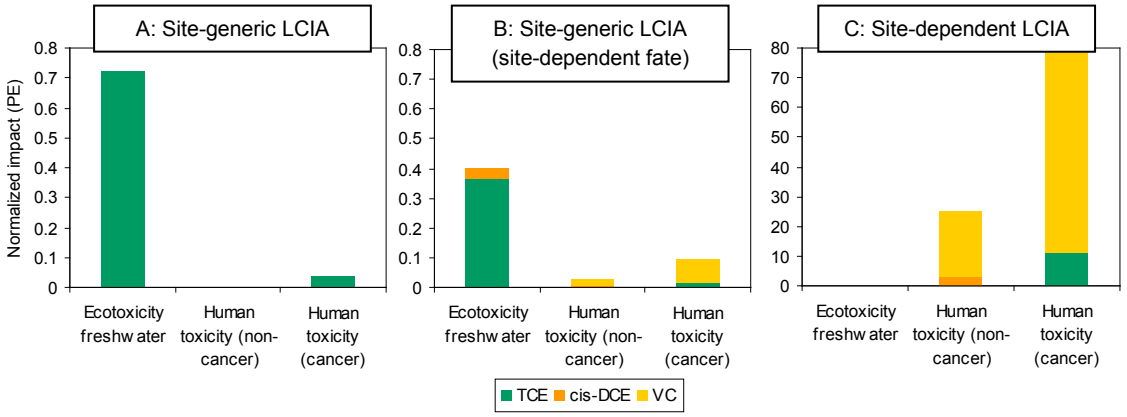


**Figure 15.** Comparison of site-generic and site-dependent assessments of primary impacts of the no action scenario for the TCE-contaminated site in Lemming et al. (III). In the site-generic assessment (A), the entire TCE mass (41 kg) is assumed emitted directly to continental freshwater in USEtox. The site-generic assessment (B) also assumes emission to the USEtox freshwater compartment, but includes the site-specific LCI result of TCE, DCE and VC emission to the aquifer (20.7, 8.8 and 2.4 kg respectively). The site-dependent assessment (C) is the original result from Lemming et al. (III), where the site-specific LCI result of emissions of TCE, DCE and VC is combined with site-specific fate and exposure parameters for the local groundwater compartment. Note the expanded scale on the figure to the right.

Ditor (2009) also studied the difference between site-generic and site-dependent assessment of primary impacts, in this case from a contaminated sediment in an estuary harbor. In this case, Ditor (2009) found that using a site-dependent assessment of primary impacts resulted in lower impacts than those of the site-generic assessment. This was mainly due to the fact that the impacted water body was not used as a drinking water source, which was assumed in the generic model. With the site-dependent assessment, the management option of monitored natural attenuation of the sediment became favorable over the remediation option due to its low primary impacts.

## 5.3 Coupling to remedial performance evaluation

Primary impact assessment deals with assessing the impacts associated with residual contamination during and after cleanup of a contaminated site. The assessment of primary impacts of contaminated site is therefore closely linked to the assessment of remedial performance of the included remediation techniques.
In Lemming et al. (III), the numerical model for contaminant fate was used both to simulate the no action with natural attenuation only and the enhanced bioremediation scenario. This made it possible to estimate the expected timeframes for reaching the remedial target as well as the amounts of

contaminants leaching to the aquifer during this period. Another advantage of using such a dynamic numerical model compared to steady state models as exemplified by the model by ScanRail Consult et al. (2000) is that it allows for evaluation of emissions of TCE, DCE and VC within a given time periods e.g. within 100 years as was done in a sensitivity scenario.

In Lemming et al. (II) a more simple approach was taken to evaluate remedial performance. Here, the performance of each technology was simply expressed as a percentage decrease in contaminant concentrations (denoted "remediation efficiency") based on literature data and was assumed to occur instantaneously. An analytical expression was used to calculate the mass discharge of PCE entering the aquifer over time. Based on the mass discharge (mass/year) to the groundwater aquifer, the exposure concentrations in abstracted drinking water were calculated by dividing with the annually abstracted volume at the downstream water supply (similar procedure as in Lemming et al., III). The uncertainty associated with the resulting exposure concentrations were investigated using first-and second-order reliability methods (FORM-SORM), which is an analytical method for forward uncertainty propagation (Ditlevsen and Madsen, 1996). Parameter uncertainty was expressed using probability density functions. The FORM-SORM calculation showed that the uncertainty associated with the remediation efficiency was the single most important parameter contributing to uncertainty of the calculated exposure concentrations for the remediation scenario, whereas source characterization parameters such as initial concentration, size of contaminated area etc. were important contributors to uncertainty in the no action scenario.

## 5.4 LCA versus risk assessment

The underlying chemical, toxicological and ecological processes used for assessment of ecotoxicity and human toxicity in LCA are to a large extent similar to those applied in methodologies for human and ecological risk assessment (here in combination referred to as RA). However, fundamental differences exist. Where LCA studies the system as a whole and addresses and aggregates a large number of impacts on different geographical and temporal scales, RA is narrower in scope focusing on a specific chemical release and its impact on individual species (Benetto et al., 2007; Udo de Haes et al., 2006). The different temporal, geographical and biological scope of ecological RA and LCA is illustrated in Figure 16.

Furthermore, RA and LCA have very different goals. LCA aims to make a relative comparison of environmental impacts of the assessed systems relying to a large extent on best estimate and average data. RA, however, aims to make a conservative assessment of the actual risk associated with the release of a chemical to a specific environment using site-specific data to model fate and exposure of the chemical (Benetto et al., 2007; Udo de Haes et al., 2006).

Site-generic ecotoxic and human toxic impact assessment is carried out for a standardized environment often representing a continental scale as discussed previously. Site-dependent primary impact assessment includes a larger amount of site-specific data in the fate and exposure modeling and therefore has a higher environmental relevance and resemblance with RA than the site-generic assessment. Yet, given the different aims of LCA and RA, a site-dependent LCA cannot replace a RA e.g. regarding decisions on whether or not a site constitutes an inacceptable risk to the surrounding environment and should be remediated. Thus, the inclusion of primary impacts in LCA does not render superfluous the use of risk assessment.

The models applied for the site-specific fate modeling in Lemming et al. (II; III) were initially developed for groundwater risk assessment purposes to be used at higher knowledge levels (cf. Section 2.2.3), and are less conservative than the proposed Danish guidelines. However, if they should be used for groundwater RA the calculated groundwater concentrations (and not the LCA impact result) should be used as a basis for comparison with groundwater MCLs to determine if the site constitutes a risk.

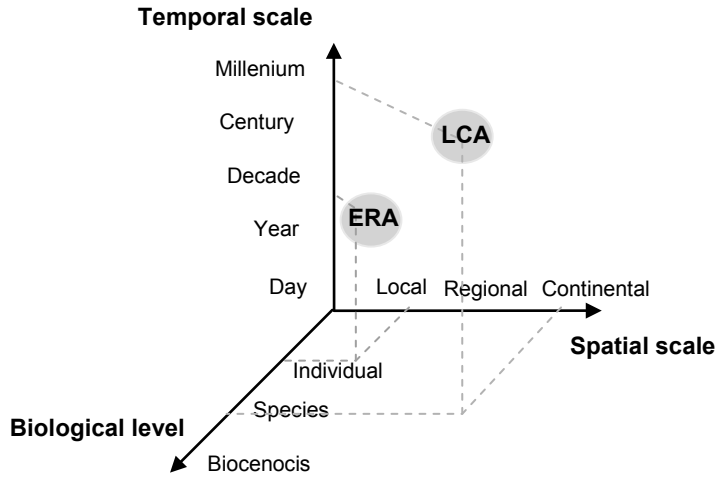


**Figure 16.** A comparison of ecological risk assessment (ERA) and LCA in terms of assessment scales (Modified after Payet, 2008)

## 5.5 Comparison of primary and secondary impacts

A clear advantage of including assessment of primary impacts in LCA of remediation projects is that it allows for a direct comparison of primary and secondary impacts as the primary toxic impacts are evaluated using the same indicators as the secondary toxic impacts and expressed in the same units.

As mentioned in Section 3.4.2 normalization of impacts can facilitate a comparison across impact categories. It should however be noted that normalization may introduce a bias between non-toxic and toxic impacts as the normalization references for toxic impacts are potentially underestimated. This is due to the fact that a very large number of chemicals are involved in the background emission scenario that is the basis of the calculation of the normalization factors. Moreover, the quality of the normalization factors suffers from incomplete chemical inventories and from lacking characterization factors for some chemicals (Heijungs et al., 2007; Sleeswijk et al., 2008).

Many of the reviewed studies including primary impact assessment did both a normalization and a weighting to a final index (Cadotte et al. 2007, Toffoletto et al., 2005; Godin et al., 2004). This procedure is good for illustrative purposes as it simplifies the result to a weighted primary and secondary impact, respectively. At the same time, however, it makes the result less transparent and may give a misleading result e.g. due to the inherent bias in normalization references and the fact that the applied weighting factors may not represent the preferences of the decision-maker.

In Lemming et al. (III) the normalized toxic impacts were generally higher than the normalized non-toxic impacts (see Figure 13), which may result from normalization bias. This was also the case for the studies by Godin et al., 2007 and Cadotte et al. (2007).

Figure 17 displays the primary and secondary human toxic impacts as quantified in the study by Lemming et al. (III). The results show that primary impacts are indeed important contributors to the overall human toxic impacts of the no action and the enhanced reductive dechlorination (ERD) scenarios, as they are responsible for 70 to 99% of the total impacts. Yet, in comparison to the secondary human toxic impacts of the in situ thermal desorption and the excavation option, the no action and ERD performs significantly better.

However, the comparison of secondary and primary toxic impacts in this study was hampered by the fact that the secondary toxic impacts were dominated to a large extent by metal releases (especially chromium) of which the characterization factors are only interim. Thus, the uncertainty of the secondary

toxic impacts was much higher than of the primary toxic impacts as these relied on recommended human toxicity effect factors and site-specific fate and exposure modeling.

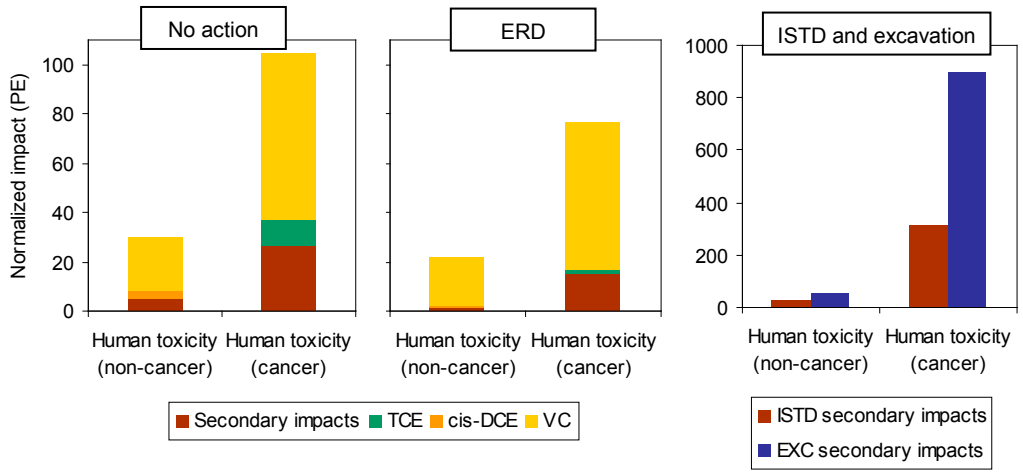


**Figure 17.** Comparison of secondary and primary human toxic impacts for no action, ERD (enhanced reductive dechlorination), ISTD (in situ thermal desorption) and excavation for the case study in Lemming et al. (III). Note the expanded scale on the figure to the right.

### *5.5.1 Monetization of impacts*

As an alternative to comparing secondary and primary impacts by their characterized, normalized or weighted impact scores on the midpoint or endpoint level, monetization can be a way to compare the primary and secondary impacts. In Lemming et al. (II), primary impacts were modeled to the endpoint expressing the number of fatalities and illnesses due to the increased cancer risk in the population from ingestion of PCE-contaminated drinking water. The Life Quality Time Allocation Index (LQTAI) (Ditlevsen and Friis-Hansen, 2007a; 2007b) was then used to obtain loss values associated with the increased risk of loss of life and health. The LQTAI model assesses human life value by requiring that any activity that reduces the life in good health must be balanced by an equivalent increase in societal productivity. Thus, it assesses the maximum productivity time that a country can allocate to avert a fatality or an injury. This time allocation can be converted to a monetary value by multiplying with a country-specific annual work-based salary based on the gross domestic product. Doing this, Lemming et al. (II) estimated loss values of approximately 11 million euro and 1 million euro for averting a cancer fatality and a 2-year disease incidence, respectively.

The monetization of secondary environmental impacts was done using a simplified approach assuming a unit cost for each weighted impact quantified in

the LCA. For environmental impacts the unit cost was based on the alternative costs for $CO_2$ reduction and for non-renewable resources the market price of oil was used as a unit cost proxy. For the case study presented in Lemming et al. (II), the monetized secondary impacts by far exceeded the monetized primary impacts of the three remediation scenarios, whereas the primary impacts of the no action scenario were comparable to the secondary impacts of remediation. Monetization of environmental impacts is inherently a very difficult task and including a monetization step, introduces additional uncertainty on top of the uncertainty already associated with the LCA result. As previously mentioned, LCA methodologies including a monetization step have already been developed, but the inherent uncertainty of such procedures might be the reason that they are rarely applied in published LCA studies. However, the advantage of monetization is that it eases interpretation of results as it merges all the impact indicators to a single monetary unit, which can easily be grasped by decision makers and compared to other costs e.g. direct remediation costs as done in Lemming et al. (II) (see Figure 18). The comparison of such costs expressing societal costs and direct economic expenses should, however, be done with caution.

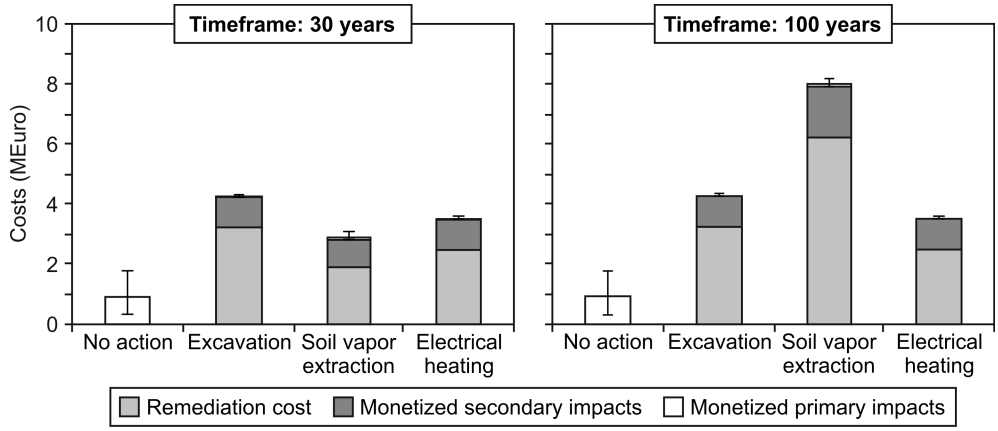


**Figure 18.** Remediation cost, monetized secondary impacts and monetized primary impacts of three remediation scenarios and the no action scenario using a timeframe of 30 and 100 years respectively. Error bars mark the 5$^{th}$ and 95$^{th}$ percentile of the monetized primary impacts. From Lemming et al. (II).

## 5.6 Findings for primary impacts of site remediation

- Primary impacts have typically been assessed using site-generic LCIA models representing a continental scale and excluding the groundwater compartment. Soil contaminants have therefore generally been assigned as emissions to surface soil or surface water compartments.

- Chloroethenes are dense liquids that have the potential of migrating to deep soil layers and groundwater. Generic fate models poorly represent the fate of chloroethenes as they exclude the migration to the groundwater compartment and do not account for formation of chlorinated degradation products of which vinyl chloride is particularly problematic in terms of toxicity and carcinogenicity.

- Site-dependent assessments of chloroethene-contaminated sites including the groundwater compartment have been suggested either by using a steady state model (ScanRail Consult et al., 2000) or dynamic analytical or numerical models of higher site-specificity (Lemming et al., II; III).

- An advantage of the dynamic models is that they can be used for evaluation of primary impacts within certain time periods, e.g. the near-term impact (0-100 years) and the long-term impact (> 100 years). Furthermore, they can be setup to include formation of degradation products and be useful for estimation of the timeframe of long-term biological remediation methods (as done in Lemming et al., III).

- The inclusion of primary impacts in LCA does not replace site-specific risk assessment as the aim and scope of the two tools are different. Including primary impacts in the LCA makes it possible to compare secondary and primary impacts using the same set of "units". This can facilitate a more fair comparison between remediation methods with substantially different timeframes for reaching the remedial target and thereby different primary impacts during cleanup.

- The assessment of primary impacts can also be used to compare the environmental impacts of a no action versus a remediation scenario to see whether remediation is an overall environmental benefit. However, such comparisons entail a discussion of the internal weighting between primary and secondary impacts.

- Lemming et al. (II) included monetization to combine secondary impacts, primary impacts and remediation costs in one measure. Although monetization of life cycle impacts has large advantages in terms of interpretability and comparability especially for non-LCA experts, the methods are still quite immature and the values assigned to the different impacts disputed, and further development and standardization of valuation methods is required.

# 6 Conclusions and perspectives

## 6.1 LCA for environmental evaluation of remediation

Environmental assessments are increasingly used in decision support for technology selection for remediation of contaminated sites. Life cycle assessment (LCA) is an environmental assessment tool that seeks to compile all relevant environmental flows during the life cycle of a product or a service and translate them into a number of environmental impacts.

This PhD thesis has investigated the use of LCA for environmental assessment of remediation of contaminated sites. This was done with a focus on chloroethene-contaminated sites and the integration of primary impacts (related to the contaminants) and secondary impacts (related to the remediation) in the LCA. Based on the thesis, including the three papers, the following key findings have been made. These findings are related to the LCA methodology as well as the applicability of LCA for decision-support for environmental assessment of remediation.

- LCA is a useful tool for environmental assessment of remediation methods for contaminated sites. Overall, LCA has the advantage that it encompasses a wide range of impacts instead of only focusing on one, e.g. global warming. This reduces the risk of burden-shifting. It can be stressed that LCA given the broad scope, large data use and uncertainty cannot be expected to provide absolute results, but rather relative results of environmental impacts associated with remediation alternatives.

- Critical issues of the goal and scope phase have been identified. The delineation of system and time boundaries for the assessment can have important implications for the result and should be stated clearly. Using a rather short timeframe (< 50 years) as done the majority of previous studies is found to be too short for chloroethene-contaminated sites that may impact the surrounding environment for centuries. It is furthermore recommendable to include a cleanup target in the functional unit to ensure that the compared options perform equally well. Alternatively, efficiency differences can be dealt with in the assessment of primary impacts.

- Life cycle assessments comparing remediation methods with different time scales and/or remediation efficiency can benefit from including primary impacts (i.e. local toxic emissions from the site) in addition to the secondary impacts (i.e. those generated by the remediation activities). This gives a more fair and complete comparison of the remediation options.

- Assessment of secondary impacts is relatively straightforward as it can be done using the existing LCA methodology. However, important choices need to be made such as choice of impact assessment model(s) and whether a consequential or attributional LCA is the aim. Furthermore, production data for many remedial amendments for in situ treatment are not available in generic LCI databases. This necessitates additional data collection or data estimation.

- Primary impact assessment has typically been done by assigning the residual contamination as an emission to soil or freshwater (surface water) and the use of generic characterization factors. However, such site-generic assessments poorly reflect the fate of chloroethenes at contaminated sites and it has been proposed to use a site-dependent assessment accounting for the site-specific transport to groundwater, degradation reactions and local exposure parameters.

- Life cycle inventories of remediation techniques are very dependent on the remediation timeframe associated with the techniques, which can be difficult to assess especially for long-term scenarios. Instead of using arbitrary timeframes in such cases it is beneficial to combine the LCA with the use of site-specific performance models as these can provide important inputs to the inventory of secondary as well as primary emissions.

- Based on a case study, in situ bioremediation of TCE by enhanced reductive dechlorination (ERD) is found to be a promising low-impact remediation option for clay till sites compared with the alternatives of in situ thermal desorption (ISTD) and excavation with off-site soil treatment. The longer timeframe of ERD compared to the other remediation options, resulted in significant primary toxic impacts during remediation. These were, however, lower than the secondary toxic impacts generated in the ISTD and excavation scenario.

- In addition to using LCA for ranking of remediation methods based on their environmental performance, an important use is identification of environmental hotspots of remediation systems. Based on these, improvement options by changing to other materials, suppliers, soil treatment site locations etc. may be identified.

- The comprehensiveness of a life cycle assessment implies that conducting an LCA requires large amounts of data and is time-consuming. Furthermore, the fact that LCA is a multi-indicator tool makes it less easy to interpret the results and use it for a ranking of remedial alternatives. However, the life cycle perspective and the inclusion of multiple indicators is also the strength of the tool. Single-indicator assessments such as "carbon footprint" (essentially equal to the global warming indicator in LCA) are more easily interpreted, but cannot be seen as an overall indicator of the environmental burdens associated with a remediation technology.

## 6.2 Future research directions

This PhD project dealt with life cycle assessment of contaminated site remediation with a focus on chloroethene-contaminated sites. The following issues for future research and investigations were identified:

- **Compilation of topic-specific inventory data and tool development.** In order to enhance the usability of LCA for decision-support for remedy selection, the data collection phase could be eased by establishment of a LCI database of production processes relevant for remediation-specific amendments and components. This data could be integrated with default technology design data that could be used for a first iteration of the secondary impacts.

- **Development of methods for primary impact assessment.** The methods for assessment of primary impacts could be further elaborated to take other exposure routes into account (inhalation, soil ingestion, dermal contact) as well as groundwater discharge to surface water. If to be integrated in a tool together with secondary impact assessment, however, the approaches should be kept simple.

- **General LCA methodology improvement.** The evaluation of toxic impacts has a high importance in LCA of site remediation. Future improvements of characterization models for toxic emissions are therefore important especially regarding heavy metals, which are currently very uncertain. Further development regarding site-dependency and integration of the groundwater compartment in LCIA models will also be beneficial for LCA of site remediation. In addition, methods for inclusion of water and land use are evolving and are also relevant for this topic.

- **Monetization and integration of environmental assessments with other decision parameters.** As mentioned in the introduction, environmental assessment is one aspect of a decision-support system for holistic remedy selection. Thus, multi-criteria methods for integration of LCA results with other decision criteria such as remedy cost, disruption of neighbors, remediation time and cleanup certainty is an issue for future attention. Further development and standardization of methods for monetization of life cycle impacts is another issue for future attention as this could enhance interpretability and be a basis for comparison with other decision criteria.

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# 8 Appendices

**I** Lemming, G., Hauschild, M.Z. and Bjerg, P.L. Life cycle assessment of soil and groundwater remediation technologies: literature review. International Journal of Life Cycle Assessment 2010, 15 (1), 115-127.

**II** Lemming G., Friis-Hansen P. and Bjerg P.L. Risk-based economic decision analysis of remediation options at a PCE-contaminated site. Journal of Environmental Management 2010, 91 (5), 1169-1182.

**III** Lemming G., Hauschild, M.Z., Chambon, J., Binning, P.J., Bulle, C., Margni, M. and Bjerg P.L. Life cycle assessment as a decision support tool for evaluation of contaminated site remediation alternatives. Submitted manuscript.

The papers are not included in this web-version, but can be obtained from the library at DTU Environment. Contact library@env.dtu.dk or Department of Environmental Engineering, Technical University of Denmark, Miljoevej, Building 113, DK-2800 Kgs. Lyngby, Denmark.

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