SUPPORTING INFORMATION

Environmental Impacts of Remediation of a Trichloroethene-contaminated Site: Life Cycle Assessment of Remediation Alternatives

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A. Life cycle inventory

A.1 Overview of included activities and estimations made

Inventory data for the remediation systems were based on modeling and the input from consultants and contractors likely to undertake the work on the site. Enhanced reductive dechlorination (ERD) has already been initiated at the study site and inventory data for this scenario is largely based on the actual site data. An overview of activities included in the life cycle inventory of each remediation system is given in Table S1, which also lists the approximate duration of each remediation phase. Specific inventory data and estimations and assumptions made are provided in Table S2 and Table S3. The ecoinvent processes applied for the LCA modeling using the software SimaPro (version 7.1.8) are presented in Table S4.

Phase	NoA	ERD	ISTD	EXC
Installation Approx. duration		6 monitoring wells in upper aquifer (5 m deep) 4 monitoring wells in regional aquifer (10 meter deep) 5 site visits Transport of materials and drill rig to site (5 days)	20 heater wells 20 extraction wells 20 heaters 10 temperature sensors 560 kg expanded polystyrene insulation 60 m aboveground steel gas collection pipes 15 site visits Transport of materials and drill rig to site (15 days)	Removal of 140m^2 pavement Installation of sheet pile wall (32 tons steel) 5 site visits Transport of machinery and materials to site (5 days)
Operation Approx. duration		5000 kg sugar cane molasses transported by ship from Brazil 6700 kg bioculture by truck from the Netherlands 35 site visits Mixing, pumping and injection at 56 injection points for each injection round Transport of Geoprobe (5 days per injection round, 7 injection rounds)	Heating of soil Ventilation of soil Water pumping 260 kg activated carbon 30 site visits (~3 months/100 days)	Excavation and backfilling of soil (1120m^3) 20 site visits Off site soil treatment Soil disposal Transportation of soil to treatment (700m^3, 100km) Transportation of soil to disposal (700m^3, 50km) (~ 1 month at site)
Monitoring Approx. duration	1200 site visits 6000 water samples 6000 lab analyses (1200 years)	52 site visits 232 water samples 10 soil samples 232 lab analyses (water) 50 lab analyses (soil) Transport of soil sampler (38 years)	8 soil samples 30 lab analyses (soil) 1 site visit Transport of soil sampler (1 day)	8 soil samples 181 lab analyses (soil) 1 site visit Transport of soil sampler (1 day at site, 1 day at soil treatment center)
Dismantling				Asphalting (140m^2) Removal of sheet pile wall
Primary impacts	Leaching of TCE and degradation products	Leaching of TCE and degradation products	None	None

Table S1. Overview of activities included in the modeling of each of the 4 systems. Upstream processes such as production of components, energy, infrastructure and transport are not mentioned specifically, but are also included in the LCA

Relevant for technologies	Description of estimation/assumption made
NoA, ERD, ISTD, EXC	Distance of round trip for consultants for supervision and monitoring is set to 90 km based on the actual distance between the site and the consultant.
NoA, ERD, ISTD, EXC	The transportation distance of machinery and materials used at site was set to 100 km (roundtrip) to represent transport in Denmark. Transportation of steel, plastic, concrete and asphalt from regional storage to Denmark was added to the ecoinvent processes using standard transport distances from (*1*)
NoA, ERD, ISTD, EXC	Laboratory analyses: Energy and material use and emissions from groundwater and soil analyses was estimated from CIRAIG laboratory practices and standard analytic method descriptions
ERD, EXC, ISTD	Transportation of sand and gravel from Danish gravel pits for backfill and well screens set at 100 km.
ERD, ISTD	Energy for uptake of wells (monitoring wells/heating wells) after termination assumed negligible
NoA	No new groundwater wells are constructed. Monitoring takes place in existing wells
ERD	*Material use for monitoring wells (5 m deep in upper aquifer and 10 m deep in regional aquifer):* Polyethylene (PE) use per meter depth of pipe ($Ø_{outer}$63 mm, $Ø_{inner}$51.4 mm, density 960 kg/m^3): 1 kg/m Bentonite use per well in upper aquifer (4.5 m depth, Ø150 mm, density 950 kg/m^3): 76 kg Bentonite use per well in regional aquifer (8 m depth, Ø150 mm, density 950 kg/m^3): 111 kg Screen sand use per well in upper aquifer (1.5 m screen): 33 kg Screen sand use per well in regional aquifer (2 m screen): 44 kg Steel use per well cover plate: 5 kg
ERD	Organic sugar cane molasses from Brazil and bioculture produced in the Netherlands was selected as electron donor and bioaugmentation culture respectively based on what was actually used at the site. The amount of bioculture needed was based on the bioculture-to-molasses ratio (3.3) actually applied for the first injection at the site. Thus it is assumed that 2000 kg of molasses and 6700 kg of bioculture is added in the first injection round. The remaining 3000 kg of molasses are divided between the remaining 6 injection rounds. See molasses demand calculation in section A4.
ERD	The inventory of bioculture production (*2*) includes energy consumption, organic additives (0.1-0.5 g/L of lactate and acetate) and inorganic additives (1 g/L of N, P, Mg, Ca) as seen in Table S4. The organic and inorganic additives were represented by acetic acid and ammonium nitrate phosphate respectively, which were the closest substances available in ecoinvent. The contribution to impacts from these additives was found to be negligible.
ERD	The density of the bioculture is set equal to water (1 kg/L) as it is very dilute. The mass-based content of bacteria is approximately 0.0042 g/kg of culture (1E12 cells/L, 4.2E-15 g/cell). Due to the very low organic content of the bioculture, the methane generation potential from the anaerobic degradation of it is omitted in the analysis.
ERD	The ecoinvent process for cane sugar includes a negative emission (i.e. an uptake) of metals from the agricultural soil to the sugar plants. However, as this uptake can vary substantially depending on local soil properties and because the metals will be released again when the molasses is amended to soil, we simplified the analysis by excluding the uptake as well as the subsequent release of metals. The approximate metal concentrations in molasses based on information from supplier are: Ca: 5.4 g/kg; Na: 0.7 g/kg; K: 49 g/kg; Mg: 4 g/kg; Fe: 150 mg/kg; Cu: 4.6 mg/kg; Mn: 13 mg/kg; Zn: 7.8 mg/kg; Se: 0.4 mg/kg. The human toxic and ecotoxic effects of these potential emissions were evaluated with USEtox assuming that they are emitted directly to freshwater. The calculated impact indicates that these metal emissions are negligible for the ERD result. However, a better assessment of these local effects would require a site-specific model of fate, and use of site-specific exposure parameters as done for the fate of chlorinated ethenes.

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ERD	The end-of-life stage for PE pipes (50 kg) from monitoring wells is not included due to negligible

	impact
ERD	Estimated weight of mixing tank: 100 kg. Not included in inventory except for transport to site as assumed many times
ISTD	Steel use for wells, filters and heaters based on data from (*3,4*) *Material use per meter of heater well (total depth 8 meter).* Steel use for outer pipe: (Ø80 mm, thickness 4 mm, density 7850 kg/m^3): 7.5 kg/m. Stainless steel use for inner pipes (Ø72 mm, thickness 4 mm, density 8000 kg/m^3): 6.8 kg/m *Materials use per meter of extraction filter, heater and temperature sensor.* Stainless steel use for filter (Ø20 mm, thickness 4 mm, density 8000 kg/m^3): 1.6 kg/m Stainless steel use for heater (Ø10 mm, solid and U-shape, density 8000 kg/m^3): 1.2 kg/m Stainless steel use for temperature sensor (Ø10 mm, solid, density 8000 kg/m^3): 0.6 kg/m *Gravel and concrete use per well:* Gravel use per well: 142 kg (7.5 meter of gravel pack in Ø150 mm boring) Concrete use per well: 15 kg (0.5 meter of concrete) *Steel for aboveground gas collection pipes:* 12 meter of Ø80 mm pipes: 90 kg 48 meter of Ø20 mm pipes: 76 kg
ISTD	*Use of activated carbon:* Estimations based on adsorption data from supplier (*5*) Vapor treatment: 200 kg (assumed to absorb 20% of weight = 40 kg TCE) Water treatment: 60 kg (assumed to absorb 3% of weight = 1.8 kg TCE)
ISTD	The inventory for activated carbon production was based on data from (*6*) with addition of an end-of-life stage assuming that the coal was transported to a power plant and substituted an equal amount of hard coal. The chlorinated incineration products from the contaminants adsorbed to the activated carbon were assumed fully removed by the flue gas treatment of the power plant
ISTD	End-of-life stage for EPS insulation (560) kg not included as impact estimated to be negligible.
ISTD	The activated carbon container, electricity cables, transformer and equipment cabins where not included in inventory (except the transport to the site) as only a minor part of their total service life is ascribed to this project, and their contribution is hence negligible
EXC	The soil transportation distance from site to treatment (100 km) and from treatment to disposal (50 km) was based on the actual distance to most likely receivers of the excavated soil (treatment facility and disposal site).
EXC	Steel use for sheet pile wall (*7*). (depth 9.5 m, thickness 9 mm, surface area 450 m2, density 7850 kg/m^3): 31.8 tonnes. Only 1/3 of the steel production is ascribed to this project as the steel profiles are expected to be reused directly an average of two times (*7*).
EXC	End-of-life stage for removed pavement (140m^2) not included as impact estimated to be negligible
EXC	Inventory for soil treatment facility from (*8,9*). *Material and energy use per tonne of soil treated:* Steel: 0.3 kg/tonne PE: 0.023 kg/tonne PVC: 0.6 kg/tonne Gravel: 22 kg/tonne Electricity: 1.2 kWh/tonne Diesel use for site construction and layout in piles: 0.07 L/tonne Diesel use for treatment (turning of piles): 1 L/tonne
EXC	Loss of TCE to air during excavation and soil treatment was disregarded in the inventory as the contribution to impacts (ozone formation, human and ecotoxicity) was found to be negligible

Table S2. List of estimations and assumptions for the life cycle inventory not mentioned in the article or in table S1

Relevant for technologies	**Energy consumption data**
ERD, ISTD, EXC	Diesel use for well drilling and soil core sampling (*10*): 1.75 L diesel per meter drilled
NoA, ERD	Estimated electricity use for groundwater sampling (0.65 kw, 0.6 h): 0.39 kWh/per screen
ERD	Bioculture production: 1.2 kWh per liter of culture. Estimated based on data from (*2*). Density of bioculture: 1 kg/L
ERD	Geoprobe diesel use for injection: 10 L/day (*11*). Number of injection sites per day: 10 (with injections at 25 cm intervals from 3-8 meters below surface) (*11*).
ISTD	Estimated electricity use for pumping of groundwater during heating (0.75 kW, 2356 h, 3.3months): 1770 kWh
ISTD	Energy for heating of soil: 400 kWh/m^3 of soil Electricity use for ventilation, cooling, condensation: 20 kWh/m^3 of soil The energy use was estimated based on experience data from (*12*) from other remediated sites with similar geology and water content. The applied value represents the expected energy use for a clay till site with a low water flow in the heated zone. For a site with a higher water flow, the energy use can be up to 500 kWh/m^3 of soil (*12*).
EXC	Diesel for excavation and backfilling: 1.8 L/m^3 soil (includes diesel for excavation, backfilling and miscellaneous) (*13*).
EXC	Diesel use for installation of sheet pile wall: (40 L/hour, 4 tonnes installed per hour): 10 L/hour The same diesel use is assumed for wall removal (*7*).
EXC	Asphalting: 0.11 liter diesel per m^2 (From Road-Res model (*14*)) Removal of asphalt: 0.04 liter diesel per m^2

Table S3. Energy consumption data used in the inventor

Process	**Ecoinvent processes used for electricity consumption**
Danish electricity	Average: *Electricity, low voltage, at grid/DK* Marginal: *Electricity, hard coal, at power plant/NORDEL** (*loss due to transmission and distribution, hexafluoride use, transmission and distribution network infrastructure added to process to represent low voltage electricity at grid)
Dutch electricity	*Electricity, low voltage, at grid/NL*
Process	**Ecoinvent processes used for production of 1 kg**
Steel product	*Steel, electric, un- and low-alloyed, at plant/RER*: 1 kg *Hot rolling, steel/RER*: 1 kg *Steel product manufacturing, average metal working*/RER: 1 kg *Steel, converter, low-alloyed at plant/RER*: 0.1 kg (steel loss) *Transport, lorry >16t, fleet average/RER*: 0.1 tkm *Transport, freight, rail/RER:* 0.2 tkm
Stainless steel product	*Steel, electric, chromium steel 18/8, at plant/RER** (*all metal raw material assumed from scrap): 1kg *Hot rolling, steel/RER*: 1 kg *Chromium steel product manufacturing, average metal working/RER*: 1 kg *Steel, converter, chromium steel 18/8, at plant/RER:* 0.1 kg (steel loss) *Transport, lorry >16t, fleet average/RER*: 0.1 tkm *Transport, freight, rail/RER:* 0.2 tkm

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PE product	*Polyethylene, HDPE, granulate, at plant/RER:* 1 kg *Extrusion, plastic pipes/RER:* 1 kg *Transport, lorry >16t, fleet average/RER*: 0.1 tkm *Transport, freight, rail/RER*: 0.2 tkm
EPS	*Polystyrene, extruded (XPS), at plant/RER*: 1kg *Transport, lorry >16t, fleet average/RER*: 0.1 tkm *Transport, freight, rail/RER:* 0.2 tkm
Gravel	*Gravel, round at mine/CH*: 1 kg *Transport, lorry >16t, fleet average/RER*: 0.1 tkm
Organic molasses	*Molasses from sugar cane, at sugar refinery/BR** (*The CO_2 uptake during plant growth was removed as the same amount will be emitted when it is degraded. Pesticide use was removed from the inventory to represent the organically grown sugar cane, which was actually used for the molasses production. Lastly, heavy metal uptake from the soil during plant growth was removed as explained in Table S2) *Transport, lorry >16t, fleet average/RER:* 0.2 tkm *Transport, transoceanic freight ship/OCE:* 10.458 tkm
Bioculture	*Electricity, low voltage, at grid/NL*: 1.2 kWh *Acetic acid, 98% in H2O, at plant/RER*: 0.3 g *Ammonium nitrate phosphate, as N, at regional storehouse/RER:* 1 g *Transport, lorry 3.5-16t, fleet average/RER*: 0.727 tkm
Concrete	*Concrete, normal, at plant/CH*: 0.00045 m^3 *Transport, lorry >16t, fleet average/RER*: 0.05 tkm
Activated carbon	*Hard coal mix, at regional storage/UCTE*: 3 kg *Hard coal, burned in industrial furnace 1-10MW/RER**: 48 MJ (only emissions, coal use removed) *Electricity, medium voltage, production UCTE, at grid/UCTE*: 1.6 kWh *Natural gas, burned in industrial furnace low-NO > 100kW/RER*: 13.2 MJ *Transport, lorry >16t, fleet average/RER* 0.6 tkm *Transport, lorry >16t, fleet average/RER* 0.1 tkm *Transport, lorry 3.5-7.5t EURO5/RER* 0.1 tkm Avoided: *1 kg hard coal mix at regional storage*/UCTE
Bentonite	*Bentonite, at processing/DE*: 1 kg *Transport, lorry >16t, fleet average/RER*: 0.6 tkm
Asphalt	*Mastic asphalt, at plant/CH:* 1 kg *Transport, lorry >16t, fleet average/RER*: 0.05 tkm
Process	**Ecoinvent processes applied for transport and non-road diesel use**
Transport	Soil transport: *Transport, lorry >16t, fleet average/RER U** (*Diesel consumption per tkm increased by 20.5% based on diesel consumption data regarding a full and empty lorry respectively collected from a Danish transport company (*15*)). Person transport: *Transport, passenger car, diesel, fleet average 2010/RER*
Emissions from non-road diesel use	Emissions from: *Excavation, hydraulic digger, low sulphur diesel/RER U* Used for excavator, drill rig, Geoprobe etc. Diesel use for different work types, see Table S3
	Other ecoinvent processes used
Soil landfill	*Process-specific burdens, inert material landfill/CH*

Table S4. List of ecoinvent processes used and adjustments made. Ecoinvent process names are given in italic. Processes with adjustments are marked with an asterisk and the change explained in parentheses.
Processes marked with RER are representative of average Europe production and supply situation. Country specific processes are marked with a country code: DK: Denmark, BR: Brazil, CH: Switzerland, NL: The Netherlands, DE: Germany. UCTE and NORDEL are electricity mixes for larger regions, namely western and northern Europe respectively.
Note that 1 tkm is a measure of transported mass (in tonnes) times the distance (in km).

A.3 Numerical reactive transport model for primary impacts

A numerical reactive transport model developed by Chambon et al. (*16*) and further developed in Chambon et al. (*17*) was used to estimate the timeframes for 98% mass removal with natural attenuation (NoA) and enhanced reductive dechlorination (ERD) at the site. Furthermore, the model was used to estimate the mass discharge of TCE and degradation products (cis-DCE and VC) to the regional limestone aquifer. The sequential dechlorination of TCE is described using Monod kinetics, which includes the concentration and growth of the specific degraders (*Dehalococcoides*). The degradation rate, r_i (mole.$L^{-1}d^{-1}$) has the following general form:

$$r_i = \frac{\mu_i \cdot X / Y \cdot C_i}{C_i + K_i}$$

Where C_i is the concentration of the chlorinated ethene i (mole L^{-1}), μ_i is the maximum growth rate of i (d^{-1}), X is the dechlorinating biomass concentration (cells L^{-1}), Y is the specific yield (cells μmol^{-1}) and K_i is the half velocity coefficient of i (mole L^{-1}). The sequential degradation is described by a set of coupled differential equations where the degradation of the mother product results in the production of the daughter product:

$$\frac{dC_i}{dt} = -r_i + r_{i+1}$$

Where r_i is the degradation rate of the chlorinated ethene i and r_{i+1} is the production rate of chlorinated ethene i via the degradation of the higher chlorinated ethene. In the no action scenario, degradation of chlorinated ethenes is assumed only to take place in and along the vertical fractures in the clay. In the enhanced reductive dechlorination scenario, horizontal degradation zones corresponding to the depths of injection (each 25 cm over the depth) are assumed (see illustration of degradation zones in Figure 1). In both cases, the degradation zones extend 5 cm into the clay matrix around the natural fracture (NoA) or the injection depth (ERD). The extent of the bioactive zones in the clay till matrix was based on experience from another Danish clay till site subject to ERD (*18*). The biomass concentration of dechlorinating bacteria of the ERD scenario is based on measurements done at the study site, where the ERD method was actually applied. For the NoA scenario a biomass concentration of 20 times lower was assumed, based on a calibration of model results to measured concentrations of degradation products in the aquifer.

The degradation model is combined with a transport model including sorption and diffusive transport in the clay matrix and vertical advective-dispersive transport in the fractures. The numerical model is solved in the Comsol Multiphysics software. Model input parameters are listed in Table S5 and Table S6. In addition, contaminated volume and mass are important parameters. The model estimates concentration profiles of TCE and the degradation products (cis-DCE and VC) over time in the soil matrix. The mass discharge to the aquifer is calculated based on the concentrations of TCE, cis-DCE and VC at the fracture outlet to the aquifer (see results in Figure 1).

Geological and hydrological parameters		**Value**
Net recharge rate to aquifer [mm/year]		100
Fracture spacing *2B* [m]		2
Fracture aperture *2b* [μm]		22
Vertical hydraulic gradient I_v [m/m]		1
Bulk hydraulic conductivity K_b [m/s]		$3.2*10^{-9}$
Matrix porosity ϕ		0.3
Bulk density ρ_b [kg/L]		1.9
Tortuosity τ		0.3
Free diffusion coefficient $D*_i$ [m^2/y]		
	TCE	0.020
	DCE	0.022
	VC	0.026
	Ethene	0.033
Sorption coefficient Kd_i [L/kg]		
	TCE	0.6
	DCE	0.12
	VC	0.04
	Ethene	0
Dispersivity in fracture [m]		0.1

Table S5. Model parameters related to transport. The fracture aperture and the bulk hydraulic conductivity are calculated based on the water balance of the clay till, cf. to (*17*) for details.

Microbial parameters		**Value**
Maximum growth rate μ_i [1/d]		
	TCE	2
	DCE	0.38
	VC	0.14
Specific yield *Y* [cell μmol^{-1}]		$5.2*10^8$
Biomass concentration *X* [cell/L]	NoA	$5*10^7$
Biomass concentration *X* [cell/L]	ERD	10^9

Table S6. Model parameters related to microbial degradation

A.4 Substrate demand calculation for ERD

To stimulate the *in situ* reductive dechlorination of TCE via cis-DCE to VC and finally ethene, a sufficient amount of an organic substrate (electron donor) is required to react with all electron acceptors in the source zone. Native electron acceptors in the treated soil volume are divided into those that are dissolved such as O_2, NO_3^-, SO_4^{2-} etc. those which are in the solid phase (sorbed) such as Fe(III) and Mn(IV). During fermentation, the organic substrate (in this case sugar cane molasses) generates hydrogen, which is the actual electron donor in the reaction with the chlorinated ethenes. Table S7 presents the calculation of substrate demand for the study site. The calculation is carried out in accordance with guidelines in (*19*). For this site a H_2 demand of 56 kg is estimated for reduction of all electron acceptors. According to (*19*), the H_2 production potential for sucrose (which is the main sugar constituent in cane molasses) is 0.047 kg/kg. This corresponds to a production of 8 moles of H_2 per mole of sucrose. Due to uncertainty in e.g. solid phase electron acceptor concentrations, microbial efficiency etc., safety factors are often used in the substrate demand calculations and the guideline propose safety factors from 2 to 5. Here a safety factor of 2 was applied, resulting in a required amount of sucrose of 2400 kg. The sugar content of the sugar cane molasses used as substrate is 46% according to the supplier[1]. Hence, the necessary amount of molasses is approximately 5000 kg.

			Stoichiometric relation (mg H_2/mg acceptor)	H_2 demand (kg)	Substrate demand (kg sucrose)	Substrate demand with SF of 2 (kg sucrose)	Molasses demand with safety factor (kg molasses)
Dissolved electron acceptors							
	Conc. (mg/L)	Mass (kg)					
Oxygen	1	1.1	0.125	0.14	3.0	6.07	13.2
Nitrate	200	228.2	0.08	18.3	388	777	1689
Sulfate	330	376.5	0.08	30.1	641	1282	2787
Sorbed electron acceptors							
	Conc. (mg/kg DS)	Mass (kg)					
Iron (FeIII)	207	275.31	0.02	5.51	117	234	509
Chlorinated solvents							
	Conc. (mg/kg DS)	Mass (kg)					
TCE	31.1	41.3	0.05	2.1	43.9	87.9	191
SUM				56	1193	2387	**5189**

Table S7. Calculation of substrate demand. The total mass of dissolved electron acceptors takes into consideration the inflow of these during a period of 38 years. SF: safety factor. Conc.: Concentration. DS: Dry solids.

A.5. Estimation of methane generation from substrate decomposition

[1] The approximate composition of the rest of the mass is water (25%), ash (13%), metals (6%), salt (4.6%), crude protein (4%) and other carbohydrates (2.5%). Based on information from supplier and the literature (*20*).

Methane generation from substrate degradation was included in the inventory of greenhouse gas emissions. The anaerobic substrate decomposition is a fairly complex process. Here it was simplified by the following set of reactions:

- Hydrolysis of sucrose to lactate: $C_{12}H_{22}O_{11} + H_2O \rightarrow 4CH_3CHOHCOO^- + 4H^+$
- Lactate fermentation to acetate (*21*): $CH_3CHOHCOO^- + 2H_2O \rightarrow CH_3COO^- + HCO_3^- + 2H_2 + H^+$
- Methanogenesis from acetate: $CH_3COO^- + H^+ \rightarrow CH_4 + CO_2$

Thus, for each mole of sucrose, 4 moles of methane is generated. This corresponds to a methane generation of 0.19 kg per kg of sucrose or 0.09 kg per kg of molasses.

Some of the substrate may not react with any electron acceptor (due to the safety factor) and is assumed degraded to methane and carbon dioxide after the following equation, where 6 moles of methane is produced per mole of sucrose:

$$C_{12}H_{22}O_{11} + H_2O \rightarrow 6CO_2 + 6CH_4$$

This corresponds to a methane generation of 0.28 kg per kg sucrose or 0.13 kg per kg molasses.

Assuming that 50 percent of the molasses reacts after each scheme presented above, the average methane generation rate is 0.11 kg methane per kg molasses. Carbon used for biomass increase is not subtracted in this calculation. Furthermore, no methane oxidation in the unsaturated soil is assumed as the gas is expected to be transported quickly to the surface via gravel structures or wells. Carbon dioxide from biogenic sources (in this case molasses) is not accounted for in the LCA as it balances with the carbon dioxide taken up by the plant from the atmosphere during photosynthesis; however biogenic methane is accounted for as this has a direct global warming potential apart from the contribution through oxidation to carbon dioxide.

B. Calculation of human toxicity via groundwater ingestion (primary impacts)

The mass discharge of the chlorinated ethenes from source to groundwater $J_i(t)$ (g/yr) is calculated using the numerical model for transport and degradation of chlorinated ethenes in fractured clay till described in section A.3.

Worst case exposure concentrations, $C_i(t)$ (g/m^3), in the groundwater abstracted at the downstream well field with a pump rate, Q, of 2.5E6 m^3/year are estimated assuming that all mass is captured in the water supply well and that no further degradation takes place during the transport to the well:

$$C_i(t) = \frac{J_i(t)}{Q}$$

The accumulated intake of each contaminant due to groundwater ingestion $m_{ing,i}$ is calculated by integrating the concentration times the ingested volume (1.4 L/day (*22*)) over a period of 38 years for the ERD scenario and 1200 years for the no action scenario:

$$m_{ing,i} = \int_0^t C_i(t) \cdot V dt$$

The number of illness cases (cancer or non cancer) is then calculated by multiplying with the USEtox human health effect factors for TCE, DCE and VC (listed in Table S8) and multiplying with the size of the affected population (POP), which is assumed constant at 44 000:

- Number of cancer cases summed for intake of all chlorinated ethene, i:

$$CTU(cancer) = POP \cdot \sum_{i=1}^{3} m_{ing,i} \cdot HHEF_{i,cancer}$$

- Number. of non-cancer cases summed for intake of all chlorinated ethene, i:

$$CTU(noncancer) = POP \cdot \sum_{ii=1}^{3} m_{ing,i} \cdot HHEF_{i,noncancer}$$

The human health effect factors express the number of cases per kg of a chemical ingested. The toxicity evaluation is based on ED50 values and assumes a linear dose-response function up to a probability of 0.5. (*23*)

CAS	**Name**	**Human health effect factor [cases/kg_intake]**	
		Ingestion	
		Cancer	non-cancer
79-01-6	Trichloroethene	2.94E-03	n/a
75-35-4	1,1-Dichloroethene	5.00E-51	3.97E-02
156-60-5	1,2-Dichloroethene (trans)	n/a	2.66E-02
75-01-4	Chloroethene (vinyl chloride)	1.52E-01	8.03E-01
156-59-2	1,2- Dichlorethene (cis)	Not in Usetox database[1]	

[1] The average values for 1,1 DCE and 1,2 trans DCE was used for non-cancer effects, no cancer effect was assumed.

Table S8. USEtox human health effect factors for TCE, DCE and VC (USEtox version 1, January 2010)

C. Life cycle impact assessment

C.1 Applied impact assessment models

EDIP2003 (*24*) is the impact assessment method applied for the categories global warming, ozone formation, acidification and eutrophication. EDIP2003 uses a 100 year time horizon for calculation of global warming and the characterization factors for global warming represent the 2007 values from IPCC.

Respiratory inorganics cover respiratory impacts related to emissions of particulate matter ($PM_{2.5\text{-}10\mu m}$, $PM_{<2.5\mu m}$) and inorganics (NO_x, SO_2, and NH_3). Midpoint characterization factors (in unit kg PM2.5-eq/kg emissions) were calculated based on endpoint characterization factors from Humbert et al. (*25*) and adapted to European conditions using the guidance provided (*25*).

USEtox[TM] (version 1.0, January 2010) is used for primary and secondary human toxicity and ecotoxicity (*22*). The USEtox characterization factors were imported to the SimaPro software, which was used for the life cycle modeling. Primary impacts from leaching of contaminants on-site were calculated separated from SimaPro as only the effect factors in Table S8 from the characterization factors were applied. Fate of the chlorinated ethenes was modeled using the site-specific reactive transport model (see section A.3) including degradation product generation, and exposure parameters were adapted to the actual number of people exposed to the contaminated drinking water (see section B).

C.2 Normalization references and weighting factors

The applied normalization references expressing the average annual impacts per capita for non-toxic and toxic impacts are presented in Table S9. For simplification, aquatic eutrophication (in terms of kg N and kg P respectively) were combined to one impact score expressed in terms of kg NO_3^- (*24*). Resource consumption is translated into person reserves according to updated normalization and weighting references for 2004 (*26*) as listed in Table S10.

Impact category	LCIA method	Unit	Value	Base
Global Warming	EDIP2003	kg CO_2 eq/pers/yr	7728	World
Ozone formation (Human)	EDIP2003	person.ppm.h/pers/yr	2.84	EU27
Ozone formation (Vegetation)	EDIP2003	m2.ppm.h/pers/yr	59701	EU27
Acidification	EDIP2003	m^2/pers/yr	392	EU27
Terrestrial eutrophication	EDIP2003	m^2/pers/yr	1367	EU27
Aquatic eutrophication EP(N)	EDIP2003	kg N/pers/yr	8.33	EU27
Aquatic eutrophication EP(P)	EDIP2003	kg P/pers/yr	0.28	EU27
Aquatic eutrophication [1]	EDIP2003	kg NO_3^-/pers/yr	44.0	EU27
Respiratory inorganics	Humbert et al., 2010 (*25*)	kg PM2.5 eq/pers/yr	6.49	EU27
Ecotoxicity freshwater	USEtox	CTU_e/pers/yr	4744	EU27
Human toxicity (non-cancer)	USEtox	CTU_h/pers/yr	8.16E-04	EU27
Human toxicity (cancer)	USEtox	CTU_h/pers/yr	4.97E-05	EU27

[1] Weighted average based on the normalization references for aquatic eutrophication (kg N) and aquatic eutrophication (kg P) respectively as 62/14 g NO3-/gN * 8.33 + 32 g P/ g N *0.28.

Table S9. Applied normalization references representing the average impact from a world citizen (global warming) or a European (other impacts) per person per year. All values are calculated for the base year 2004 (*27*).

Ressource	Normalization reference, kg/pers/yr	Weighting factor
Brown coal	2.64E+02	3.93E-03
Coal	6.02E+02	8.04E-03
Natural gas	3.53E+02	1.50E-02
Oil	6.06E+02	2.39E-02
Aluminium	4.52E+00	6.78E-03
Copper	2.27E+00	3.09E-02
Chromium	8.26E-01	2.12E-02
Iron	9.80E+01	7.81E-03
Manganese	1.72E+00	2.89E-02
Molybdenum	2.17E-02	1.62E-02
Nickel	2.19E-01	2.26E-02
Uranium	5.62E-03	1.02E-02

Table S10. Applied normalization references and weighting factors for resource consumption. The normalization references represent the average resource use for an average world citizen expressed per person per year for the base year 2004, and the weighting factors represent the reciprocal supply horizon of each resource (*26*).

D. Additional results

D.1. Characterized results

Table S11 lists the characterized results before normalization. For simplification, ozone formation impact on vegetation has been left out of the results presented in the paper as they exhibit the same trend as the ozone formation impacts on humans. Table S12 shows the contribution of TCE and the degradation products cis-DCE and VC to the primary toxic impacts. VC-formation is clearly the dominant cause of primary impact both to human toxic cancer and non-cancer effects especially in the ERD scenario. Table S13 lists the resource consumption inventory result.

Indicator	Unit	NoA	ERD	ISTD	EXC
Global warming	kg CO_2 eq	29064	29678	201396	120364
Ozone formation (Human)	person.ppm.h	11.1	29.0	55.3	116.0
Ozone formation (Vegetation)	m^2.ppm.h	158379	388827	770747	1699745
Acidification	m^2	1394	1305	9275	10513
Terrestrial eutrophication	m^2	1741	2415	7348	21635
Aquatic eutrophication	kg NO_3^-	32.4	45.0	141.9	401.7
Respiratory inorganics	kg PM2.5 eq	16.6	14.7	115.3	102.0
Ecotoxicity freshwater	CTU_e	82507	22300	304452	902432
Sec. human toxicity (non-cancer)	CTU_h	3.88E-03	1.48E-03	2.04E-02	4.70E-02
Sec. human toxicity (cancer)	CTU_h	1.31E-03	7.61E-04	1.56E-02	4.46E-02
Pri. human toxicity (non-cancer)	CTU_h	2.03E-02	1.61E-02		
Pri. human toxicity (cancer)	CTU_h	3.90E-03	3.02E-03		
Human toxicity (non-cancer)[1]	CTU_h	2.44E-02	1.76E-02	2.04E-02	4.70E-02
Human toxicity (cancer)[1]	CTU_h	5.21E-03	3.78E-03	1.56E-02	4.46E-02

[1]Sum of secondary and primary toxic impacts

Table S11. Characterized life-cycle impacts

	Human toxicity (non-cancer) CTU_h	Fraction of total impact	Human toxicity (cancer) CTU_h	Fraction of total impact
NoA				
TCE	n/a	-	5.50E-04	0.14
Cis-DCE	2.63E-03	0.13	n/a	-
VC	1.76E-02	0.86	3.35E-03	0.87
Sum	2.03E-02		3.90E-03	
ERD				
TCE	n/a	-	5.59E-05	0.02
Cis-DCE	4.76E-04	0.03	n/a	-
VC	1.56E-02	0.97	2.97E-03	0.98
Sum	1.61E-02		3.02E-03	

Table S12. Contribution of TCE, cis-DCE and VC to primary toxic impacts

Resource	Unit	NoA	ERD	ISTD	EXC
Brown coal	kg	1151	791	10128	7587
Coal	kg	5071	1840	72729	10152
Natural gas	kg	1582	1562	16874	6637
Oil	kg	4913	2662	5687	40704
Aluminium	kg	22.0	4.4	28.2	78.4
Copper	kg	12.9	5.3	138.8	30.0
Chromium	kg	3.27	1.28	49.59	32.65
Iron	kg	551	275	1359	6022
Manganese	kg	0.88	0.80	4.66	30.1
Molybdenum	kg	1.12	0.92	6.99	32.4
Nickel	kg	10.4	5.86	124	145
Uranium	kg	0.09	0.04	0.48	0.45

Table S13. Resource use inventory result

D.2. Detailed resource consumption result

The resource consumption in person reserves (PR) divided into fossil energy carriers and scarce metals is summarized in Table S14. A more detailed presentation of the results for resource consumption divided into energy types and individual metals are given in Figure S1. The use of fossil energy carriers is approximately 2 PR both for ISTD and excavation, although ISTD has a higher global warming potential. This is due to the fact that whereas the energy consumption for ISTD relies on mainly coal and natural gas-based Danish electricity, excavation and transportation consume large amounts of crude oil, which has a shorter supply horizon than coal. The use of scarce metals is high for both methods due to the use of steel and stainless steel for heating wells and pipes in ISTD and steel for the sheet pile wall used around the excavation pit. Alloying metals for steel and stainless steel (molybdenum, nickel and chromium) are the main contributors due to their scarcity. ERD has the lowest consumption of both fossil energy and metals.

Resource consumption	Unit	NoA	ERD	ISTD	EXC
Fossil energy carriers	PR	0.35	0.21	2.1	2.1
Metals	PR	2.6	2.1	22	42

Table S14. Resource use in person reserves (PR) for no action and the three remediation alternatives

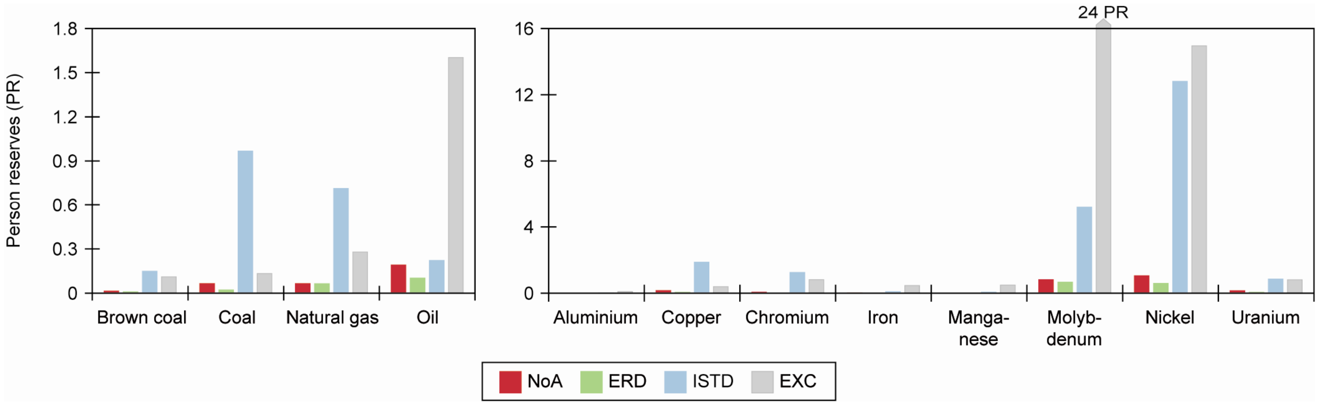

Figure S1. Resource consumption in person reserves (PR). Note the different y-axes of the two figures.

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