

Open Archive Toulouse Archive Ouverte (OATAO)

OATAO is an open access repository that collects the work of Toulouse researchers and makes it freely available over the web where possible.

This is an author-deposited version published in: <u>http://oatao.univ-toulouse.fr/</u> Eprints ID: 5254

Official URL: http://dx.doi.org/10.1007/s11367-011-0366-7

To cite this version:

Busset, Guillaume and Sangely, Matthieu and Montréjaud-Vignoles, Mireille and Thannberger, Laurent and Sablayrolles, Caroline *Life cycle assessment of Polychlorinated Biphenyl contaminated soil remediation processes*. International Journal of Life Cycle Assessment . ISSN 0948-3349

Any correspondence concerning this service should be sent to the repository administrator: staff-oatao@inp-toulouse.fr

Life cycle assessment of polychlorinated biphenyl contaminated soil remediation processes

Guillaume Busset • Matthieu Sangely • Mireille Montrejaud-Vignoles • Laurent Thannberger • Caroline Sablayrolles

Abstract

Purpose A life-cycle assessment (LCA) was performed to evaluate the environmental impacts of the remediation of industrial soils contaminated by polychlorobiphenyl (PCB). Two new bioremediation treatment options were compared with the usual incineration process. In this attributional LCA, only secondary impacts were considered. The contaminated soil used for the experiments contained 200 mg of PCB per kilogram.

Methods Three off-site treatment scenarios were studied: 1) bioremediation with mechanical aeration, 2) bioremediation with electric aeration and 3) incineration with natural gas. Bioremediation processes were designed from lab-scale, scale-up and pilot experiments. The incineration technique was inspired by a French plant. A semi-quantitative uncertainty

G. BussetCATAR-CRITT Agroressources,4 Allee Emile Monso,31030 Toulouse, France

M. Sangely · L. Thannberger VALGO,81 rue Jacques Babinet,31100 Toulouse, France

M. Sangely · C. Sablayrolles
Universite de Toulouse, INP, LCA (Laboratoire de Chimie Agro-Industrielle), ENSIACET,
4 Allee Emile Monso,
31030 Toulouse, France

M. Sangely · M. Montrejaud-Vignoles · C. Sablayrolles (🖂) INRA, LCA (Laboratoire de Chimie Agro-Industrielle), 31029 Toulouse, France e-mail: caroline.sablayrolles@ensiacet.fr analysis was performed on the data. Environmental impacts were evaluated with the CML 2001 method using the SimaPro software.

Results and discussion In most compared categories, the bioremediation processes are favorable. Of the bioremediation options, the lowest environmental footprint was observed for electric aeration. The uncertainty analysis supported the results that compared incineration and bioremediation but decreased the difference between the options of aeration. The distance of transportation was one of the most sensitive parameters, especially for bioremediation. At equal distances between the polluted sites and the treatment plant, bioremediation had fewer impacts than incineration in eight out of 13 categories.

Conclusions The use of natural gas for the incineration process generated the most impacts. Irrespective of the aeration option, bioremediation was better than incineration. The time of treatment should be taken into account. More precise and detailed data are required for the incineration scenario. More parameters of biological treatments should be measured. LCA results should be completed using ecological and health risk assessment and an acceptability evaluation.

Keywords Attributional LCA · CML-method · Environmental evaluation · Midpoint category · Polychlorinated biphenyl

1 Introduction

The management of contaminated soil requires the selection of the most adapted technology from a wide range of options (Suer et al. 2004). Remediation techniques take place on site, either in situ or ex situ, or off-site and include thermal treatments, biological treatments, soil washing, landfill, electrodialysis, bioleaching, biosparging treatments, chemical treatments (such as oxidation or reduction) and solvent extraction among many others (Cadotte et al. 2007; Lemming et al. 2010). In practice, ex situ techniques appear to be the most commonly used (Lemming et al. 2010). These technologies also diverge in their results. They could lead to immobilization, separation, concentration or destruction of the pollutants (Rahuman et al. 2000). The primary differences lie in their technology, but their cost, efficiency and duration are also considered.

Pollutants are either inorganic, such as metals, or organic, and their physical and chemical properties, such as volatility, persistence, solubility and conductivity influence the choice of remediation technique.

Until now, polychlorobiphenyl (PCB) contaminants have most often been destroyed by incineration. However, the dedicated incinerators used for this process require a large amount of energy to limit dioxin formation, and few efficient alternatives are available. Chemical oxidation has shown a low efficiency (Zhou et al. 2004). Supercritical water oxidation exhibits high destruction efficiency but requires high pressure and temperature conditions (Zhou et al. 2004). A phytoremediation technique using methylated- α -cyclodextrins has been the subject of a recent study (Shen et al. 2009). The results are conclusive: the impact of methylated- α -cyclodextrins must be investigated. To investigate the biological breakdown of PCBs, Sangely et al. (2009) have tested the combination of Phanerochaete crysosporium, a fungus capable of breaking down PCBs under anaerobic conditions, and Burkholderia xenovorans, a bacterium implicated in PCB breakdown under aerobic conditions. The combination of aerobic and anaerobic steps has given rise to a new process of bioremediation of PCBcontaminated soils and has been developed on both the laboratory scale and as a pilot project.

Life-cycle assessment (LCA) appears to be a method well adapted for the evaluation of the impacts of remediation techniques (Morais and Delerue-Matos 2009). LCA can be attributional or consequential, particularly in the soil remediation domain (Lesage et al. 2007a; Lesage et al. 2007b). Attributional LCA evaluates the primary impacts from residual contamination and/or the secondary impacts from the technique life cycle. Consequential LCA takes into account environmental and economic impacts after remediation (Volkwein et al. 1999). Most authors have limited their studies to secondary impacts (Lemming et al. 2010). LCA has been applied to contaminations of lead (Page et al. 1999), polycyclic aromatic hydrocarbons, chromium and mineral oils (Volkwein et al. 1999), sulfur (Blanc et al. 2004), diesel fuel (Toffoletto et al. 2007; Cadotte et al. 2007) and trichloroethene (Lemming et al. 2010). These studies show that LCA is a relevant management tool for evaluation of the environmental impacts of soil remediation techniques of different pollutants.

A life-cycle assessment was undertaken to compare different treatments of PCB waste in Ohio, USA. This complete study investigated environmental impacts and economic, technologic and health risks (Morris et al. 2000). At that time, biological treatments were only in the R&D stage; therefore, they were not included among the evaluated techniques. Another recent LCA investigated PCB treatment techniques but compared a high-temperature process with a base-catalyzed decontamination (Hu et al. 2011). No LCA has been performed on the new biological process used in this study.

The objectives of this study are (a) to evaluate, via attributional LCA methodology, the potential environmental impacts of the bioremediation process for PCB-contaminated soils, as recently established by Sangely et al. (2009), and (b) to compare the bioremediation impacts to the impacts of the current incineration technique.

2 Methodology

The life-cycle assessment was undertaken using the ISO14040 (2006) and ISO 14044(2006) standards

2.1 LCA goal and scope

The evaluated system's function was to restore soil PCB contamination levels to waste acceptance criteria (50 mg kg⁻¹ of soil) in hazardous (French class 1) waste landfill sites in France (http://www.ineris.fr/aida/?q=consult_doc/consultation/ 2.250.190.28.8.2283). The reference flow was taken as the amount of moist soil (20% moisture) that can be excavated in 1 day under pilot-project conditions; this quantity corresponds to 600 t per day. Laboratory results have shown a potential of PCB degradation in soil of 556 µg kg⁻¹ per day. Therefore, the PCB concentration can be reduced from 200 mg kg⁻¹– 50 mg kg⁻¹ of soil in 265 days (Sangely 2010). The functional unit was therefore defined as treating 600 t of PCBcontaminated moist soil (20% moisture) to reduce its PCB concentration from 200 mg kg⁻¹–50 mg kg⁻¹ of soil.

The processes taken into consideration for the studied systems included excavation and transport to the landfill site after the treatment phase. A detailed description is given in the next section. For all of these processes, infrastructure construction, worker transport and landfill site maintenance were not taken into account, primarily because the share of impacts by soil remediation treatments was negligible. The remediation activity of PCB-contaminated soils is not the most important part of the enterprises activity. Systems boundaries are discussed in "Section 4".

2.2 Life-cycle inventory

2.2.1 Systems description

The life-cycle assessment was used to compare two PCBcontaminated soil remediation processes: soil incineration and biological treatment. Three scenarios were defined: BM, treatment by bioremediation with mechanical aeration; BE, treatment by bioremediation with electric aeration; and Inc, treatment by incineration.

Biological treatment The biological treatment is an innovative and original process based on experimental laboratory-scale and pilot-scale trial results. The treatment's procedure consisted of alternating aerobic and anaerobic phases. The aerobic conditions favor the development of the bacteria B. xenovorans, and the anaerobic conditions favor the fungi P. crysosporium; each is capable of partially breaking down PCB. Bacteria broke down the less-chlorinated PCBs, whereas fungi broke down the more highly chlorinated PCBs (Sangely et al. 2009). When the bacteria and fungi were broken down in tandem, PCB was broken down to the target concentration or lower. In practice, the treatment of PCB-contaminated soils required three cycles; each cycle consisted of 2 months under anaerobic conditions and 1 month under aerobic conditions.

Excavation, the first phase, was followed by transport to the bioremediation site, where the soil was immediately put onto a waterproof concrete platform (not taken into account in the LCA). This soil was then covered with a low-density polyethylene sheet, supplied with nitrates and flooded with water to create anaerobic conditions. The aerobic phase was facilitated by soil aeration. The two techniques being studied were given two different bioremediation scenarios. These scenarios were designed to allow a comparison of two technical alternatives and determine the best one. The first technique (BM) involved turning the soil over four times per cycle using a 5-t mechanical digger. The second technique (BE) involved the electrical pumping of air through the soil for 25% of the aerobic phase. An 11-kW compressor was used. The first anaerobic/aerobic cycle was followed by two more identical cycles. After three cycles, the soil was transported to the nearest hazardous waste landfill site. For this stage, the residual amount of PCB met the landfill's waste acceptance upper limitation criteria. It was considered as an emission to the soil. A flow diagram of the bioremediation procedure and the two aeration options is shown in Fig. 1.

The potential direct emissions from anaerobic and aerobic PCB decomposition were not known and were not considered.

Treatment by incineration Soil treatment by incineration consisted of excavation followed by transport to the incineration site. At the incineration site, the soil was put into a rotating oven where soils and other organochloride wastes were burned at a high temperature (1200°C) (Séché 2010). The gaseous waste was burned in a second combustion at 1200°C, followed by a rapid cooling to 70°C to avoid the formation of dioxins and furans (Séché 2010). The gas was then washed with sodium hydroxide in two gas-liquid contactors. Dust was then removed by a Venturi followed by an electric filter (Séché 2010). Waste water was treated with lime and complexing and flocculating agents. Solid residues from the incinerated soils and the wash-water treatment sludges were sent to a hazardous waste landfill. Although not all of the PCB was destroyed by incineration, we assumed that there were no emissions due to the very low residual concentration. The incineration procedure and process flowchart is shown in Fig. 2.

2.2.2 Data collection

Inventory data about the bioremediation processes were taken from laboratory-scale and pilot-scale experiments. When results from the pilot scale were not available, laboratory-scale data from Sangely's dissertation were used for extrapolation (Sangely 2010).

Data about incineration were taken from the "Registre Français des émissions Polluantes (*French pollutant emission register*)" website (http://www.pollutionsindustrielles. ecologie.gouv.fr/IREP/index.php), which provided information on the main direct emissions for the overall process at the incineration site. A large amount of natural gas was used for the incineration; the LCA impact from its production was taken into account by calculating the quantity of natural gas Eq. 1 from the amount of CO_2 emitted.

Volume of natural gas used per incineration calculated from the quantity of CO_2 emitted:

$$V_{\rm gnv} = \frac{m_{\rm CO_2} \times I_{\rm p}}{\alpha} \tag{1}$$

where

 V_{gnv} is the volume of natural gas assumed to be consumed by the functional unit in cubic meter m_{CO2} is the mass of CO₂ emitted in kg UF⁻¹

- $I_{\rm p}$ is the percentage $\rm CO_2$ emitted attributed to the natural gas combustion
- α is the conversion coefficient (mass of CO₂ per volume of burned natural gas: in kilogram per cubic meter).

Finally, all the product and energy inventory data used in the procedure were obtained from the Ecoinvent European data table.



Fig. 1 Life cycle of the bioremediation procedure with the two aeration options proposed

2.2.3 Uncertainty analysis

Uncertainty analysis was applied to all inventory data according to the method in Frischknecht et al. (2007). For each data set, six parameters were qualitatively evaluated on a scale of 1–6, and an uncertainty factor was attributed to each evaluation using a correspondence table. The evaluated parameters and the corresponding uncertainty factors are given in Table 1 (Jolliet et al. 2005). If a parameter did not apply to the data, it was assigned a value of 1. The variance was calculated using Eq. 2.

Variance calculation:

$$V_{95\%} = \exp\sqrt{\sum_{n=1}^{7} \ln^2 U_n}$$
 (2)

where

- U_1 the uncertainty factor for the reliability parameter
- U_2 the uncertainty factor for the exhaustivity parameter
- U_3 the uncertainty factor for the temporal correlation parameter

- U_4 the uncertainty factor for the geographical correlation parameter
- U_5 the uncertainty factor for the technological correlation parameter
- U_6 the uncertainty factor for the sample size parameter
- U_7 the basic uncertainty factor. It depends on the emissions' measuring and modelling techniques.

The uncertainty factors have no units.

The Ecoinvent data used were primarily evaluated as a function of the different correlations. The relative uncertainty of the data was found using Eq. 3.

Calculation of relative uncertainties:

$$I_{\%} = \left(\sqrt{V_{95\%}} - 1\right) \times 100 \tag{3}$$

where $I_{\%}$ is the relative uncertainty of the data expressed as percent.

2.3 Impact assessment

Impact evaluations were made using the CML 2001 calculation method for 13 chosen midpoint impact categories:



Fig. 2 Life cycle of the incineration procedure

human toxicity, terrestrial ecotoxicity, photochemical oxidation (low NOx), ionizing radiation, freshwater sedimentary ecotoxicity, freshwater aquatic ecotoxicity, marine sedimentary ecotoxicity, marine aquatic ecotoxicity, ozone layer depletion (Steady state), global warming (100 year horizon), eutrophication, acidification and abiotic depletion.

The results were normalized with factors from Western Europe obtained in 1995 (Huijbregts et al. 2003) (Table 2). The inventory and impact calculations were made using Microsoft Excel and SimaPro[®] software.

2.4 Iso-distance impacts calculation

The soil transportation distance was a relevant parameter in this study; therefore, the influence of the location of polluted soil was calculated. To this end, Δd defines the difference in distance from which the impacts of the two scenarios compensated for each other in a given category. Only the transport-related impacts had a linear relation to distance. Equation 4 provides the iso-impacts distance. If $\Delta d < 0$, the distance favored scenario 3 (incineration); if $\Delta d > 0$, the distance favored scenario 1 (bioremediation).

Table 1	Uncertainty factors
correspo	nding to possible
quality s	cores for each parameter
(Jolliet e	t al. 2005)

Quality score	Reliability	Exhaustivity	Temporal correlation	Geographical correlation	Technological correlation	Sample size correlation	
1	1	1	1	1	1	1	
2	1.05	1.02	1.03	1.01	-	1.02	
3	1.1	1.05	1.1	1.02	1.2	1.05	
4	1.2	1.1	1.2	_	1.5	1.1	
5	1.5	1.2	1.5	1.1	2	1.2	

Table 2Normalisation factorsfor West Europe, 1995(Huijbregts et al. 2003)

Impact category	Normalisation factor	Unit
Global warming (100 year horizon)	$2.08 10^{-13}$	kg CO ₂ eq. y ^{-1}
Ozone layer depletion (steady state)	$1.20 10^{-08}$	kg CFC-11 eq. y^{-1}
Human toxicity (infinite)	$1.32 10^{-13}$	1.4-dichlorobenzene eq. y^{-1}
Terrestrial ecotoxicity (infinite)	$2.12 10^{-11}$	kg 1.4-dichlorobenzene eq. y ⁻¹
Freshwater aquatic ecotoxicity (infinite)	$1.98 10^{-12}$	kg 1.4-dichlorobenzene eq. y ⁻¹
Freshwater sedimentary ecotoxicity (infinite)	$1.93 10^{-12}$	kg 1.4-dichlorobenzene eq. y ⁻¹
Marine aquatic ecotoxicity (infinite)	$8.81 \ 10^{-15}$	kg 1.4-dichlorobenzene eq. y ⁻¹
Marine sedimentary ecotoxicity (infinite)	9.62 10^{-15}	kg 1.4-dichlorobenzene eq. y^{-1}
Photochemical oxidation (low NOx)	$1.58 10^{-10}$	kg ethylene eq. y^{-1}
Eutrophication	$8.02 10^{-11}$	kg PO_4^{3-} eq. y ⁻¹
Acidification	$3.66 10^{-11}$	kg SO ₂ eq. y^{-1}
Abiotic depletion	$6.74 10^{-11}$	kg Sb eq. y^{-1}
Ionising radiation	$2.06 10^{-05}$	Daly y^{-1}

Calculation of the iso-impact distances:

$$\Delta d = d_2 - d_1 = \frac{I_{\text{scenario1}}(d_1 = 0) - I_{\text{scenario2}}(d_2 = 0)}{I_{\text{transport}}(d = d_1 = d_2 = 1)}$$
(4)

where

Δd	is the difference in distance travelled
	between the contaminated site and the
	treatment centre
d_1 and d_2	the distance travelled between the
	contaminated site and the treatment centre
	for scenario 1 and scenario 2 respectively
Iscenario1 and	the impact of scenario 1 and of scenario 2,
I _{scenario2}	respectively, in the case where the
	contaminated site is treated in situ
I _{transport}	the unitary impact of the transport of soil
-	over 1 km.

3 Results

3.1 Flow comparisons between the scenarios

Table 3 gives the main inputs necessary for each of the three scenarios. The amount of diesel fuel consumed covered transport and handling of the soil. The stated uncertainties were calculated using Eqs. 2 and 3. Bioremediation procedures required more diesel fuel, which was directly related to the distance traveled between the contaminated site and the treatment centers. In all scenarios, the truck capacity and mass of soil were the same; therefore, the frequency of transport did not vary. As a consequence, it had no influence on diesel consumption. Conversely, incineration consumed more water, especially for the treatment of the gaseous effluents.

Incineration produces direct emissions, and Table 4 shows the emission data declared for a hazardous waste

(including PCB-contaminated soils) incineration plant. This procedure was responsible for many of the direct emissions of heavy metals and chlorinated products into water. In the air, carbon dioxide was emitted from the combustion of the natural gas used for the incineration. The presence of residual dioxins/furans and PCB indicated that PCB was not totally destroyed by incineration. The uncertainty in these data was 40%, as calculated from the Ecoinvent uncertainty database concerning techniques for the determination of these types of emissions. However, there was much less uncertainty in the quantity of CO_2 produced; it was generally calculated from reliable and proven models.

3.2 Impact analysis

3.2.1 Bioremediation: mechanical versus electric aeration

Table 5 shows the results of the impact evaluations for the three scenarios. The biological treatment processes all exhibited the same impact on freshwater aquatic ecotoxicity $(1.3 \times 10^3 \text{ kg } 1,4\text{-dichlorobenzene eq.})$. The impact on human toxicity and ionizing radiation was greater for electric than for mechanical aeration because of the contribution of nuclear power in French electricity production. The other ten impact categories all favored electric aeration. With regard to the uncertainty and compared with the incineration results, both biologic techniques could be considered equivalent.

3.2.2 Bioremediation versus incineration

Because the orders of magnitude are the same between the two biological techniques, the comparison of these techniques with incineration led to identical conclusions, irrespective of the bioremediation scenario. Only the electric aeration scenario was evaluated because it is the best scenario in the French context. The incineration technique had

Table 3 Direct intrants (Sangely 2010)

Intrants	Units	Bioremediation with mechanical aeration		Bioremediation with electric aeration		Incineration	
		Value	Uncertainty (%)	Value	Uncertainty (%)	Value	Uncertainty (%)
Diesel fuel	kg/UF	1.5 10 ⁴	45	1.4 10 ⁴	45	2.65 10 ³	40
Water	m ³ /UF	$1.8 \ 10^2$	33	$1.8 \ 10^2$	33	1.11 10 ³	7
Electricity	kWh/UF	_	_	5.9 10 ³	43	_	_
Nitrates	kg/UF	$1.8 \ 10^3$	10	$1.8 \ 10^3$	10	_	_
Low density polyethylene	kg/UF	$7.7 \ 10^2$	23	$7.7 \ 10^2$	23	_	_
Distance between contaminated site and remediation centre ^a	km	750	10	750	10	40	10
Distance between remediation centre and hazardous waste landfill ^a	km	290	10	290	10	170	10
Mass-kilometre	tkm/UF	6.6 10 ⁵	15	6.6 10 ⁵	15	$1.25 \ 10^5$	15
Natural gas	m ³ /UF	-	-	-	-	7.70 10 ⁵	34

^a Distance corresponds to specific case study. Bioremediation centre and corresponding hazardous waste landfill are located respectively in Lacq and in Graulhet (West-Southern France). Incinerator and corresponding hazardous waste landfill are located respectively in Saint-Vulbas and in Drambon (East-Southern France).

a greater impact than did bioremediation on the depletion of abiotic resources, the ozone layer, photochemical oxidation, marine ecotoxicity (aquatic and sedimentary) and global warming. For incineration and bioremediation, the impacts were similar for acidification and human toxicity. Finally, the bioremediation impacts exceeded those for incineration in eutrophication, terrestrial toxicity, freshwater ecotoxicity (aquatic and sedimentary) and ionizing radiation (Fig. 3). Furthermore, normalized results showed a higher weight for abiotic depletion, global warming and marine (aquatic and sedimentary) ecotoxicity (see Fig. 3). Other impact categories did not appear with significant weight. These results confirmed that the environmental impacts of biological treatment were less than those of incineration.

The impact on global warming was nine times greater for incineration than for the biological soil treatment procedures, producing 6.5×10^5 and 7.2×10^4 kg eq. CO₂, respectively. Carbon dioxide (CO₂), nitrous oxide (N₂O) and methane (CH₄) contributed the most to this effect (Table 6). The incineration stage of the incineration scenario was responsible for 94% of the impact on climate warming. For bioremediation, the transport phase contributed 75% of this impact (Fig. 4). This result partially confirmed the conclusions of Diamond et al. (1999). The production of nitrates used for the biological

Table 4 Direct emissions fromincineration: quantities andemission compartment	Substance	Emission compartment	Quantity emitted per functional unit in kg ^a	Uncertainty (%)
	Dioxins and furans (DRIRE Rhône-Alpes 2003)	Air	$1.5 \ 10^{-6}$	40
	Polychlorinated biphenyls (PCB)	Air	$3.0 \ 10^{-3}$	40
	Total CO ₂	Air	6.1 10 ⁵	4
	Arsenic and its compounds (As)	Eau	$5.0 10^{-1}$	40
	Polychlorinated biphenyls (PCB)	Eau	$2.1 \ 10^{-3}$	40
	Cadmium and its compounds (Cd)	Eau	$4.0 10^{-1}$	40
	Chlorine (total Cl)	Eau	$5.4 10^4$	40
	Organohalogen compounds (AOX)	Eau	$4.5 \ 10^{1}$	40
	Copper and its compounds (Cu)	Eau	1.7	40
	Fluorine (total F)	Eau	$1.0 \ 10^2$	40
	Mercury and its compounds (Hg)	Eau	$1.8 10^{-1}$	40
	Nickel and its compounds (Ni)	Eau	$5.0 10^{-1}$	40
	Lead and its compounds (Pb)	Eau	$5.0 10^{-1}$	40
	Trichlorobenzenes (TCB)	Eau	$3.0 10^{-3}$	40
^a Source: http://www.pollution	Tetrachloroethylene (PER-perchloroethylene)	Eau	$1.5 10^{-1}$	40
sindustrielles.ecologie.gouv.fr/ IREP/index.php	Zinc and its compounds (Zn) (2007)	Eau	6.2	48

Table 5 Score of intermediary impacts for each scenario

Impact category	Reference unit	Bioremediation with mechanical aeration		Bioremediation with electric aeration		Incineration	
		Mean	Standard error (%)	Mean	Standard error (%)	Mean	Standard error (%)
Global warming (100 year horizon)	kg CO ₂ eq.	7.5 10 ⁴	16	7.2 10 ⁴	15	6.5 10 ⁵	6
Ozone layer depletion (steady state)	10^{-3} kg CFC-11 eq.	9.3	16	8.9	15	21.6	33
Human toxicity (infinite)	10 ⁴ kg 1.4-dichlorobenzene eq.	1.2	20	1.0	16	1.1	34
Terrestrial ecotoxicity (infinite)	kg 1.4-dichlorobenzene eq.	106	14	158	23	35	29
Freshwater aquatic ecotoxicity (infinite)	10 ³ kg 1.4-dichlorobenzene eq.	1.3	15	1.3	14	0.6	29
Freshwater sedimentary ecotoxicity (infinite)	10 ³ kg 1.4-dichlorobenzene eq.	3.2	15	3.1	14	1.4	29
Marine aquatic ecotoxicity (infinite)	10 ⁶ kg 1.4-dichlorobenzene eq.	4.9	16	4.6	14	9.6	33
Marine sedimentary ecotoxicity (infinite)	10 ⁶ kg 1.4-dichlorobenzene eq.	4.2	16	4.0	14	8.2	33
Photochemical oxidation (low NOx)	kg ethylene eq.	4.3	20	3.8	16	8.4	33
Eutrophication	kg PO_4^{3-} eq.	43.8	19	38.3	15	14.2	27
Acidification	kg SO ₂ eq.	230	18	207	16	139	30
Abiotic depletion	kg Sb eq.	446	17	427	16	$1.7 \ 10^3$	33
Ionising radiation	10^{-5} Daly	3.5	16	18.9	38	2.1	21



HT Inf. = Human Toxicity Infinite

TE Inf. = Terrestrial Ecotoxicity Infinite

Ph Ox = Photochemical Oxidation (Low Nox)

IR = Ionizing Radiation

FWSET Inf. = Freshwater Sedimentary Ecotoxicity Infinite

FWAET Inf. =Freshwater Aquatic Ecotoxicity Infinite

MSET Inf. = Marine Sedimentary Ecotoxicity Infinite

MAET Inf. = Marine Aquatic Ecotoxicity Infinite

OLD = Ozone Layer Depletion (Steady state) GW = Global Warming (100 yr horizon)

Eut = Eutrophication

Aci = Acidification

AD = Abiotic Depletion



Impact category	Reference unit	Substances					
		Air	Water	Soil			
Global warming (100 year horizon)	kg CO ₂ eq.	Fossil CO ₂ , nitrous oxide, fossil methane	-	-			
Ozone layer depletion (steady state)	kg CFC-11 eq.	HCFC-22, halon 1301, halon 1211	_	-			
Human toxicity (infinite)	kg 1.4-DB eq.	Vanadium, HAP, nitrogen oxides, nickel, dioxins, copper, chrome VI, cadmium, benzene, arsenic	HAP, barium, barytite	-			
Terrestrial ecotoxicity (infinite)	kg 1.4-DB eq.	Vanadium, nickel, mercury, arsenic	-	Zinc, chrome VI, barium			
Freshwater aquatic ecotoxicity (infinite)	kg 1.4-DB eq.	Vanadium, nickel	Zinc ions, vanadium ions, HAP, nickel ions, copper ions, cobalt, beryllium, barium	Zinc, barium			
Freshwater sedimentary ecotoxicity (infinite)	kg 1.4-DB eq.	Vanadium, nickel	Vanadium ions, nickel ions, cobalt, beryllium, barium, barytite	Barium			
Marine aquatic ecotoxicity (infinite)	kg 1.4-DB eq.	Vanadium, nickel	Vanadium ions, nickel ions, cobalt, barium, barytite	Barium			
Marine sedimentary ecotoxicity (infinite)	kg 1.4-DB eq.	Vanadium, nickel	Zinc ions, vanadium ions, HAP, nickel ions, copper ions, cobalt, barium	Zinc, barium			
Photochemical oxidation (low NOx)	kg C_2H_4 eq.	Toluene, propane, pentane, fossil methane, hexane, heptane, ethane, carbon monoxide, butane	_	-			
Eutrophication	kg PO_4^{3-} eq.	Nitrogen oxides, ammonia	Phosphate, chemical oxygen demand	-			
Acidification	kg SO ₂ eq.	Sulfur dioxide, nitrogen oxides, ammonia	-	_			
Abiotic depletion	kg Sb eq.	-	_	Petrol, natural gas, coal			
Ionising radiation	Daly	Radon 222, carbon 14	-	-			

Table 6 Characterisation of substances contributing over 1% to intermediary impacts

treatment was responsible for 21% of greenhouse gas emissions. Conversely, the excavation undertaken for soil handling did not generate significant impacts (see Fig. 4).

Oil, natural gas and coal were decreased the most (see Table 6) during the bioremediation process. The consumption of oil, natural gas and coal for incineration and bioremediation was 138, 2992 and 24 GJ and 720, 120 and 45 GJ, respectively. The difference in quantities of oil could be linked, for

the most part, to the different soil transport distances (see Table 4). The high quantities of natural gas used for incineration were for the process itself.

Ecotoxicological impacts on marine, freshwater and terrestrial categories were due to vanadium, nickel, zinc and barium (see Table 6). In addition to these substances, human toxicity was subject to the effects of polycyclic aromatic hydrocarbons (PAHs), dioxins (mainly incineration), benzene and some







Incineration

heavy metals (copper, mercury, cadmium, chromium(VI), arsenic). These emissions were related to transport (Spielmann et al. 2007). The most important impact phases of the bioremediation scenario were transport and nitrates production; these phases accounted for a total of almost 90% of the impacts. For the incineration scenario, natural gas production accounted for more than 70% of the impact.

Ionizing radiation was a very strong impact from the bioremediation scenario because of electric aeration. More than 80% of the impact was due to the production of electricity (see Fig. 4).

Finally, acidification, ozone-layer depletion and photochemical oxidation were also largely subject to the influence of transport under bioremediation, with impacts greater than 70%. Overall, natural gas production was responsible for almost 90% of impacts in these categories (see Fig. 4).

4 Discussion

4.1 Distance of transport

Transport was an important impact factor (see Fig. 4), especially for the bioremediation scenarios. However, the baseline hypothesis used a transport distance of 750 km for bioremediation against 40 km for incineration. Therefore,

Table 7 Critical distances for iso-impacts

it seemed relevant to simulate variations in the distance that separated a contaminated site for the two remediation centers. From the results, in the case of global warming, it was "better" for the environment to biologically treat soil up to a distance of approximately 12,600 km over that of the incineration center (Table 7). In the same vein, for depletion of resources, the balance favored bioremediation at a distance disparity of 4700 km. However, for ionizing radiation, there were fewer impacts of incineration compared with biological treatment with mechanical aeration, up to a distance difference of approximately 8000 km. The iso-impact distance differences were larger in absolute values when bioremediation was favorable.

4.2 Electricity production mix

Electric aeration depends on the electricity production mix. French production is very specific, with 77% of the production from nuclear power, which is an energy that features a low carbon footprint. To test this hypothesis, the bioremediation scenario with electric aeration was calculated with the European electricity mix and compared to the French reference. Figure 5 shows that eight impacts remained the same, irrespective of the production mix. The impact on ionizing radiation was twice as high for the French mix as for the European mix, which confirmed that nuclear power

Impact category	Impacts		Critical distances in kilometres					
	Unit	Bioremediation (mechanical aeration) distance = 0	Bioremediation (electric aeration) distance = 0	Incineration PCB contaminated soil distance = 0	Transport	$d_{ m biomeca}-d_{ m inc}$	$d_{ m bioelec} - d_{ m inc}$	$d_{ m biomeca}-d_{ m bioelec}$
Global warming	kg CO ₂	38739	35881	651632	49	-12618	-12677	59
(100 year horizon) Ozone layer depletion (steady state)	eq. kg CFC- 11 eq.	0	0	0	0	-2386	-2442	56
Human toxicity (infinite)	kg 1.4-DB	8611	7040	10834	4	-534	-912	378
Terrestrial ecotoxicity (infinite)	kg 1.4-DB	78	129	34	0	1177	2545	-1369
Freshwater aquatic	kg 1.4-DB	933	902	562	1	716	656	60
Freshwater sedimentary ecotoxicity (infinite)	eq. kg 1.4-DB eq.	2230	2152	1356	1	693	631	62
Marine aquatic ecotoxicity (infinite)	kg 1.4-DB	2935975	2729575	9491286	2550	-2571	-2652	81
Marine sedimentary ecotoxicity (infinite)	kg 1.4-DB	2554687	2373609	8151181	2174	-2574	-2658	83
Photochemical oxidation (low NOx)	kg C ₂ H ₄	2	2	8	0	-2310	-2539	230
Eutrophication	kg PO ₄ eq.	25	20	13	0	475	256	219
Acidification	kg SO ₂ eq.	130	108	134	0	-25	-197	172
Abiotic depletion	kg Sb eq.	214	195	1667	0	-4709	-4771	62
Ionising radiation	Daly	0	0	0	0	50	8121	-8070

is importance in French production. The final four categories (freshwater and marine water ecotoxicity (aquatic and sedimentary)) differed by less than a factor two. In regards to this sensitivity analysis, the electricity production mix did not significantly influence the environmental impacts of bioremediation with electrical aeration. The LCA results could be applied across Europe.

4.3 Limits

Bioremediation occurs on a simple platform composed primarily of concrete with a pump (in the case of electric aeration) and a few piezometers. Incineration requires the construction of a complex plant with a multitude of inputs. Nevertheless, incineration facilities for the treatment of PCB and other hazardous wastes are not built exclusively for contaminated soil. The share of impacts from the burning of soil might be negligible. In addition, biological treatment requires 9 months, whereas incineration of the same quantity of soil requires a few hours to a few days. Land-use evaluation could also be relevant because biological treatment would require a larger surface to treat a high quantity of soil. The remediation market increased in the 2000s in France; however, the actual market/demand for decontamination remains very difficult to ascertain. Therefore, it has not been possible to include infrastructure impact in the limits of this work (Suez 2006).

Apart from natural gas, the main inputs of the incineration procedure, such as the lime and caustic soda, were not possible to estimate.

The potential gas emissions (CO_2 for the aerobic phase, CH_4 for the anaerobic phase) of the biological breakdown of soil are not known. The influence of the soil composition on homogenization during aeration is also unknown. The new biological process and the tried-and-tested incineration process must be compared with respect to the robustness and reliability of each technique.

Only secondary impacts have been evaluated in this lifecycle assessment. The "do nothing" scenario, which includes the primary impacts and an evaluation of the tertiary impacts, has not been explored. To this end, other methodologies, such as ecological risk assessment or health risk assessment, could be applied to complete the LCA results (Payet 2008).

The stability of the deposited soil or clinker at a landfill could be measured to extend the system boundary.

The CML 2001 method was chosen even though it does not evaluate the impact of PCB emissions into the environment. Nevertheless, the calculation of the latter using another method, such as IMPACT 2002+, produces a relatively similar picture with PCBs having less influence on the overall results.

5 Conclusions

A life-cycle assessment of three remediation scenarios for PCB-contaminated soils was analyzed. The attributional analysis highlighted the importance of soil transport, particularly for the two bioremediation processes. The incineration phase of the third procedure is responsible for the majority of the impacts. Biological treatment appears more environmentally friendly, especially in terms of global warming and depletion of abiotic resources. Furthermore, bioremediation with mechanical aeration has greater impacts than electric aeration. Nonetheless, the relative difference between these two scenarios remains small because of the preponderance of the transport factor and because of the small influence of the electricity production mix on the results. The results of the LCA of the two technologies for PCB-contaminated soil remediation showed that this study could be a relevant basis on which to choose a soil remediation technique according to environmental criteria.

The study and its limits demonstrate the necessity for indepth knowledge of the incineration procedure inputs and of

Fig. 5 Influence of electricity mix on environmental impacts of bioremediation techniques with electric aeration. European electricity mix (*BE/RER*) and French electricity mix (*BE/FR*) are compared



the technical parameters of bioremediation processes. LCA results should also be completed with other methodologies to answer questions about ecological risks, health risks and social acceptability.

Acknowledgements We would like to thank the Agence Nationale pour la Recherche et la Technologie (*French National Agency for Research*) for their financial support under the aegis of a CIFRE thesis contract (n° de convention 0046/2007).

References

- Blanc A, Metivier-Pignon H, Gourdon R, Rousseaux P (2004) Life cycle assessment as a tool for controlling the development of technical activities: application to the remediation of a site contaminated by sulfur. Adv Environ Res 8(3–4):613–627
- Cadotte M, Deschênes L, Samson R (2007) Selection of a remediation scenario for a diesel-contaminated site using LCA. Int J Life Cycle Assess 12(4):239–251
- Diamond ML, Page CA, Campbell M, McKenna S, Lall R (1999) Lifecycle framework for assessment of site remediation options: method and generic survey. Environ Toxicol Chem 18(4):788–800
- Frischknecht R, Jungbluth N, Althaus H-J, Doka G, Dones R, Hischier R, Hellweg S, Nemecek T, Rebitzer G, Spielmann M (2007) Overview and methodology. Final report Ecoinvent data v2.0, No. 1. Swiss Centre for Life Cycle Inventories, Dübendorf
- Hu X, Zhu J, Ding Q (2011) Environmental life-cycle comparisons of two polychlorinated biphenyl remediation technologies: incineration and base catalyzed decomposition. J Hazard Mater 191(1–3):258–268
- Huijbregts MAJ, Breedveld L, Huppes G, de Koning A, van Oers L, Suh S (2003) Normalization figures for environmental life-cycle assessment: The Netherlands (1997/1998), Western Europe (1995) and the world (1990 and 1995). J Clean Prod 11(7):737–748
- Jolliet O, Saadé M, Crettaz P (2005) Analyse du cycle de vie. Comprendre et réaliser un écobilan. Collection Gérer l'Environnement. Presse Polytechniques et Universitaires Romandes, Lausanne
- Lemming G, Hausshild MZ, Bjerg PL (2010) Life cycle assessment of soil and groundwater remediation technologies: literature review. Int J Life Cycle Assess 15(1):115–127
- Lesage P, Ekvall T, Deschênes L, Samson R (2007a) Environmental assessment of brownfield rehabilitation using two different life cycle inventory models Part 1: methodological approach. Int J Life Cycle Assess 12(6):391–398
- Lesage P, Ekvall T, Deschênes L, Samson R (2007b) Environmental assessment of brownfield rehabilitation using two different life cycle inventory models Part 2: case studies. Int J Life Cycle Assess 12(7):497–513

- Morais SA, Delerue-Matos C (2009) A perspective on LCA application in site remediation services: critical review of challenges. J Hazard Mater 175(1–3):12–22
- Morris M, Yuracko K, Govers RA (2000) Life cycle analysis for treatment and disposal of PCB waste at Ashtabula and Fernald. http://www.ornl.gov/~webworks/cpr/v823/rpt/108437.pdf Accessed 31 August 2011
- Page CA, Diamond ML, Campbell M, McKenna S (1999) Life cycle framework for assessment of site remediation options: case study. Environ Toxicol Chem 18(4):801–810
- Payet J (2008) Integrating Multiple Scale Impact Assessment on ecosystems for contaminated site management (MuSA Project). Deliverable 2.1: Similarities and differences between EcoRA and LCA. SNOWMAN project SN01/18. www.snowman-era.net/ downloads/MUSA deliverable21.pdf Accessed 5 December 2010
- Rahuman M, Pistone L, Trifirò L, Miertus S (2000) Destruction technologies for polychlorinated byphenils (PCB). http://www.clu-in. org/s.focus/c/pub/i/834/ Accessed 31 August 2011
- Sangely M, Sablayrolles C, Vialle C, Strehaiano P, Thannberger L, Montrejaud-Vignoles M (2009) Polychlorinated biphenyls fractioning in aqueous bioremediation assay with *Phanerochaete chrysosporium*. Int J Environ Anal Chem 89(8):849–856
- Sangely M (2010) Biodegradation of polychorinated biphenyls. Dissertation, INP-University of Toulouse, Toulouse
- Séché Environnement (2010) Savoir-faire spécifiques http://www. groupe-seche.com/FR/Savoir-faires-specifiques_47.html Accessed 2 December 2010
- Shen C, Tanga X, Cheemaa SA, Zhanga C, Khana MI et al (2009) Enhanced phytoremediation potential of polychlorinated biphenyl contaminated soil from waste recycling area in the presence of randomly methylated cyclodextrins. J Hazard Mater 172(2–3): 671–1676
- Spielmann M., Bauer C, Tuchschmid M (2007) Life cycle inventories of transport services. Final report Ecoinvent v2.0 No. 14. Swiss Centre for Life Cycle Inventories, Dübendorf
- Suer P, Nilsson-Påledal S, Norrman J (2004) LCA for site remediation: a literature review. Soil Sediment Contam 13(4):415–425
- Suez Environnement (2006) Grands savoirs environnementaux: dépollution des sites—terres polluées. http://www.suez-environnement. fr/gse/depollution/fr/pdf/dossier_information.pdf Accessed 30 August 2011
- Toffoletto L, Deschênes L, Samson R (2007) LCA of ex-situ bioremediation of diesel-contaminated soil. Int J Life Cycle Assess 10 (6):406–416
- Volkwein S, Hurtig HW, Klopffer W (1999) Life cycle assessment of contaminated sites remediation. Int J Life Cycle Assess 4(5):263– 274
- Zhou W, Anitescu G, Tavlarides LL (2004) Supercritical fluid extraction–oxidation technology to remediate PCB-contaminated soils/ sediments: an economic analysis. Environ Prog 23(3):222–231