

Pilot-Scale Decontamination of Soil Polluted with As, Cr, Cu, PCP, and PCDDF by Attrition and Alkaline Leaching

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Abstract: Recently, an efficient and promising process was developed to allow the removal of As, Cr, Cu, pentachlorophenol (PCP), and polychlorodibenzo-dioxins and furans (PCDDF) from soil using alkaline leaching. The present study evaluates the performance and the robustness of this decontamination process for the treatment of four different polluted soils by attrition and alkaline leaching at a pilot scale. The attrition process carried out on the coarse fraction (>0.125 mm) allowed the removal of 24–42% of As, 0–13% of Cr, 23–46% of Cu, 0–85% of PCP, and 17–64% of PCDDF from the different contaminated soils. Removal yields of 87–95% of As, 50–72% of Cr, 73–84% of Cu, 52–100% of PCP, and 27–73% of PCDDF were obtained after three leaching steps ($[\text{NaOH}] = 1$ M; $[\text{Cocamidopropylbetaine—BW}] = 3\%$ (w/w); $t = 2$ h; pulp density $[\text{PD}] = 10\%$ [w/v]) conducted on the fine fraction (<0.125 mm). The performance of both attrition and alkaline leaching processes seemed to be influenced by the nature of the soil and the type and initial level of contaminants present in the soils. However, the entire leaching process seemed to be highly efficient, allowing the simultaneous reduction of concentrations of inorganic and organic contaminants. The cost, including direct and indirect costs, were estimated between US\$214 and 454 per ton of treated soil, depending on the nature of the soil and the initial level of contamination. DOI: 10.1061/(ASCE)EE.1943-7870.0001255. © 2017 American Society of Civil Engineers.

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Introduction

Over the last few decades, soil contaminated by organic and/or inorganic compounds have become a major concern, affecting human health and causing a serious threat to the environment (Napola et al. 2006; Mouton et al. 2009). The primary reasons for soil contamination are improper industrial discharge, inappropriate disposal of wastes, mine tailings, the use of pesticides, combustion and the industry of wood preservation, and certain natural resources (Kulkarni et al. 2008; Mao et al. 2015). In North America, numerous sites contaminated by both organic and inorganic compounds

requiring remediation have been listed, including wood preservation industry sites. Since the 1970s, preservative agents have been applied to wood structures to protect them from insects and fungal attacks and enhance their service lifetime by 20–50 years (Janin 2009). Over the last few decades, the most commonly used wood preservative agents are chromated copper arsenate (CCA) and pentachlorophenol (PCP). The leaching of these preservative agents [As, Cr, Cu, PCP, and polychlorodibenzo-dioxins and furans (PCDDF)] from treated wood structures led to the contamination of soil by both organic and inorganic compounds. Indeed, some researchers highlighted that the inappropriate management and/or storage of treated wood during the last few decades is responsible for the contamination of several sites across the world (Cooper and Ung 1997; Stefanovic and Cooper 2005; Hasan et al. 2010). Usually, these soils are contaminated by metals (As, Cr, Cu) and organic compounds, such as PCP and PCDDF (Reynier 2012).

Currently, the only available options for the remediation of soils contaminated by both organic and inorganic compounds are limited to excavation, followed by thermal desorption to destroy organic contaminants, followed by the immobilization of inorganic contaminants or excavation, followed by disposal at off-site secured waste landfill sites (Dermont et al. 2008). However, these management options are becoming nonpreferable owing to the regulations regarding incineration and landfill leachate controls, which are becoming increasingly stringent (Reynier et al. 2013). In the last few years, various processes including thermal, biological, and physicochemical technologies have been the subject of several studies to allow the rehabilitation of soils contaminated by As, Cr, Cu, PCP, and PCDDF (Mouton et al. 2009). The thermal methods showed good results for PCP and PCDDF removal, but these technologies are inefficient for the removal of metals from contaminated soils (Lecomte 1998; Reynier 2012; Dufresne 2013). Kasai et al. (2004) showed that a thermal desorption method that

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allows more than 98.9% removal of PCDDF in a laboratory-scale experiment could be efficiently used for the remediation of contaminated soil. Bioremediation methods usually referred to the use of microorganisms such as fungi and bacteria to break down complex organic contaminants into simpler compounds such as CO₂, H₂O, CH₄, and chlorine (Doyle 2008). However, these methods required long periods to remove or degrade organic contaminants, ranging from a few weeks to a few months, and their applicability to soils contaminated by metals and PCDDF is restricted (Lecomte 1998). Soil washing is another method of treatment that can be used to remediate both organic and inorganic contaminants using physicochemical treatment methods. Chemical extraction can be achieved using different reagents including inorganic (sulfuric, nitric, phosphoric acids) or organic acids (acetic, citric acid) (Subramanian et al. 2010; Lafond et al. 2012), chelating agents [ethylenediaminetetraacetic acid (EDTA), ethylenediamine disuccinic acid (EDSS)] (Rivero-Huguet and Marshall 2011; Pocięcha and Lestan 2012; Voglar and Lestan 2013), or surfactants [Tween 80 (TW), cocamidopropylhydroxysultaine (CAS), cocamidopropylbetaine (BW), Brij 35, and Brij 98] (Mouton et al. 2009; Rivero-Huguet and Marshall 2011; Reynier 2012; Torres et al. 2012). Physical separation may be performed alone or in combination with chemical treatments to enhance the performance of contaminant removal from coarse soil fraction while reducing the operational costs. Physical separation methods such as mechanical screening, froth flotation, magnetic separation, attrition scrubbing and hydrodynamic classification can be efficiently used to remove metals from soil and concentrate these contaminants into small quantities of soil, especially for the rehabilitation of sites with large amounts of contaminated soil owing to the low operational costs of these technologies (Dermont et al. 2008). The most common physical separation method used for the decontamination of coarse particles of soil contaminated by both organic and inorganic compounds is attrition (Taillard 2010; Bisone 2012). Indeed, mechanical rotation used during the attrition processes caused collision between large particles that released the contaminants and concentrated them into the fine fraction (Bergeron et al. 1999). An efficient leaching process using flotation in acidic media (sulfuric acid) in the presence of an amphoteric surfactant (BW) was identified to remove organic compounds and metals from soil contaminated by As, Cr, Cu, PCP, and PCDDF. According to the authors, the results showed that removal yields of 82–93% of As, 30–80% of Cr, 79–90% of Cu, and 36–78% of PCP were obtained from soil initially containing [As]_i = 50–250 mg/kg, [Cr]_i = 35–220 mg/kg, [Cu]_i = 80–350 mg/kg, and [PCP]_i = 2.5–30 mg/kg (Reynier et al. 2013). Reynier (2012) also highlighted that more than 60% of As, 32% of Cr, 77% of Cu, and 87% of PCP can be removed from contaminated soil after three leaching steps, 2 h each, carried out at 80°C with pulp density (PD) fixed at 10% (w/w) in the presence of sodium hydroxide (0.5 M) and a surfactant [(BW) = 2%(w/w)]. However, the effluents produced during this leaching process contained high concentrations of contaminants and required a treatment to allow their recycling into the leaching process or their discharge in municipal sewers. Over the last few decades, precipitation and coagulation of metals have been the primary methods used for the removal of inorganic and organic contaminants from acidic or basic industrial effluents. Typically, metals can be precipitated as hydroxides, sulfides, or carbonates from the effluents. Previous studies showed the efficiency of ferric salts for the removal of As, Cr, and Cu from effluents (Blais et al. 2008; Coudert et al. 2014).

The objective of this study was to evaluate the performance and the robustness of an alkaline leaching process to remove As, Cr, Cu, PCP, and PCDDF from the fine soil fraction (<0.125 mm)

combined with an attrition process for the removal of contaminants from coarse particles (>0.125 mm) at a pilot plant scale to reduce the costs of decontamination.

Material and Methods

Soil Sampling and Characterization

Four soil samples (D1, G2, S1, and S3) contaminated by As, Cr, Cu, PCP, and PCDDF were collected from different industrial areas. For each site, excavation of contaminated soils was performed using an excavator at a depth of 0–30 cm. More than 25 kg of soils were collected and stored in high-density polyethylene (HDPE) containers. Soils were then wet-sieved for 20 min through four different sieves (12, 4, 1, and 0.125 mm) using a mechanical Sweco to determine the particle-size distribution of each soil and collect the different fractions used in this study (>12, 4–12, 1–4, 0.125–4, and <0.125 mm). For the determination of inorganic (As, Cr, Cu) and organic (PCP and PCDDF) contaminant contents in the different soil fractions, soil samples were crushed using a Frissh ball mill (Pulverisette model 6) to obtain homogenous samples.

Attrition Experiments

Fig. 1 shows a diagram of the entire process used for the decontamination of the four soils contaminated by As, Cr, Cu, PCP, and PCDDF. Attrition experiments were applied to the coarse fractions (4–12, 1–4, and 0.125–1 mm) of four different soil samples, which represented between 87 and 95% (w/w) of the total soil. Soil particles larger than 12 mm were not treated by attrition because of the low levels of inorganic (As, Cr, Cu) and organic (PCP, PCDDF) contaminations. The attrition process consisted of three 20-min attrition steps performed in a 10-L capacity stainless steel reactor equipped with three deflectors and a mechanical stirrer (Light EV1 P25 AXFLOW, New York). The mixing speed was fixed at 1,700 rpm. Attrition experiments were carried out onto 2 kg of soil sample mixed with tap water (pH around 7) to obtain a 40% PD (w/w). After each 20-min attrition step, treated soils were separated from the water using 2, 0.5, and 0.125 mm sieves for the 4–12, 1–4, and 0.125–1 mm soil fractions, respectively. The treated soil was then washed with 4 L of tap water before being reintroduced to the attrition process to undergo the next attrition step. After the third attrition step, the soil samples were transferred in a 20-L plastic reactor and were then rinsed with 4 L of tap water using a Karcher electric sprayer (140 kg/cm², Québec, QC, Canada) before being sieved. Approximately 2 L of tap water was sprayed on the different sieves (less than 0.125 mm for the 0.125–1 mm fraction, less than 0.5 mm for the 1–2 mm fraction, and less than 1 mm for the 2–4 mm fraction) to rinse each treated soil fraction. After the attrition process, each fraction of the different soils studied were dried at 60°C in a vacuum oven and analyzed to determine the residual concentrations of inorganic and organic contaminants.

Leaching Experiments

Leaching experiments were performed on the fine fraction (<0.125 mm) of the four different soils studied. The alkaline conditions of leaching (temperature, pH, PD, number of steps) were optimized in a previous study (Reynier 2012). The leaching process consisted of three leaching steps, 2 h each, followed by one rinsing step of 15 min. For all leaching assays, the PD was fixed at 10% (w/v) for the leaching steps and 28% (w/v) for the rinsing steps.

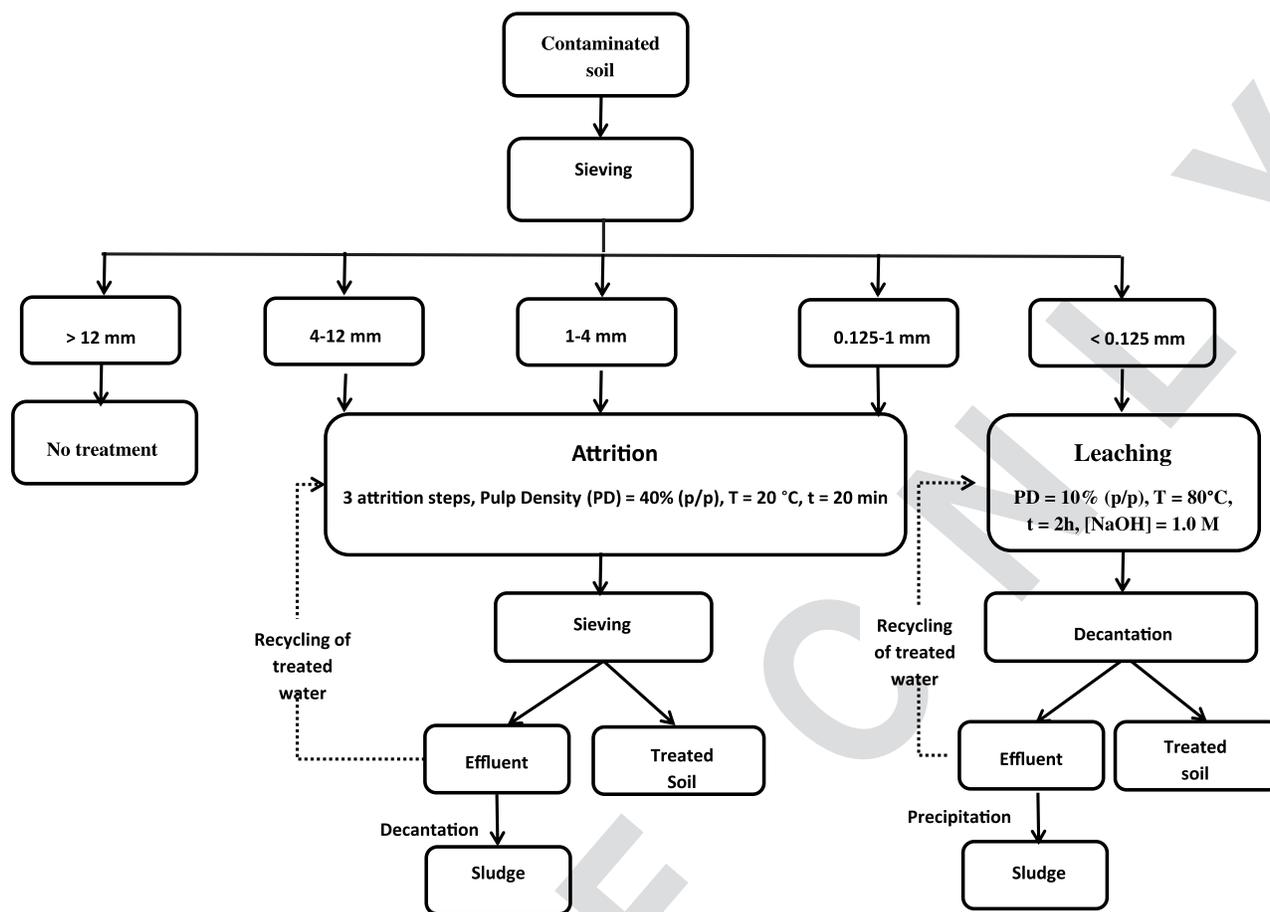


Fig. 1. Diagram of the treatment process applied for the rehabilitation of four different soils contaminated by As, Cr, Cu, PCP, and PCDDF

FI:1

184 Leaching experiments were performed in a 25-L capacity stainless
 185 steel reactor by mixing 2 kg of soil with 18 L of alkaline solution
 186 (1 M NaOH, Laboratories MAT, Quebec, Canada). The initial pH
 187 value of the pulp was around 13–13.5 all along the experiments
 188 (without any addition of acid or basis). To enhance the removal
 189 of organic contaminants, an amphoteric surfactant (BW) was added
 190 before each washing step, and its concentration was fixed at 3%
 191 (w/v). The leaching steps were conducted at $80 \pm 7^\circ\text{C}$ using a hot-
 192 plate (Thermo Scientific Remote-Control Hotplates, Montreal, QC,
 193 Canada), whereas the rinsing steps were carried out at room tem-
 194 perature. During the leaching and rinsing steps, the mixing speed
 195 was fixed at 1,700 rpm using a mechanical stirrer (Light EV1 P25
 196 AXFLOW, New York, NY). After each leaching and rinsing step,
 197 solid-to-liquid separation was carried out by removing most of the
 198 liquid using a lamella settler with a capacity of 20 L, a settling area
 199 of 0.44 m^2 , a length of 21 cm, and a height of 32 cm (Fiberglass, 14
 200 lamella, Plastiver, Québec, QC, Canada). After the leaching and the
 201 rinsing steps, soil samples were collected and then dried at 60°C
 202 before being analyzed to determine the residual concentrations of
 203 PCP, PCDDF, and metals.

204 Alkaline Leachates Treatment by Precipitation

205 Leachate treatment was performed by precipitation–coagulation
 206 using 1 L of the effluent emerging from the first leaching step
 207 to concentrate the contaminants present in small amounts of sludge.
 208 The precipitation–coagulation experiments were performed in
 209 Imhoff cones. A solution of sulfuric acid (93% H_2SO_4) was used
 210 to reduce the pH of the leaching solution from 13.0–13.5 to

7.5–8.0. Between 2.95 and 4.30 g/L of a solution of ferric sulfate
 [9.65% of iron (w/w), Chemco, Saint-Augustin-de-Desmaures,
 QC, Canada] was added to the solution under agitation to improve
 the removal of As. Indeed, some research showed that the addition
 of ferric ions during the precipitation allowed the formation of a
 precipitate of iron arsenate ($\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$) and the adsorption
 of As ions onto ferric hydroxide molecules, thus improving the
 removal of As from effluents (Coudert et al. 2014). The addition
 of ferric sulfate led to an acidification of the solution that depended
 on the amount added. To avoid any resolubilization of As, Cr, Cu,
 and PCP resulting from the acidification observed, a solution of sodium
 hydroxide [$[\text{NaOH}] = 0.5 \text{ M}$] was then added to adjust the final
 pH of the solution between 7.0 and 7.3. The solution was then left
 to settle overnight, and the sludge produced was then collected and
 dried at 60°C . The concentrations of inorganic and organic contam-
 inants were determined in the different sludge samples collected.

Analytical Methods

Metal Analysis

228 Metal contents in the soil before and after treatment were deter-
 229 mined in the laboratories at the National Institute of Scientific Re-
 230 search (INRS) by inductively plasma coupled to atomic emission
 231 spectroscopy (ICP-AES) (Vista Ax CCO simultaneous ICP-AES,
 232 Varian, Mississauga, ON, Canada) after digestion of 0.5 g of dry
 233 soil samples in the presence of nitric and hydrochloric acids (HNO_3
 234 and HCl) according to Method 3030I (APHA 1999). Each soil sam-
 235 ple was digested and analyzed in triplicate. During each series of
 236

Table 1. Concentrations of Contaminants Measured in the Different Soil Fractions from Four Different Soils Studied

T1:	Soil	Fraction	Soil proportion (w/w) (%)	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	PCP (mg/kg)
T1:1	D1	>12 mm	22.8	38.4	26.0	50.0	0.36
T1:2		4–12 mm	23.8	37.2	134	61.0	0.04
T1:3		1–4 mm	22.5	19.6 ± 1.6	183 ± 4	88.0 ± 3.4	0.04
T1:4		0.125–1 mm	19.2	45.3 ± 4.8	143 ± 9	136 ± 6	0.23
T1:5		<0.125 mm	11.7	286 ± 22	374 ± 44	559 ± 73	4.43
T1:6		Entire soil	100	64.2	150	137	0.66
T1:7	G2	4–25 mm	42.1	52.2	24.0	98.0	0.15
T1:8		1–4 mm	19.4	70.5 ± 3.5	75.0 ± 2.8	134 ± 8	0.95
T1:9		0.125–1 mm	19.2	83.8 ± 5.3	153 ± 8	168 ± 8	0.13
T1:10		<0.125 mm	19.2	776 ± 6	575 ± 8	956 ± 7	5.95
T1:11		Entire soil	100	201	164	283	1.41
T1:12	S1	>12 mm	22.7	41.8	17.0	60.0	5.03
T1:13		4–12 mm	21.0	37.0	16.0	53.0	1.89
T1:14		1–4 mm	39.2	34.1 ± 0.9	258 ± 2	67.0 ± 1.4	6.17
T1:15		0.125–1 mm	13.4	52.2 ± 1.7	247 ± 8	89.0 ± 0.1	8.56
T1:16		<0.125 mm	3.70	544 ± 80	401 ± 59	681 ± 111	54.7
T1:17		Entire soil	100	57.7	156	88.1	7.12
T1:18	S3	>12 mm	15.6	7.33	7.00	27.0	2.10
T1:19		4–12 mm	23.4	7.05	7.00	25.0	8.40
T1:20		1–4 mm	27.9	7.19 ± 0.40	7.19 ± 0.20	7.05 ± 0.30	18.4
T1:21		0.125–1 mm	24.3	75.0 ± 4.3	66.0 ± 3.6	135 ± 8	21.3
T1:22		<0.125 mm	8.74	664 ± 8	509 ± 29	830 ± 11	191
T1:23		Entire soil	100	81.1	65.2	117	29.3
T1:24							

237 analysis, digestion blanks, reference certified soil samples [CNS
238 392-050, PQ-1, lot No. 7110C513, CANMET, Canadian Certified
239 Reference Materials Project (CCRMP)], and certified standard so-
240 lutions (Multi-elements standard, Catalog No. C00-061-403, SCP
241 Science, Lasalle, QC, Canada) were analyzed to ensure the quality
242 of the analysis.

243 Organic Contaminant Analysis

244 PCP contents in the soil before and after treatment were determined in
245 **10** the laboratories according to the Centre d'expertise en analyse envi-
246 ronnementale du Québec (CEAEQ) method (CEAEQ 2013) after the
247 solubilization of PCP from 20 g of soil by Soxhlet extraction in the
248 presence of methylene chloride (300 mL). PCP was then transferred
249 to an aqueous phase (solution of sodium hydroxide at 20 g/L) by
250 liquid/liquid extraction to perform a derivatization step in the presence
251 of anhydrous acetate and a solution of potassium carbonate (75%, v/
252 v). After 12 h, a liquid/liquid extraction step was carried out using
253 methylene chloride. A solution of phenanthren-*d*₁₀ (internal standard)
254 was added, and then PCP analysis was performed by gas chromatog-
255 raphy with mass spectroscopy (GC-MS) (Perkin Elmer, model
256 Clarus 500, column type Rxi-17, 30 m × 0.25 mm × 0.25 μm).

257 The analysis of the 17 toxic congeners of PCDDF was per-
258 formed according to the CEAEQ method MA. 400-D.F. 1.1
259 (CEAEQ 2011) by an external laboratory (Wellington Laboratories,
260 Guelph, ON, Canada).

261 pH Measurements

262 **11** The pH was determined using a pH-meter (Accumet Model 915)
263 equipped with a double junction Cole-Parmer electrode with an Ag/
264 AgCl reference cell. Before each series of measurements, certified
265 buffer solutions (pH = 2, 4, 7, and 10) were used to calibrate the
266 pH-meter. The total solid contents were measured according to the
267 APHA method 2540D (APHA 1999).

268 Economic Analysis

269 The direct and indirect costs related to the treatment of the four
270 different soils contaminated by As, Cr, Cu, PCP, and PCDDF

271 by attrition (>0.125 mm soil fraction) and alkaline leaching
272 (<0.125 mm) were estimated for a decontamination plant able to
273 treat 15,000 t of soil per year (operating period: 350 d/year; treat-
274 ment capacity: 48 t/d; operating efficiency factor: 90%). The
275 decontamination process plant was designed in countercurrent
276 mode, which means that the effluents produced were recycled into
277 the decontamination process to reduce the consumption of chem-
278 icals and water. In the economic analysis, the operating cost in-
279 cluded the costs related to the use of chemical products such as
280 H₂SO₄ [US\$80/t for a solution at 93% (w/w)], BW (US\$1/kg),
281 Fe₂(SO₄)₃ (US\$200/t), and NaOH (US\$500/t); the consumption
282 of electricity (US\$0.07/kWh), water (US\$0.50/m³), and fuel
283 (US\$3.50/t); and the labor (US\$25/h). The costs related to the
284 transport and disposal of contaminated soils (US\$120/t), highly
285 contaminated soils (US\$500/t for the transport, thermal destruc-
286 tion, and landfilling), and inorganic and organic hazardous wastes
287 including sludge coming from the precipitation-coagulation
288 (US\$500/t) were also included in the estimation of the direct costs.
289 The indirect costs included the administrative staff, insurance and
290 taxes, research and development, and capital costs. The capital
291 costs were evaluated using a 10-year reimbursement period for
292 all equipment required for the treatment of soils by attrition and
293 leaching and for the treatment of leachates by precipitation with
294 a 5% annual interest rate.

295 Results and Discussion

296 Soil Characteristics

297 Table 1 presents the concentration of As, Cr, Cu, PCP measured in
298 each soil fraction (>12, 4–12, 1–4, 0.125–1, and <0.125 mm) of
299 the four different soils studied and the soil fraction proportion. Ac-
300 cording to the particle-size distribution of the different soils, the
301 coarse particles (>0.125 mm) represented the majority of the soils
302 with proportions ranging from 80.8 to 96.3%, whereas fine par-
303 ticles (<0.125 mm) represented only between 3.7 and 19.2% of

Table 2. Concentrations of Each Dioxin and Furan Measured in the Different Soil Fractions from Four Different Soils Studied

T2:2 T2:3	Dioxin or furan	Toxicity factor ^a	Associated TEQ (ng/kg)				Associated TEQ (ng/kg)			
			D1(B)	G2(B)	S1(A)	S3(C)	D1(B)	G2(B)	S1(A)	S3(C)
			0.125–12 mm	0.125–25 mm	0.125–12 mm	0.125–12 mm	<0.125 mm	<0.125 mm	<0.125 mm	<0.125 mm
T2:4	Furans									
T2:5	2378-TCDF	0.1	0.18	0.40	0.24	1.43	4.59	2.50	3.38	9.98
T2:6	12378-PeCDF	0.05	0.44	0.87	0.64	3.66	8.80	5.30	5.70	19.4
T2:7	23478-PeCDF	0.5	4.15	9.25	6.50	33.2	92.5	46.8	65.0	266
T2:8	123478-HxCDF	0.1	9.88	28.2	23.8	204	203	226	239	1,480
T2:9	123678-HxCDF	0.1	9.10	22.8	14.1	118	221	140	154	775
T2:10	234678-HxCDF	0.1	16.7	53.3	29.1	305	361	376	298	321
T2:11	123789-HxCDF	0.1	0.41	2.33	1.03	10.7	6.78	9.75	8.55	2,020
T2:12	1234678-HpCDF	0.01	37.3	130	87.2	644	846	896	897	4,080
T2:13	1234789-HpCDF	0.01	2.58	10.7	6.03	61.0	57.1	78.4	62.6	393
T2:14	OCDF	0.001	21.4	83.6	65.6	389	405	674	576	2,020
T2:15	Dioxins									
T2:16	2378-TCDD	1	7.00	18.6	5.10	27.4	104	135	67.5	176
T2:17	12378-PeCDD	0.5	49.0	135	63.0	332	1,060	785	715	2,300
T2:18	123478-HxCDD	0.1	22.8	78.6	42.1	268	532	554	377	1,590
T2:19	123678-HxCDD	0.1	40.4	158	78.9	834	910	996	846	6,360
T2:20	123789-HxCDD	0.1	44.5	147	75.3	584	1,010	958	804	3,860
T2:21	1234678-HpCDD	0.01	131	774	303	3,010	2,680	4,520	2,720	12,800
T2:22	OCDD	0.001	123	837	276	2,360	2,840	5,830	2,630	7,300
T2:23	Total		520	2,489	1,078	9,185	11,342	16,233	10,469	45,770

^aFrom OTAN and CDSM (1988).

all soils. For soil samples G2, S1, and S3, the texture class of the entire soil is silty sand, whereas the texture class of D1 is sand.

The concentrations of As, Cr, Cu, and PCP measured in all soil samples were very different among the four soils studied, ranging from 57.7 to 201 mg As/kg, from 65.2 to 164 mg Cr/kg, from 88.1 to 283 mg Cu/kg, and from 0.66 to 29.3 mg PCP/kg. The distribution of the contaminants in the four different soils studied showed that the finer fraction (<0.125 mm) was most contaminated than the coarser fraction (>0.125 mm). According to Anderson et al. (1999), the increase of surface area, cationic exchange potential in the fine fraction, and the innate shape of silts and clays are important parameters favoring the attraction of contaminants to the fine fractions of soils. Dermont et al. (2008) also reported that the contaminants are generally associated with fine particles (clay and silt), which are more potentially reactive as they have a higher surface area than coarser particles.

Table 2 presents the toxicity factors and the associated toxic equivalent quantity (TEQ) values for each dioxin and furan congener present in the coarse fraction (>0.125 mm) and the fine fraction (<0.125 mm) of the four different soils. These results showed that the 17 congeners known to be toxic were present in the coarse and fine fractions of all soils. According to these results, the fixation of PCDDF seemed to more important and favorable on the finer fraction for the different soils; the concentrations of PCDDF were 5–22 times higher in the fine fraction (2,990–45,770 ng TEQ/kg) than in the coarse fraction (520–1,340 ng TEQ/kg). A comparison of the concentrations of dioxin and furans substituted at the same position showed that the concentrations of dioxin compounds seemed to be higher than those of furan compounds in the different soil fractions and for all of the soils studied.

Efficiencies of Attrition Treatment on the Coarse Fraction (>0.125 mm)

Three attrition steps 20 min each, carried out at room temperature with a pulp density fixed at 40% (w/w), were applied to the coarse fraction (4–12, 1–4, and 0.125–1 mm) of the different soils studied.

Performance of Attrition on Inorganic and Organic Contaminant Removal

Table 3 show the concentrations of As, Cr, Cu, PCP, and PCDDF measured in the recombined soil fraction (>0.125 mm) before and after treatment by attrition and the associated removal yields. After the treatment by attrition, the residual concentrations of inorganic contaminants in the coarse fraction (>0.125 mm recombined soil fraction) ranged from 19 to 37 mg As/kg, from 22 to 195 mg Cr/kg, and from 41 to 67 mg Cu/kg. The entire attrition process allowed the removal of 24–42% of As, 0–13% of Cr, and 23–46% of Cu. According to a study by Williford et al. (1999), a pretreatment by attrition allowed similar removal yields (26.8% for Cr) to those observed in the present study. The low efficiencies of As, Cr, and Cu removal observed during the attrition process can be

Table 3. Concentrations of Contaminants Measured in the Recombined 0.125–12 mm or 0.125–25 mm before and after Treatment by Attrition

Soils	Contaminants				
	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	PCP (mg/kg)	PCDDF (ng/kg)
	Before attrition				
D1	34.0	153	92.0	0.09	520
G2	64.0	67.0	123	0.36	2,489
S1	38.0	186	67.0	2.15	1,078
S3	28.0	25.0	53.0	11.8	8,885
	After attrition				
D1	26.0	133	63.0	0.05	332
G2	37.0	87.0	67.0	1.18	2,055
S1	22.0	195	46.0	0.33	387
S3	19.0	22.0	41.0	7.92	3,213
	Removal yield (%)				
D1	24	13	32	44	36
G2	42	0	46	0	17
S1	42	0	31	85	64
S3	32	12	23	33	64

Note: Three Attrition Steps, PD = 40%, T = 20°C, t = 20 min.

Table 4. Sludge Production and Contaminant Concentrations Measured in the Sludge Produced during the Treatment of the 0.125–12 mm or 0.125–25 mm Soil Fractions by Attrition

T4:1	Soils	D1	G2	S1	S3
T4:2	Dry sludge production (kg/t)	115	329	194	121
T4:3	As content (mg/kg)	69.0	110	84.0	73.0
T4:4	Cr content (mg/kg)	247	32.0	163	42.0
T4:5	Cu content (mg/kg)	229	219	125	119
T4:6	PCP content (mg/kg)	0.31	0.01	7.18	32.8
T4:7	PCDD/F content (ng TEQ/kg)	1,400	3,220	3,000	30,620

Note: Three Attrition Steps, PD = 40%, T = 20°C, t = 20 min.

attributed to the fact that the solubilization of these metals is unfavorable at pH = 7, and only a small proportion of the inorganic contaminants is fixed to the fine particles agglomerated to the coarse particles and dislodged by attrition. Usually, attrition is used as a pretreatment to enhance the performance of inorganic contaminant removal by gravimetric separation technologies and not as a treatment itself. However, the attrition process developed seemed to be efficient enough to remove As, Cr, and Cu from the coarse fractions and allow the potential reutilization of the treated soils, depending on the current national regulations.

Concerning the organic contaminants, the residual PCP and PCDDF concentrations measured in the coarse fraction ranged from 0.05 to 7.92 mg/kg and from 332 to 3,213 ng TEQ/kg, respectively. According to these results, the attrition process seemed to be more efficient for the removal of PCP and PCDDF from Soils S1 and S3, with removal yields ranging from 33 to 85% for PCP and 64% for PCDDF, than from Soils G2 and D1, with removal yields varying between 0 and 44% for PCP and between 17 and 36% for PCDDF. These results showed that the effect of attrition could be influenced by the nature of the soil and the type of contaminant. In the case of Soils G2 and D1, the performance of attrition to remove organic contaminants from the coarse particles might be improved by the use of an amphoteric surfactant, thus enhancing the solubilization of these hydrophobic organic contaminants.

Attrition Sludge Production

The attrition steps produced sludge that represented between 0.7 and 6.1% for the fraction 0.125–1 mm, between 8.1 and 31% for the fraction 1–4 mm, and between 8.0 and 20% for the fraction 4–12 mm of the total mass of the four different soils studied. Except for Soil S1, the amount of sludge produced during attrition increased as the size of the particles treated by attrition increased. This effect can be a result of the more important disintegration of agglomerated particles from the coarser soil fraction during attrition.

Table 4 presents the amount of sludge produced during attrition treatment of the coarse fraction (>0.125 mm) and the inorganic and organic contaminant contents measured in the sludge. Depending on the soils treated by attrition, the total amount of sludge produced varied between 11.5 and 32.9% of all soil treated (coarse and fine fraction). The treatment by attrition of the coarse fraction, which represented between 80.8 and 96.3% of the soil, allowed the concentration of both inorganic and organic contaminants in small amounts of sludge, except for Cr. The attrition sludge can be disposed of in landfill sites or must be treated or disposed of in secured landfill sites, depending on the residual concentrations of contaminants and the regulations.

These results highlighted that attrition is an inexpensive and promising technique to simultaneously remove inorganic and organic contaminants from the coarse particles of soil and concentrate

Table 5. Concentrations of Contaminants Measured in the Soil Fraction <0.125 mm before and after Leaching Treatment

Soils	Contaminants					
	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	PCP (mg/kg)	PCDDF (ng/kg)	
Before leaching						
D1	286	374	559	4.43	11,342	T5:2
G2	776	575	965	5.9	16,233	T5:1
S1	554	401	681	54.9	10,469	T5:3
S3	664	509	830	191	45,800	T5:4
After leaching						
D1	30.0	168	149	2.12	7,439	T5:5
G2	99.0	286	234	0.60	11,917	T5:6
S1	33.0	196	174	22.3	2,778	T5:7
S3	31.0	141	129	0.28	15,391	T5:8
Removal yield (%)						
D1	90	55	73	52	34	T5:9
G2	87	50	76	90	27	T5:10
S1	94	51	74	59	73	T5:11
S3	95	72	84	100	66	T5:12

Note: Three Leaching Steps, PD = 10%, T = 80°C, t = 2 h, (NaOH) = 1.0 M.

them into small amounts of sludge, even if its performance seemed to vary, depending on the nature of both soil and contaminant and the initial levels of organic and inorganic contaminants.

Efficiencies of Chemical Leaching Treatment on the Fine Fraction

Three leaching steps 2 h each were applied to the fine soil fraction (<0.125 mm) of four different soils at 80 ± 7°C to evaluate the removal efficiencies of both organic and inorganic contaminants. The pulp density was fixed at 10% (w/w), and the leaching solution was composed of NaOH (1 M) and an amphoteric surfactant [(BW) = 3% (w/w)].

Performance of the Leaching Process on Inorganic and Organic Contaminant Removal

Table 5 shows the contents of As, Cr, Cu, PCP, and PCDDF measured in the fine soil fraction (<0.125 mm) before and after treatment by alkaline leaching and the associated removal yields. These results highlight the necessity to develop an efficient method of decontamination to allow the simultaneous removal of both organic (PCP and PCDDF) and inorganic (As and Cu) contaminants.

After three alkaline leaching steps, the residual concentrations of inorganic contaminants in the fine fraction (<0.125 mm) ranged from 30.0 to 99 mg As/kg, from 141 to 286 mg Cr/kg, and from 129 to 234 mg Cu/kg. These results show that the entire leaching process was quite effective in solubilizing inorganic contaminants, especially As and Cu, with removal yields ranging from 87 to 95% for As and from 73 to 84% for Cu, whereas it seemed to be relatively less effective in the removal of Cr (from 50 to 72% of Cr removed) from the different soils studied. These removal yields were slightly higher than those observed by Reynier et al. (2014) after three 2-h leaching steps carried out at 80°C in the presence of NaOH (0.75 M) and BW [3% (w/w)] with a PD fixed at 10% (w/w). Indeed, these authors obtained removal yields between 70 and 89% for As, between 23 and 66% for Cr, between 59 and 71% for Cu, and between 77 and 90% for PCP. The difference observed can be attributed to the fact that the concentration of NaOH used in the present study was fixed at 1 M, thus increasing the solubilization of metals under anionic forms. According to Reynier et al. (2015), As was mainly solubilized as AsO_4^{3-} and $HAsO_4^{2-}$ (<1%) during

Table 6. Concentration of Inorganic and Organic Contaminants in the Sludge Obtained after Treatment of the Leachate by Precipitation–Coagulation with Ferric Sulfate

T6:1	Soils	D1	G2	S1	S3
T6:2	Dry sludge production (kg/t)	33	30	11	37
T6:3	As content (mg/kg)	975	2,550	1,900	2,350
T6:4	Cr content (mg/kg)	964	1,280	930	1,500
T6:5	Cu content (mg/kg)	1,690	2,820	2,020	2,710
T6:6	PCP content (mg/kg)	11.1	20.0	140	699
T6:7	PCDD/F content (ng TEQ/kg)	23,400	24,200	30,800	128,000

alkaline leaching [(NaOH) = 0.75 M], whereas Cr and Cu were mainly solubilized as $\text{Cr}(\text{OH})_4^-$ and CrO_2^- for Cr and as $\text{Cu}(\text{OH})_4^{2-}$ and $\text{Cu}(\text{OH})_3^-$ (<1%) for Cu.

Concerning the organic contaminants, the residual PCP and PCDDF concentrations measured in the fine fraction ranged from 0.3 to 22 mg/kg and from 2,780 to 15,391 ng TEQ/kg (Table 5), respectively. According to these results, the alkaline leaching process seemed to be highly efficient for the removal of PCP and PCDDF from the different soils studied, except for the removal of PCDDF from Soils G2 and D1 and the removal of PCP from Soil S1. The alkaline leaching process developed in the present study allowed the removal of 52–100% of PCP and 27–73% of PCDDF. These removal yields were slightly lower than those observed by Reynier et al. (2014) after a similar leaching process carried out on 1–6 mm contaminated soil fraction (>92% for PCP and >81% for PCDDF). The differences observed could be explained by the fact that it is more difficult in the present study to efficiently remove organic contaminants from the fine fraction owing to their very high initial contents of PCP and PCDDF.

According to results of the present study, the performance of the leaching process in the removal of organic contaminants might be influenced by the nature of the soil and/or the type of contaminants. Indeed, the performance of the leaching process seemed to be more variable for the removal of PCP and PCDDF from soil than for As, Cr, and Cu. However, these results highlighted that the use of NaOH in combination with an amphoteric surfactant BW is efficient to simultaneously reduce the concentration of As, Cr, Cu, PCP, and PCDDF from soils with different levels of contamination.

Leachates Treatment by Precipitation and Sludge Production

The primary disadvantage of using chemical processes to remove contaminants from soils is the production of large amounts of alkaline leachates that should be treated to concentrate the contaminants into small amounts of sludge. Table 6 presents the amount of sludge produced during the treatment of alkaline leachates by precipitation–coagulation and the inorganic and organic contaminant contents measured in the sludge.

Depending on the fine soil fraction treated by leaching, the total amount of sludge produced during the treatment of leachates by precipitation–coagulation varied between 1.1 and 3.7% (w/w) of all soil treated. The treatment by precipitation–coagulation of alkaline leachates concentrated the inorganic and organic contaminants, especially PCDDF, in small amounts of sludge, thus reducing the costs related to waste disposal. Indeed, the treatment of leachates by precipitation–coagulation at pH = 7 in the presence of ferric ions allowed the precipitation of metals as $\text{Cr}(\text{OH})_3$, $\text{Cu}(\text{OH})_2$, $\text{Cu}_3(\text{AsO}_4)_2 \cdot 2\text{H}_2\text{O}$ or $\text{FeAs}_2\text{H}_2\text{O}$ or the adsorption of metals onto ferric hydroxides or oxy-hydroxides (Reynier et al. 2015). Moreover, the precipitation of ferric hydroxides or oxy-hydroxides enhances the removal of hydrophobic organic contaminants such

Table 7. Direct and Indirect Costs Related to the Treatment by Attrition (>0.125 mm) and Alkaline Leaching (<0.125 mm) of Different Soils Contaminated by As, Cr, Cu, PCP, and PCDDF

Soil sample	D1	G2	S1	S3	T7:1
Direct operating costs (US\$/t)					T7:2
Chemicals					T7:3
Surfactant (BW)	10.11	9.27	3.37	11.29	T7:4
Sodium hydroxide	22.47	20.60	7.49	25.09	T7:5
Sulfuric acid	11.51	10.55	3.84	12.85	T7:6
Ferric chloride	0.04	0.04	0.01	0.05	T7:7
Labor					T7:8
Operating and maintenance staff	23.03	13.03	23.03	23.03	T7:9
Utilities					T7:10
Electricity	3.03	3.40	3.16	3.51	T7:11
Process water	0.05	0.04	0.02	0.05	T7:12
Fuel	13.51	12.38	4.50	15.09	T7:13
Transport and disposal of soil (C-D) (120 US\$/t)	17.33	106	5.77	127	T7:14
Transport and disposal of hazardous wastes and highly contaminated soils (US\$500/t)	82.26	148	154	90.19	T7:15
Total direct operating cost	213.27	355.50	230.00	331.25	T7:16
Indirect and general costs (US\$/t)					T7:17
Administrative staff	3.47	3.45	3.45	3.45	T7:18
Insurance and taxes	18.19	18.30	13.20	19.41	T7:19
Capital cost					T7:20
Depreciation	67.71	70.14	50.62	74.41	T7:21
Debt service	43.84	44.11	31.83	46.80	T7:22
Total indirect and capital costs	158.04	165.27	122.49	172.29	T7:23
Net cost	371.31	520.77	352.49	503.55	T7:24

as PCP and PCDDF, allowing their adsorption by electrostatic interactions (van der Waals interactions) on the flocs produced.

The contaminant contents measured in the sludge produced during precipitation–coagulation varied between 975 and 2,550 mg As/kg (dry basis), between 930 and 1,500 mg Cr/kg, between 1,690 and 2,820 mg Cu/kg, between 11.1 and 699 mg PCP/kg, and between 23,400 and 128,000 ng TEQ/kg, depending on the initial level of contamination present in the fine soil fraction (<0.125 mm). Owing to the large amounts of both organic and inorganic contaminants measured in sludge, these residues should be properly and safely managed according to the regulations established in the country.

Economic Analysis

Table 7 presents the cost related to the treatment of different soils contaminated by As, Cr, Cu, PCP, and PCDDF by attrition (>0.125 mm soil fraction) and alkaline leaching (<0.125 mm soil fraction) (Scenario 1), whereas Table 8 presents the cost related to the treatment of the coarse fraction by attrition and the landfilling of the fine fraction (Scenario 2). The total cost, expressed in US\$/t, include the direct, indirect, and capital costs estimated for the decontamination of 15,000 t of contaminated soils per year.

The global cost related to the decontamination of different soils contaminated by As, Cr, Cu, PCP, and PCDDF ranged from US\$353 to US\$521/t for Scenario 1 and from US\$235 to US\$443/t for Scenario 2. As expected, the main parameters impacting the decontamination cost are the performance of both the attrition and alkaline leaching processes and the ultimate amounts of heavily contaminated soil and hazardous wastes (metallic sludge) to be appropriately disposed of. Indeed, if the attrition or alkaline leaching processes did not sufficiently reduce both organic and inorganic contaminants, the soil fraction should be

Table 8. Direct and Indirect Costs Related to the Treatment by Attrition (>0.125 mm) and Disposal of the Fine Fractions (<0.125 mm) of Different Soils Contaminated by As, Cr, Cu, PCP, and PCDDF

T8:1	Soil sample	D1	G2	S1	S3
T8:2	Direct operating costs (US\$/t)				
T8:3	Chemicals				
T8:4	Surfactant (BW)	0.00	0.00	0.00	0.00
T8:5	Sodium hydroxide	0.00	0.00	0.00	0.00
T8:6	Sulfuric acid	0.00	0.00	0.00	0.00
T8:7	Ferric chloride	0.00	0.00	0.00	0.00
T8:8	Labor				
T8:9	Operating and maintenance staff	10.2	10.2	10.2	10.2
T8:10	Utilities				
T8:11	Electricity	2.93	3.18	2.93	3.34
T8:12	Process water	0.00	0.00	0.00	0.00
T8:13	Fuel	0.00	0.00	0.00	0.00
T8:14	Transport and disposal of soil (C-D)	0.00	69.93	0.00	106
T8:15	Transport and disposal of hazardous wastes and highly contaminated soils (500 US\$/t)	151	282	156	164
T8:16	Total direct operating costs	179.73	374.17	185.37	292.11
T8:17	Indirect and general costs				
T8:18	Administrative staff	1.53	1.53	1.53	1.53
T8:19	Insurance and taxes	5.51	6.16	5.53	6.03
T8:20	Capital costs				
T8:21	Depreciation	21.13	23.62	21.19	23.11
T8:22	Debt service	13.29	14.86	13.33	14.54
T8:23	Total indirect and capital costs	55.31	68.80	55.68	64.13
T8:24	Net costs	235.04	442.97	241.05	356.24

disposed of in an appropriate secured landfill site, increasing the decontamination cost. The amounts of sludge produced during the attrition and precipitation of leachates also significantly impacted the cost of decontamination, ranging from US\$82 to US\$154/t for Scenario 1 and from US\$151 to US\$282/t for Scenario 2. These results highlighted that the treatment of the fine fraction by leaching allowed a significant reduction in the volume of highly contaminated soil to be appropriately disposed of. However, the chemical cost related to the treatment of the fine fraction by alkaline leachates and the treatment of effluents by precipitation were estimated to be US\$15–50/t, depending on the nature of the soil and the initial level of contamination. These costs revealed the necessity of treating only a small proportion of the contaminated soil and to favor physical decontamination methods such as attrition for the coarse particles to reduce the direct and investment costs.

The indirect and capital costs accounted for between 32 and 43% of the total decontamination cost for Scenario 1 and between 15 and 24% for Scenario 2. The differences observed were primarily a result of the highest investment cost obtained when the fine fraction of the soils was treated by alkaline leaching (approximately US\$10 million) compared to the investment costs of Scenario 2 (approximately US\$4 million).

According to these results, Scenario 2 seemed to be less expensive than Scenario 1 for all soils studied (US\$353–521/t for Scenario 1 versus US\$235–443/t for Scenario 2). A technico-economic evaluation has been made with different treatment plant capacities varying from 48 t per day (15,000 t per year) and 100 t per day (31,500 t per year). The results presented in Table 9 show that the increased treatment plant capacities from 48 to 100 t/day significantly reduced the decontamination costs from US\$353–521/t to US\$299–455/t for Scenario 1 and from US\$233–443/t to US\$214–421/t for Scenario 2. An increase in the treatment capacities of the decontamination plant from 15,000 t/year to

Table 9. Determination of the Direct and Indirect Costs Related to the Decontamination of Different Soils Contaminated by As, Cr, Cu, PCP, and PCDDF Depending on the Treatment Plant Capacity

Scenario	Soil	Total cost (US\$/t)		T9:2
		(15,000 t/year)	(31,500 t/year)	
1	D1	371.31	305.25	T9:3
	G2	520.77	454.84	T9:4
	S1	352.49	299.62	T9:5
	S3	503.55	434.61	T9:6
2	D1	235.04	214.39	T9:7
	G2	442.97	421.16	T9:8
	S1	241.05	220.37	T9:9
	S3	356.24	334.56	T9:10

31,500 t/year allowed a reduction in the total costs ranging from US\$21 to US\$69/t, depending on the soil and the decontamination process. The estimated global cost was much lower than the actual cost (US\$600/t) related to the secure landfilling of these contaminated soils (Reynier 2012). In other words, the entire decontamination process, with or without the treatment of the fine fraction by alkaline leaching, is highly competitive with current disposal options (thermal desorption and landfilling or incineration in a cement kiln) (Reynier et al. 2013).

Conclusion

The inappropriate management and/or storage of PCP- and CCA-treated wood over the last few decades has led to the contamination of several sites across the world. The primary contaminants found in these sites are As, Cr, Cu, PCP, and PCDDF. The present work evaluates the performance and the robustness of a decontamination process able to simultaneously remove inorganic and organic contaminants using an attrition process for the coarse particles and an alkaline leaching process for the fine particles. Satisfactory removal yields were observed for both organic and inorganic contaminants from the four different soils studied (24–42% of As, 0–13% of Cr, 23–46% of Cu, 0–85% of PCP, and 17–64% of PCDDF). The present results also highlighted that the combination of NaOH and BW is highly efficient to simultaneously remove organic and inorganic contaminants from the fine fraction of contaminated soil. This process extracted 87–95% of As, 50–72% of Cr, 73–84% of Cu, 52–100% of PCP, and 27–73 of PCDDF from different contaminated soils after three leaching steps [(NaOH) = 1 M, (BW) = 3% (w/w), $t = 2$ h, PD = 10% (w/w)]. These results highlighted that the entire leaching process is effective for simultaneous removal of inorganic and organic contaminants. However, the nature of the soil and the type and initial level of contaminants present in the soil seemed to influence the performance of both the attrition and alkaline leaching processes. The cost, including direct and indirect costs, was estimated between US\$235 and US\$521 per ton of treated soil, depending on the nature of the soil, the initial level of contamination, and the scenario applied to the fine soil fraction (alkaline leaching or secured disposal).

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