Techno-economic assessment of an hydrometallurgical process to simultaneously remove As, Cr, Cu, PCP and PCDD/F from contaminated soil

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ABSTRACT

Industrial activities lead to the contamination of large amounts of soils polluted by both inorganic and organic compounds, which are difficult to treat due to different chemical properties. The efficiency of a decontamination process developed to simultaneously remove mixed contamination of industrial soils was evaluated at the pilot-scale, as well as operating costs associated to that process to define the best remediation approach. The results showed that the treatment of the coarse fractions (> 0.250 mm) of 40 kg of soil by attrition in countercurrent mode allowed the removal of 17 to 42% of As, 3 to 31% of Cr, 20 to 38% of Cu, and 64 to 75% of polychlorinated dioxins and furans (PCDD/F). Removals of 60% for As, 2.2% for Cr, 23% for Cu, and 74% for PCDD/F were obtained during the treatment of attrition sludge (< 0.250 mm) by alkaline leaching process. However, the results of the techno-economic evaluation, carried out on a fixed plant with an annual treatment capacity of 7,560 tons of soil treated (tst), showed that the estimated overall costs for the attrition process alone [scenario 1] (CAD$ 451/tst) were lower than the costs of the process, which additionally includes an alkaline leaching step to treat attrition sludge [scenario 2] (CAD$ 579/tst). This techno-economic evaluation also showed that the process becomes competitive with current disposal options (thermal desorption and landfilling – CAD$ 600/tst) from a certain treatment capacity, which is around of 3,465 tst/yr for the scenario 1 and 6,930 tst/yr for the scenario 2. On the other hand, the techno-economic evaluations are crucial to selecting feasible decontamination process for a soil remediation project, with considerations of the type of contamination, site characteristics and cost effectiveness.

Keywords: Soil contamination, PCDD/F, Attrition, Leaching, Surfactant, Techno-economic evaluation.

1 INTRODUCTION

The contamination of soils by both metal(loid)s and organic compounds is one of the most important environmental problems as it negatively impacts the economy and threatens public safety (Chang *et al*., 2018; Liu et *al.,* 2018). Industrial activities are the main source of this mixed contamination (Mao *et al.*, 2015). In December of 2010, the number of sites listed in the inventory of contaminated sites « *Système de gestion des terrains contaminés* » in the province of Quebec (Canada) reached 8,334 sites and this number is still growing. Organic pollution was found in 73% of inventoried sites, whereas metal(loid)s and organic contamination were found in 15% of the listed sites and the rest of the sites were contaminated by metal(loid)s only (MDDEFP, 2013). Wood preserving sites are examples of areas that are contaminated by metal(loid)s (e.g. As, Cr, Cu) and organic compounds such as pentachlorophenol (PCP) and polychlorinated dioxins and furans (PCDD/F) (Kumpiene et al., 2016). The coexistence of metal(loid)s and organic compounds at wood preserving sites are the result of the use of preservatives to extend the wood's life-time and protect it from fungi, insects and weathering (PCA, 2009). The most commonly used preservatives are chromated copper crsenate (CCA) and PCP. PCPs, which were frequently used as wood preservatives in the past, contain high concentrations of PCDD/F as impurities (Verbrugge *et al.*, 2018). The leaching of these preservatives from treated wood when exposed to rainwater can cause an increase in the soil contamination of wood preserving and/or storage sites. The rehabilitation of soils contaminated by both metal(loid)s and organic compounds is a difficult challenge due to the radically different chemical properties of these compounds (Reynier *et al.*, 2013).

Nowadays, the only available option for the remediation of industrial soils dealing with mixed contamination includes thermal desorption to destroy organic contaminants followed by the immobilization of inorganic contaminants or landfilling (Metahni *et al*., 2017). Chemical leaching with an appropriate solution is one of the most efficient and rapid soil cleaning techniques to remove contaminants from the soil matrix (Piccolo *et al.*, 2019). Indeed, chemical leaching is a technology based on the mixing of contaminated soil with a water-based solution to solubilize the contaminants initially present in the soil. Numerous researchers have investigated different types of leaching solutions to remove organic and/or inorganic contaminants from soils (Befkadu et *al*., 2018, Tokunaga and Hakuta, 2002). Inorganic acids, particularly sulfuric, nitric and hydrochloric acids, are commonly used to solubilize metal(loid)s from soils or solids (Coudert et al., 2013, Guemiza et al., 2014). Several chelating agents, such as EDTA and citric acid, have also been tested for metal(loid)s removal from soils (Jiang et al., 2017, Qiao et al., 2017). Recently, the use of chemical leaching under alkaline conditions (pH > 7) has been studied at the laboratory scale. More than 90% of PCP was removed from contaminated soils when exposed to a solution of a sodium hydroxide (NaOH) with a pH of less than 12.5 for 20 minutes at a solid/liquid ratio at 1/8 (g/mL) (Xiao et al., 2008). Reynier (2012) also showed that more than 60% of As, 32% of Cr, 77% of Cu and 87% of PCP could be simultaneously removed from contaminated soils (operating conditions: 3 leaching steps of 2 h each, T = 80ºC, pulp density (PD) = 10% - w/w, [NaOH] =0.5 M, and [cocamidopropyl betaine -BW] = 2% w/w). However, this leaching process is expensive due to the application of severe leaching conditions (high concentration of leaching agent, temperature, retention time, etc.) to the entire soil, even though the coarse fractions are usually less contaminated than the finer ones.

The chemical leaching process may be preceeded by a physical separation step to reduce the volume of contaminated soils requiring chemical treatment , significantly decreasing remediation costs. Soil washing is a physical separation based on mineral processing technologies such as flotation or attrition (Fedje and Strömvall, 2019). For example, Guemiza et *al*. (2017 a) have demonstrated the removal of 56% of As, 55% of Cr, 50% of Cu, 67% of PCP and 62% of PCDD/F from the 1-4 mm soil fraction using an attrition process in the presence of BW at a concentration of 2% (w/w). The efficiency of a treatment process, operating costs, investment capital, as well as the total time required to rehabilitate a contaminated site are some of the other factors to be considered when chosing a decontamination process (Khalid *et al.*, 2017).

This article focuses on the techno-economic assessment of a physico-chemical decontamination process applied at pilot scale to treat soil contaminated by As, Cr, Cu, PCP and PCDD/F. Several decontamination scenarios have been simulated to define an efficient, robust and economically viable remediation strategy depending on the initial level of contamination of each size fraction.

2 MATERIAL AND METHODS

2.1 Soil sampling and characterization

For confidentiality reasons, soil samples are only identified as coming from an industrial area and are refered to as S3. The industrial activity generated a spatially heterogeneous soil contamination by As, Cr, Cu, PCP and PCDD/F (Metahni et *al*., 2019). Sampling was conducted on the first 0 to 15 cm depth interval over an area of 16 m^2 (4 m x 4 m). This area was chosen because of its high PCDD/F contamination, based on a previous sampling campaign completed in November of 2014 (Metahni *et al*., 2019). This sampling campaign, revealed initial PCDD/F levels of 6,678 - 11,322 and 12,625 ng dioxin toxicity equivalence (ng TEQ/kg), for exploration holes of 1 m^2 each (1 m x 1 m) located in the area of interest.

For the present study, a volume of soils of 2.4 m^3 was collected and homogenized on wooden plates using a mechanical shovel before being stored in high-density polyethylene (HDPE) containers. Subsequently, 100 kg of soil were then wet-sieved through four different sieves (12 mm, 4 mm, 1 mm and 0.250 mm) using a mechanical Sweco™ to determine the particle size distribution of the soil. Soil samples were then crushed using a Fristh ball mill (Pulverissette model 6) in order to obtain homogenous samples to subsequently determine the inorganic (As, Cr, Cu) and organic contaminant

(PCP and PCDD/F) contents in the different fractions obtained (> 12 mm, 4-12 mm, 1-4 mm, 0.250-1 mm and < 0.250 mm).

2.2 Decontamination process at pilot-scale

The attrition process applied to the coarse soil fractions (> 12 mm, 4-12 mm, 1-4 mm, 0.250-1 mm) is composed of five attrition steps ([BW] = 0 to 2% (w/w), T = 25°C, PD = 40% (w/w), t = 20 min), followed by a rinsing step (PD = 40% (w/w), T = 25°C) (Guemiza *et al.,* 2017a). This process generates an attrition sludge, which is separated from the pulp and treated soil by sieving on different sieves depending on the size of the fraction to be treated and then by flocculation-decantation. The attrition sludge, which is > 0.250 mm, was reprocessed by attrition with the coarse fractions (> 0.250 mm) of the contaminated soil, while the fine attrition sludge (< 0.250 mm) was treated by alkaline leaching. Ten attrition cycles (L1 to L10) were conducted using a counter–current attrition process (CCAP) on 40 kg of the coarse fractions (> 0.250 mm), where 4 kg of material was used for each loop, as shown in Fig. 1. The loops L1 to L4, L5 to L7, L8, and L9 to L10 were conducted on the > 12 mm, 4-12 mm, 1-4 mm and 0.250-1mm size fractions, respectively. The first series of the five attrition steps (L1) was conducted using fresh water. As mentioned by Guemiza *et al.* (2017b), wastewater from the first stage of attrition is treated by flocculation-decantation in order to remove some of the contaminants (e.g. suspended matter). The wastewaters from other attrition steps were recycled into the CCAP to reducethe consumption of freshwater. At the end of each attrition cycle (five attrition steps), the remaining soil was collected and dried at 60°C in an oven, and the concentrations of metals and PCDD/F were measured to evaluate the performance of the CCAP. In this study, post-attrition PCP monitoring was not performed as this soil had a low initial PCP concentration, ranging from 0.30 to 4.10 mg/kg in the coarse fractions (> 0.250 mm). A counter-current alkaline process (CCLP) was the applied to the fine fraction (< 0.250 mm) of the sludge generated by the attrition process. In total five leaching cycles (L1 to L5) were completed throughout the experiment, as shown in Fig. 2. This leaching process consisted of three leaching steps carried out in the presence of a surfactant (PD = 10% (w/w), [BW] = 3% (w/w), [NaOH] = 0.85 M, t = 2 h and T = 80°C) followed by two rinsing steps with clean water (PD = 10% (w/w), T = 80°C and t = 15 min). Following each leaching step, the solid and liquid fractions were separated via centrifugation. The leaching process generated large amounts of alkaline effluents with concentration of As, Cr, Cu, PCP and PCDD/F higher than the regulation for the discharge of effluents in sewers. In this study, leachates produced in the first stage of leaching process (Lix 1) were treated by precipitation followed by adsorption on activated carbon before being discharged into municipal sewers. First, a solution of sulfuric acid (93% H_2SO_4) was used to reduce the pH of the leaching solution from 13.0 –13.5 to 7.5–8.0. Then, 0.04 g/L of activated carbon (AC) (Norit ®C GRAN, Cabot Norit Activated Carbon, Belgium) was added to the solution under agitation for 20 min to improve the adsorption of metal(loid)s and organic contaminants. The wastewaters generated from the other leaching steps were re-circulated into the leaching process. For the fine fraction (< 0.250 mm) of the soil, which represents only 13% of the entire soil, was directly managed as hazardous residual materials (HRM) considering it's very high PCDD/F contamination (30,110 ng TEQ/kg).

2.3 Analytical methods

2.3.1 Determination of metal(loid)s content

The determination of total As, Cr or Cu concentrations before and after soil treatment was performed by ICP-AES (inductively plasma coupled to atomic emission spectroscopy, Vista Ax CCO simultaneous ICP-AES, Varian, Mississauga, ON, Canada) at the Institut National de la Recherche Scientifique (INRS) laboratory.The metal(loid) concentrations were determined after digestion of 0.5 g of dry soil samples in the presence of nitric and hydrochloric acids (HNO_3 and HCl) according to the Method 3030I (APHA, 1999). Each soil sample was digested in triplicate to get an average value of metal content. The accuracy of the results was addressed with the use of standard certificate soil samples (CNS 392-050, PQ-1, lot # 7110C513, CANMET, Canadian Certified Reference Materials Project, Ottawa, ON, Canada) and certified standard solutions (Multi-elements standard, Catalogue No.C00-061-403, SCP Science, Lasalle, QC, Canada).

2.3.2 Organic contaminants analysis

The PCP concentrations were determined at INRS laboratory after a Soxhlet extraction of 20 g of dry soil samples with 300 mL of methylene chloride followed by a liquid/liquid extraction step performed in the presence of sodium hydroxide at 20 g/L. Then, a PCP derivatization step was performed using anhydrous acetate and a solution of potassium carbonate (75%, v/v). After 12 h of agitation, a liquid/liquid extraction step was carried out using methylene chloride. A solution of surrogate standard (PCP- $^{13}C_6$ and 2,4,6-tribromophenol) was added during the Soxhlet extraction step and the first liquid/liquid extraction step to ensure the quality of PCP recoveries. Before analysis, a solution of Phenanthrene-D10 was added was added as internal standard. Finally, the samples were analysed by gas chromatography with mass spectroscopy (GC-MS) (Perkin Elmer, model Clarus 500, column type Rxi-17, 30 m x 0.25 mm x 0.25 µm) according to CEAEQ method (CEAEQ, 2013).

The analysis of PCDD/F concentrations was performed by the accredited Wellington Laboratory, Guelph, ON, Canada, following the CEAEQ method MA. 400-D.F. 1.1 (CEAEQ, 2011).

All contaminant removal yields (RY) were calculated using the following equation (Eq. (1)):

$$Removal\ yield\ (\%) = \left[1 - \frac{final\ contaminant\ concentration}{initial\ contaminant\ concentration}\right] * 100 \quad (1)$$

The initial and final concentrations of contaminants are expressed in mg/kg for As, Cr, Cu and PCP and in ng TEQ/Kg for PCDD/F.

2.3.3 pH and total carbon measurements

The pH of the attrition, leaching and rinsing solutions were measured using a pH-meter (Accumet Model 915, USA), equipped with a double junction Cole-Parmer electrode with an Ag/AgCl reference cell. Before each series of measurements, certified buffer solutions (pH = 2, 4, 7, and 10) were used to calibrate the pH-meter. The total solid contents were measured according to the APHA method 2540D (APHA, 1999). The total carbon (C) and organic carbon concentrtaions were analyzed according to the method CSNH 412.1 using a CHNS Leco analyzer (LECO TruSpec® Micro CHNS 932, Michigan, USA) (Hedges *et al.*, 1984).

2.4 Technico-economic assessment of the treatment process

For this study, a software was modeled to evaluate the direct and indirect costs of the process discussed in the previous section to treat soils contaminated by As, Cr, Cu, PCP and PCDD/F. This model includes more than 260 input variables used to define soil characteristics, processing steps, market parameters and exploitation, capitalization parameters, operating parameters, etc. This model was used to evaluate two different decontamination scenarios. These scenarios were considered under the most similar possible operating conditions, but for two different treatment options. Both treatment scenarios include wet-sieving. Scenario 1 consists of treating only the coarse fractions (> 0.250 mm) of soil S3 by attrition and disposing of the fine fraction (< 0.250 mm) and the attrition sludge (< 0.250 mm) as HRM (Fig. 3a). Scenario 2 consists of treating the coarse fractions (> 0.250 mm) by attrition and the attrition sludge (< 0.250 mm) by alkaline leaching, whereas the fine fraction of the soil (< 0.250 mm) is disposed of as HRM (Fig. 3b).

Prior to establishing the two scenarios to be tested, a preliminary alkaline leaching test on the fine fractions (< 0.250 mm) of the soil S3 was performed applying the same leaching conditions used for the attrition sludge. Three leaching steps were carried out in the presence of a surfactant (PD = 10% (w/w),

[BW] = 3% (w/w), [NaOH] = 0.85 M, t = 2h and T = 80°C) followed by two rinsing steps with clean water (PD = 10%, T = 80°C and t = 15 min). The results showed that despite a removal yield of 37.5% of PCDD/F, alkaline leaching was not sufficient to reduce the levels of PCDD/F (18,815 ng TEQ/Kg) present in the soil to below the threshold (TH 4 = 5,000 ng TEQ/kg for PCDD/F) (Supplementary Table 1). Indeed, the initial content of PCDD/F contamination (30,110 ng TEQ/Kg) in the fine fractions of the soil (< 0.250 mm) is 2.3 times higher than that of the attrition sludge (< 0.250 mm) (12,780 ng TEQ/Kg). Hence, no matter the treatment scenario applied (Scenario 1 or 2), the fine fraction (< 0.250 mm) must be disposed of as HRM.

2.4.1 Process diagram

The first step in the techno-economic evaluation of a treatment system is to draw up a complete process scheme describing all the stages of the treatment chain while identifying all the inputs and outputs. Inputs include the soil to be treated, water, chemical products, etc., while the outputs include the treated soil, final effluents, as well as attrition sludge, precipitation sludge, etc. Acomplete depiction of the decontamination process for soils contaminated with As, Cr, Cu, PCP and PCDD/F is shown in Fig. 3. Scenario 1, presented in Fig. 3a, involves the application of an attrition treatment (physical treatment) to decontaminate the coarse fractions (> 0.250 mm),representing approximately 87% (w/w) of the entire soil weight and the disposal of the fine fraction (< 0.250 mm) and the attrition sludge (< 0.250 mm) as HRM. Scenario 2, presented in Fig. 3b, involves the application of an attrition treatment to decontaminate the coarse fractions (> 0.250 mm) combined with an alkaline leaching treatment (chemical treatment) to treat the fine fraction of attrition sludge (< 0.250 mm). The operating conditions for both the attrition and alkaline leaching processes have been optimized by Guemiza *et al.* (2017a) and Mercier *et al.* (2017) using a surface response method.

2.4.2 Operating conditions and process exploitation

The development of a techno-economic evaluation model for a treatment system requires the definition of a large number of parameters (variables) that can be modified if necessary in order to evaluate their effect on the economic performance of the decontamination process. The variables to be defined first include operating parameters such as the operating period (d/yr), the processing capacity of the plant (tst/d), the number of hours of operation per day (h/d), the operating efficiency factor (%), as well as unit processing income ($/tst).

Basic market parameters must also be identified as variables. The main parameters of this category are: annual inflation rate (%/yr), annual interest rate (%/yr), annual discount rate (%/yr), income tax (% of gross revenue), exchange rate (US$/CAD$), and the Marshall and Swift Equipment Cost Index. An exhaustive techno-economic evaluation must also consider certain parameters related to the initial investment (capitalization parameters) such as: the amortization period (yr), the useful life of equipment (yr) as well as the working capital (% of fixed capital costs). The evaluation of the direct and indirect costs of operation requires the definition of a set of parameters. The most common ones are taken from the actual Canadian market , as well as from literature (Peters and Timmerhaus, 1991; Ulrich et *al*., 1984) and are described in Table 1. Finally, the model should also include a set of specific technical parameters defined for each step of the process, such as hydraulic retention time, solid content, consumption of chemical reagents, sludge dryness, etc.

2.4.3 Mass and volume balance sheets

Once the process scheme has been prepared and the variables have been defined, the next step is to prepare a table containing a mass and volume balance sheet, which is generatedfor a volume (m^3) or a specific mass of treated matrix (tst). The table ends with the calculation of the volumes balance sheet, wet masses and dry weights of all inputs and outputs. The sums of the inputs and outputs are then compared using the simple relationship presented in the (Eq. (2)):

$$(Out\ /\ In)(\%) = 100 * \frac{\Sigma\,(\text{outputs})}{\Sigma\,(\text{inputs})} \qquad (2)$$

A value close to 100% is desired, although a difference of ± 10 to 15% is often acceptable. Fig. 4 shows the mass and volume balance sheets for each step of the S3 soil remediation process, including the alkaline leaching of the attrition sludge (< 0.250 mm).

2.4.4 Cost modeling

Once the mass balance sheet has been established, it is then possible to switch to the dimensioning of the required equipment according to a specific scenario (e.g. for a given processing capacity). For this reason, it is necessary to specify the period of time used for each equipment per hour (TEHU) (min/h), the processing capacity of the plant (m^3/d) (FCAJ), as well as the number of hours of operation per day (FNHO) (h/d). To take into account the operational fluctuations that can be encountered at the industrial scale, a 20% Safety Factor (FASE) is often used while sizing the equipment (Remer et *al*., 1990). The multiplicative factor (FAMU) presented in (Eq. (3)), must be applied to the dimensioning of all equipment:

$$FAMU\ (m^3/\ h)\ = \frac{(\text{FCAJ})\ (m^3/\text{d}) * 1.2\ (\text{FASE}) * 60\ (\text{min/h})}{\text{TEHU}\ \ (\text{min/h}) * \text{FNHO}\ (\text{h/d})} \qquad (3)$$

This multiplicative factor identifies the factor to be applied for each cubic meter of soil matrix processed per hour. The sizing takes into account the mass balance sheet to calculate the capacity of different equipment, which must be established according to the equipment type to be considered. The capacity of the different equipment is calculated using Eq. (4).

$$Equipement\ capacity\ (tst/h), (l/h)\ or\ (m^3) \ = \frac{1(\text{tst/ h}) * 1.2 \text{ x flowin}}{1000 * (\text{kg/ tst})} \tag{4}$$

Equipment capacity is expressed in (tst/h) for solids, in (L/h) for liquids and in (m^3) for tanks and reactors. The $flow_{in}$is determined from the mass balance and is expressed in (kg) for solids and in (L) for liquids. The reactors and reservoirs are multiplied by the hydraulic retention time and the liquid flows are converted using their densities. The cost of purchase (including transport) of the various equipments (CATE) constituting the treatment chain can be estimated using the Eq. (5):

$$CATE \ = X * \ (CAP)^Y * \ (MSECI)(a/o) \tag{5}$$

Where « X » is the constant determined from a power law regression of equipment prices for different capacities (CAP), and the exponent « Y » is a scale factor. The constants « X » and « Y » are taken from the website (www.matches.com). Exponent values « Y » for other types of equipment can be obtained from other documents such as Chauvel (1981) and Remer et *al*. (1990). MSECIa is the Chemical Engineering Plant Cost Index (CEPCI) (567.5 in December 2017) and MSECIo is the original CEPCI value for the year in which equipment costs were evaluated (Chemical, 2016).

The electricity cost is based on the electric consumption of each peice of equipment (kW) multiplied by the number of hours theequipment is in use (h/d), giving the energy consumption per day (kWh/d) for each piece ofequipment. The total energy consumption is obtained by summing the individual consumption of each piece of equipment. This consumption value is then multiplied by the number of days of operation per year (FJOA) and the unit cost of electricity (FCUE), which makes it possible to obtain the annual costs in electricity (COAE) of the plant. Electric and thermal requirements were calculated separately using Eq. (6) and Eq. (7), respectively.

$$Power\ (kW) = a * (CAP)^b \tag{6}$$

$$Q_v\,(J) = m * C_v * \Delta T \tag{7}$$

The energy consumption of some basic equipment in environmental technologies is based on Eq. (6). Where « CAP » (t/h) represents the capacity of the equipment, « *a* » is the constant determined from a power law regression of energy consumption of the equipment for different capacities (CAP), and the exponent « *b* » is a scale factor. Depending on the equipment, the variables « a » and « b » are pulled from the literature, or using supplier's data for specialized equipment.

The heat energy « Q_v (Joules) » presented in Eq. (7) is obtained by multiplying the mass of balance « m (kg) » by the specific heat « C_v (J/kg.K) » and by « ΔT (Kelvin) », which represents a change of temperature.

Once equipment costs are established, the other components of the total investment costs are estimated using multiplying factors called Lang Factors. The sum of direct, indirect, construction management costs and the quota fees are combined to give the total capital costs (TCFC). Therefore, the total investment cost is obtained by adding the amount of working capital to the amount of TCFC.

Table 2 presents the main units of direct and indirect costs used for the estimation of total costs of both scenarios 1 and 2 studied. These direct operating costs include chemical products, labor (CAD$ 25/h), electricity (CAD$ 0.07/kWh), process water (CAD$ 0.5/m^3), fuel (CAD$ 3.5/MBtu), loading of truck with contaminated solids (CAD$ 2.5/tst), and transportation (CAD$ 0.15/tst.km), contaminated soil and sludge disposal, maintenance and repairs, operating supplies, laboratory charges as well as patents and royalties. The indirect costs include administrative expenses, marginal social benefits, general plant overhead, insurance and taxes, marketing and sales as well as research and development.

3 RESULTS AND DISCUSSION

3.1 Soil characteristics

Table 3 shows the initial concentrations of As, Cr, Cu, PCP and PCDD/F measured in each soil fraction (> 12, 4-12, 1-4, 0.250-1 and < 0.250 mm). The particle size distribution of the soil shows that coarse fractions (> 0.250 mm) represent the majority of the entire soil (87%), with levels of As, Cr, Cu, PCP and PCDD/F ranging from 10.5 to 32.7 mg/kg, from 26.2 to 38.8 mg/kg, from 47.9 to 91.1 mg/kg, from 0.30 to 4.10 mg/kg and from 1,450 to 7,741 ng TEQ/kg, respectively. Although the fine fraction (< 0.250 mm) only represents 13% of the soil, its initial PCDD/F concentration is very high (30,110 ng TEQ/kg). These results also showed that the concentration of both inorganic and organic contaminants increases as particle size decreases. These findings are in accordance with those of Huang *et al*. (2014), who reported that fine soil aggregates retain more inorganic contaminants due to their larger surface area and due to the presence of clay minerals, organic matter, and Fe/Mn/Al oxides forming fine-sized aggregates. The results presented in Table 3 show that the soil pH is neutral, with low amounts of organic (0.59%) and inorganic carbon (0.48%). These parameters are of great importance since they strongly influence the fixation and behavior of contaminants in the soil (Charlatchka *et al*., 2000; Subramanian, 2007; Xiao *et al*., 2016). Recent studies have also shown that the nature of the soil and the initial level of contaminants seem to influence the performance of both attrition and alkaline leaching processes (Metahni *et al*., 2017, Guemiza *et al*.,2019). Indeed, Metahni et al. (2017) have evaluated the performance of attrition (without surfactant) and alkaline leaching in the presence of a surfactant (BW) processes for the treatment of four different soils polluted by organic and inorganic contaminants. They highlighted that the attrition process is effective in simultaneously removing inorganic and organic contaminants from the coarse fractions (> 0.125 mm) of the different soils studied, with removal yields varying from 24 to 42% for As, 0 to 13% for Cr, 23–46% of Cu, 0 to 85% for PCP and 17 to 64% for PCDD/F. Removal yields of 87 to 95% of As, 50 to 72% of Cr, 73 to 84% of Cu, 52 to 100% of PCP, and 27 to 73% of PCDDF were obtained after three leaching steps conducted on the fine fraction (<0.125 mm). However, the nature of the soil and the initial level of contaminantion seemed to influence performance of both attrition and alkaline leaching processes. Guemiza et al. (2019) also demonstrated that although the content of organic matter and the initial concentrations of PCP and PCDD/F in the soil to be treated influenced the performance of the leaching process, 96 to 98% of PCP and 57% to 81% of PCDD/F were simultaneously removed from the fine fraction (<0.250 mm) through alkaline leaching.

3.2 Performance of attrition and leaching of sludge attrition for the simultaneous removal of inorganic and organic contaminants

Table 4 shows the mass proportions of the different coarse fractions (> 12, 4-12, 1-4 and 0.250-1 mm) of soil S3, as well as the mass proportion of the fine attrition sludge (< 0.250 mm) (8.2%) generated by the attrition process. The initial concentrations (mean value) of organic (PCDD/F) and inorganic (As, Cr, Cu) contaminants and the removal yields (mean value) obtained following the treatment of coarse fractions (> 0.250 mm) by CCAP and the treatment of attrition sludge (< 0.250 mm) by CCLP are presented in Table 4. In this study, post-attrition PCP monitoring was not performed for the soil as it had a low initial contamination, ranging from 0.30 to 4.10 mg PCP/kg in all coarse fractions (> 0.250 mm).

Ten loops of the CCAP (4 kg of soil for each loop) with effluent recirculation were applied to 40 kg of the coarse fractions (> 0.250 mm) in the presence of the BW surfactant resulted in the removal of 17 to 42% of As, 3.0 to 31% of Cr, 20 to 38% of Cu and 64 to 75% of PCDD/F. Although the coarse fractions, which represent the majority of the entire soil (87%), had low initial metal(loid)s concentrations, the CCAP was efficient enough to reduce them below the criteria to allow an industrial use of the remediated coarse fractions. In comparison, Guemiza *et al.* (2017b) evaluated the performance of CCAP to remove metals, PCP and PCDD/F from the 1-4 mm fraction of contaminated soil. The results showed that the contaminant removal yields obtained during 15 loops of the CCAP varied between 32 and 52% for As, 17 and 37% for Cr, 15 and 37% for Cu and 41% and 50% for PCDD/F. The removal yields of attrition observed in the present study were similar than those obtained by Guemiza *et al.* (2017b) for the removal of metal(loid)s but higher for PCDD/F. The higher removal yields of organic contaminants in the current study can be explained by the higher initial concentration of PCDD/F in the S3 soil.

Fig. 5 presents the final concentrations and the corresponding removal yields of PCDD/F measured in the coarse fractions (> 0.250 mm) of soil S3 following the 10 loops of the CCAP that included the treatment of the wastewater from the first stage of attrition (AT1). The removal yields obtained for the PCDD/F ranged from 62 to 65% with an average value of 64 ± 1% in the > 12 mm soil fraction, ranged from 67 to 70% with an average value of 69 ± 1% in the 4-12mm soil fraction, was equal to 73% in the 1-4 mm soil fraction and ranged from 64 to 76% with an average value of 75 ± 1% in the 0.250-1 mm soil fraction. Under these conditions, PCDD/F is most efficiently removed from coarse fractions (>0.250 mm). Furthermore, the recirculation of the attrition wastewater does not appear to reduce the removal yield of PCDD/F from the different fractions. These results also showed that begining the CCAP with the less contaminated fraction and finishing with the most contaminated fraction reduces the accumulation of contaminants in the leachates during the 10 CCAP loops. Similar inorganic contaminants (As, Cr, Cu) removal evolutions were observed during the CCAP for the coarse fractions (> 0.250 mm) of soil S3 without any loss of efficiency. Inorganic contaminants and PCPs were no monitered in the recirculated waters since the coarse fractions (> 0.250 mm) were initially only very slightly contaminated by As, Cr, Cu and PCP. The leachates generated from the 10 CCAP loops were, however, analyzed before and after treatment by activated carbon. Indeed, adsorption onto activated carbon is one of the most common techniques for the treatment of wastewater contaminated with organic contaminants (Reynier *et al.*,2015). The leachates initially contained 12.04 ng TEQ PCDDF/L and 1.85 ng TEQ PCDDF/L following the treatment with activated carbon, resulting in a removal of 84.6% of PCDD/F.

The attrition process applied to the coarse fractions generated a highly contaminated attrition sludge (< 0.250 mm), but representing only 8.2% of the entire soil treated. To reduce this contamination, attrition sludge were submitted to the alkaline leaching via the CCLP in the presence of a surfactant (BW) to improve the removal of hydrophobic contaminants such as PCDD/F. The average removal yields, calculated after five loops of the CCLP, were 60 ± 3%, 2.2 ± 0.9%, 23 ± 1% and 76 ± 2%, respectively for As, Cr, Cu and PCDD/F. The low removal rates obtained for Cr (2.2%) during alkaline leaching are in accordance with the results obtained by Metahni *et al.*(2017) and Mercier et *al*.(2017) and can be explained by the low availability of chromium present in the soil in mineral form. Despite low Cr and Cu removals, alkaline leaching is efficient in removing the most problematic contaminants, such as As and PCDD/F, from the highly contaminated attrition sludge.

In soil rehabilitation, removal yields are not the most important parameters under considered when determining the efficiency of a decontamination process. Most industrial countries have defined different thresholds (TH) for both organic and inorganic contaminants below which particular uses of these soils (Supplementary Table 1) are authorised. For highly contaminated sites, these thresholds are crucial because they dictate which fractions of the soil can be disposed of directly in a sanitary landfill (contaminant content between TH 3 and TH 4) and which fractions must be disposed of as HRM (contaminant content > TH 4). Indeed, the cost of managing soil fractions exceeding TH 4 is very expensive compared to soil fractions with contaminant contents between TH 3 and TH 4. According to our results, the CCAP applied to the coarse fractions (> 0.250 mm) allowed to treat all fractions greater than 4 mm, representing 50.5% of the entire soil, being below TH 3. This significant decrease in contamination will allow the reuse of these fractions in industrial applications, effectively reducing by half the amount of contaminated soils to be disposed in a sanitary landfill and therefore significantly reducing the rehabilitation costs. However, for 36.5% of coarse fractions (> 0.250 mm), residual concentrations of PCDD/F remain above TH 3, requiring disposal in a sanitary landfill. The CCLP applied to the highly contaminated attrition sludge (< 0.250 mm) decreased the PCDD/F content from x > TH 4 to TH 3 < x < TH 4, which makes it possible to dispose of this treated attrition sludge in a sanitary landfill, reducing costs related to its management.

3.3 Operating conditions and mass balance

According to the basic operating parameters presented in Table 1, both scenarios 1 and 2 take into account a fixed treatment plant, with a processing capacity of 24 tst/d, during an operation period of 350 d/y, running full time (24 h/d). An efficiency factor of 90% was used with a factor of safety on equipment sizing of 20% to be able to adjust operation in case of mechanical breakdowns of equipment. Concerning the market parameters used, they have been defined as follows: an inflation rate of 2%, an annual interest rate of 5% and an annual discount rate of 6%. The capitalization parameters taken into account in this model are a 10-year depreciation period and a 10-year equipment life, as well as a working capital of 15% of fixed capital costs. In both scenarios, it was considered that all soil fractions, as well as final waste contaminated by inorganic or organic contaminants (e.g. fine soil fraction (< 0.250 mm), attrition sludge) will be transported over a 50 km distance, while the transport distance of the mixed waste (organic and inorganic contaminants) to be sent for thermal desorption was fixed at 500 km. Disposal costs vary from CAD$ 75 to 600 per ton, depending on the nature of the soil or waste as well as the nature and the level of contaminants.

The soil remediation process comprises: i) a sieving step, ii) an attrition step to treat coarse fractions (> 0.250 mm) and iii) a leaching step of the attrition sludge (< 0.250 mm – Scenario 2 only) according to the different scenarios. The mass and volume balance sheets of inputs and outputs defined for each step are presented in Fig. 4. The results of the mass and volume balance sheets showed that 870 kg of the dry coarse fractions (> 0.250 mm) and 130 kg of the dry fine fractions (< 0.250 mm) were recovered at the end of the wet-sieving performed at ambient temperature. This pre-treatment step used 1,436 L of fresh water that can be recovered and recirculated without prior treatment (1,329 L) for the attrition process, except for the first attrition step that was treated by flocculation-decantation with 0.07 kg of cationic polymer (CMX 123) in order to remove some of the contaminants and organic matter. Subsequently, these treated waters, as well as all the effluents coming from the other attrition steps were recirculated in the attrition steps. Based on this water consumption/use, approximately 3,497 L of fresh water have been used to treat the coarse soil fractions (> 0.250 mm) using five attrition steps followed by a rinsing step in counter-current mode. At the end of the attrition process, 792 kg of dry treated soil, 88.6 kg of dry attrition sludge (< 0.250 mm) and 3,460 L of effluents were collected. The attrition sludge generated was separated into two fractions by sieving: i) the coarse attrition sludge (> 0.250 mm), which represent approximately 11% of the total soil, was sent to the contaminated soil fraction to be treated by attrition, whereas ii) the fine attrition sludge (< 0.250 mm), which represent 8.2% of the total soil, was identified as HRM in scenario 1 and treated by alkaline leaching in scenario 2. The final step of the process, which consists of treating the fine attrition sludge (< 0.250 mm) by alkaline leaching in the presence of a surfactant, yielded 88.6 kg of dry treated sludge and 914 L of contaminated effluent. Approximately 829 L of these effluents were recycled into the CCLP, while 85 L of leachates from the first leaching step (L1) were subsequently treated by precipitation-decantation followed by an adsorption step on activated carbon to be rejected in the sewers. Table 5 presents the mass and volume balance sheets of all inputs and outputs at each stage of the process obtained using the techno-economic software developped. The results of these reports showed an output/input ratio of 100.0%, 100.4% and 100.2% for the sieving, attrition and alkaline leaching steps, respectively.

3.4 Costs analysis

The management of soil pollution is a major economic challenge worldwide (Khalid *et al.*, 2017). There exist several important factors to consider when selecting an appropriated method of a soil remediation, including the site, type of contamination present, remediation objectives, remediation efficiency, cost-effectiveness, time, and public acceptability (Liu *et al*., 2018). According to Khalid *et al*. (2017), cost is the key factor determining the success and practical application of remediation process under field conditions.

Table 6 presents the costs, including the direct, indirect and estimated capital costs for a plant with a processing capacity of 7,560 tst/yr, related to the removal of As, Cr, Cu, PCP and PCDD/F from the contaminated soil S3 according to the different treatment scenarios 1 and 2. The techno-economic assessment of scenario 1 showed that the total costs related to the treatment of coarse fractions (> 0.250 mm) by attrition and the disposal of the fine fraction (< 0.250 mm) of the soil and the attrition sludge (< 0.250 mm) as HRM were estimated at CAD$ 451/tst. These costs are lower than those estimated for the scenario 2 (CAD$ 579/tst), which consisted of treating the coarse fractions (> 0.250 mm) by attrition and the fine attrition sludge (< 0.250 mm) by alkaline leaching, while disposing of the fine fraction (< 0.250 mm) as HRM. Considering the thermal desorption cost, which is around CAD$ 600/ts, a profit of CAD$ 149/tst and CAN$ 21/tst was estimated for scenarios 1 and 2, respectively, using the hydrometallurgical process .

Percentages of direct and indirect costs are presented in Fig. 6 (a) and (b) for scenarios 1 and 2, respectively. These results showed that the direct operating costs represent the major part (from 61% to 71%) of the net costs related to the treatment of soil contaminated by metal(loid)s and organic compounds. These results also highlight the impact of labor costs, as well as disposal costs associated with mixed waste on the total costs of the process, no matter which scenario is considered. Indeed, labor costs represent 13 to 18% of the process costs while transport and disposal costs of hazardous waste (organic and inorganic) represent 21 to 41% of the operating costs. But before designing a techno-economic model, it is very important to take into account the plant's processing capacity per year, the equipment service life, as well as the amortization period. The model used in the present study was designed for a fixed plant dealing with large quantities of contaminated soils. Fig. 7 shows the results of various simulations that have been performed on different processing capacities ranging from 8 to 56 tst/d. For scenario 1, the results showed that a tonnage equal to or greater than 11 tst/d (3,465 tst/yr) is required for the attrition decontamination process to become competitive with thermal desorption, while for scenario 2, a tonnage equal to or greater than 22 tst/d (6,930 tst/yr) is required to be competitive. These results also showed the positive impact of increasing the treatment capacity of the treatment plant on the total cost of the process for both scenarios. By increasing the treatment capacity of the plant from 24 to 56 tst/d, it is possible to generate a profit of CAD$ 84/tst and CAD$ 143/tst for scenarios 1 and 2, respectively. These results are in accordance with those obtained by Metahni *et al*. (2017), which demonstrated that an increase the treatment capacities of the decontamination plant to treat different soils polluted by As, Cr, Cu, PCP and PCDD/F from 15,000 tst/yr to 31,500 tst/yr resulted in a reduction in the total costs ranging from US$ 21/tst to US$ 69/tst depending on the nature of the contaminated soil and the decontamination process applied. Pasquier *et al.* (2016) also carried out a technical and economic evaluation of a mineral carbonation process developed to treat raw flue gas issued from an industrial plant. They demonstrated that a variation in the plant treatment capacity had a significant impact on the global cost of the process.

The depreciation period and the equipment service life included in the 10-year model also need to be considered. It is necessary to predict a recurrent tonnage during the entire period of operation of the plant. This would render the total costs of the decontamination process economically and comprehensively more attractive than thermal desorption.

4 CONCLUSION

This study focused on a detailed techno-economic evaluation of a decontamination process applied to sites contaminated by both organic (PCP and PCDD/F) and inorganic (As, Cr and Cu) pollutants. For comparison purposes, two treatment options were considered within this study under the most similar conditions possible: one without leaching of attrition sludge and the other one with alkaline leaching. The technical feasibility of the proposed method for the rehabilitation of a site contaminated by both As, Cr, Cu, PCP and PCDD/F has been successfully demonstrated. Indeed, the results showed that for a plant with a processing capacity of 7,560 tst/yr, the attrition of the coarse fractions (> 0.250 mm) alone and the disposal of fine residues (< 0.250 mm) as HRM was much more economic than the process including alkaline leaching of the attrition sludge (< 0.250 mm). The technical and economic evaluations have shown that this decontamination process could be competitive to other soil decontamination methods already available on the market such as thermal desorption for a plant able to treat 3,465 tst/yr for the scenario 1 and 6,930 tst/yr for the scenario 2. This opens the door to the economically feasible application of this process into the market for the rehabilitation of contaminated soil.

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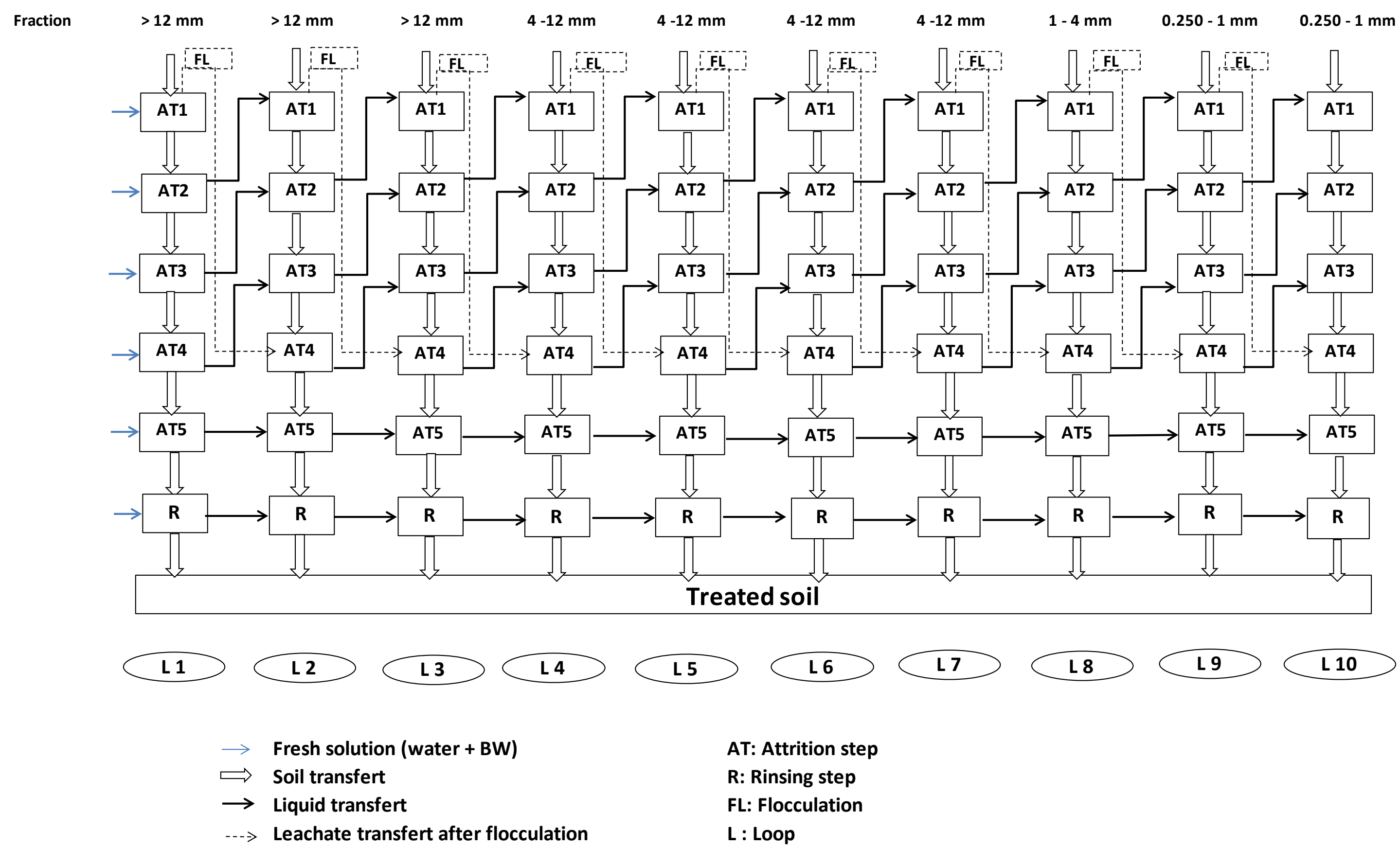
FIGURE LIST

LIST OF ABBREVIATIONS

AC	Activated carbon
As	Arsenic
AT	Attrition step
AT1	Wastewater from the first stage of attrition
BW	Cocamidopropyl betaine
C	Total carbon
CAP	Capacity of the equipment
CATE	Cost of purchase of the various equipment's
CCA	Chromated Copper Arsenate
CCAP	Counter-current attrition process
CCLP	Counter-current alkaline leaching process
CEPCI	Chemical Engineering Plant Cost Index
COAE	Annual costs in electricity of the plant
Cr	Chromium
Cu	Copper
EDTA	Ethylenediaminetetraacetic acid
FAMU	Multiplicative factor
FASE	Safety Factor
FCAJ	Processing capacity of the plant
FCUE	Unit cost of electricity
FJOA	Number of days of operation per year
FL	Flocculation
FNHO	Number of hours of operation per day
GC-MS	Gas chromatography with mass spectroscopy
H_2O_2	Hydrogen peroxide
HCl	Hydrochloric acid
HDPE	High-density polyethylene
HNO3	Nitric acid
HRM	Hazardous residual materials
ICP-AES	Inductively plasma coupled to atomic emission spectroscopy
In	Inputs
INRS	Institut National de la Recherche Scientifique
L	Loop
L1	First leaching step
LIX	Leaching step
NaOH	Sodium hydroxide
Out	Outputs
PCDD/F	Polychlorinated dioxins and furans
PCP	Pentachlorophenol
PD	Pulp density
Q	Heat energy
R	Rinsing step
RY	Removal yield
T	Temperature

TCFC	Total capital costs
TEHU	Period of time used for each equipment per hour
TEQ	Dioxin toxicity equivalence
TH	Threshold
TP	Total precipitation
TPH	Total petroleum hydrocarbon


Fraction
> 12 mm
> 12 mm
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4 -12 mm
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4 -12 mm
1 - 4 mm
0.250 - 1 mm
0.250 - 1 mm
FL
AT1
AT2
AT3
AT4
AT5
R
Treated soil
L 1
L 2
L 3
L 4
L 5
L 6
L 7
L 8
L 9
L 10
Fresh solution (water + BW)
Soil transfert
Liquid transfert
Leachate transfert after flocculation
AT: Attrition step
R: Rinsing step
FL: Flocculation
L : Loop

Fig. 1

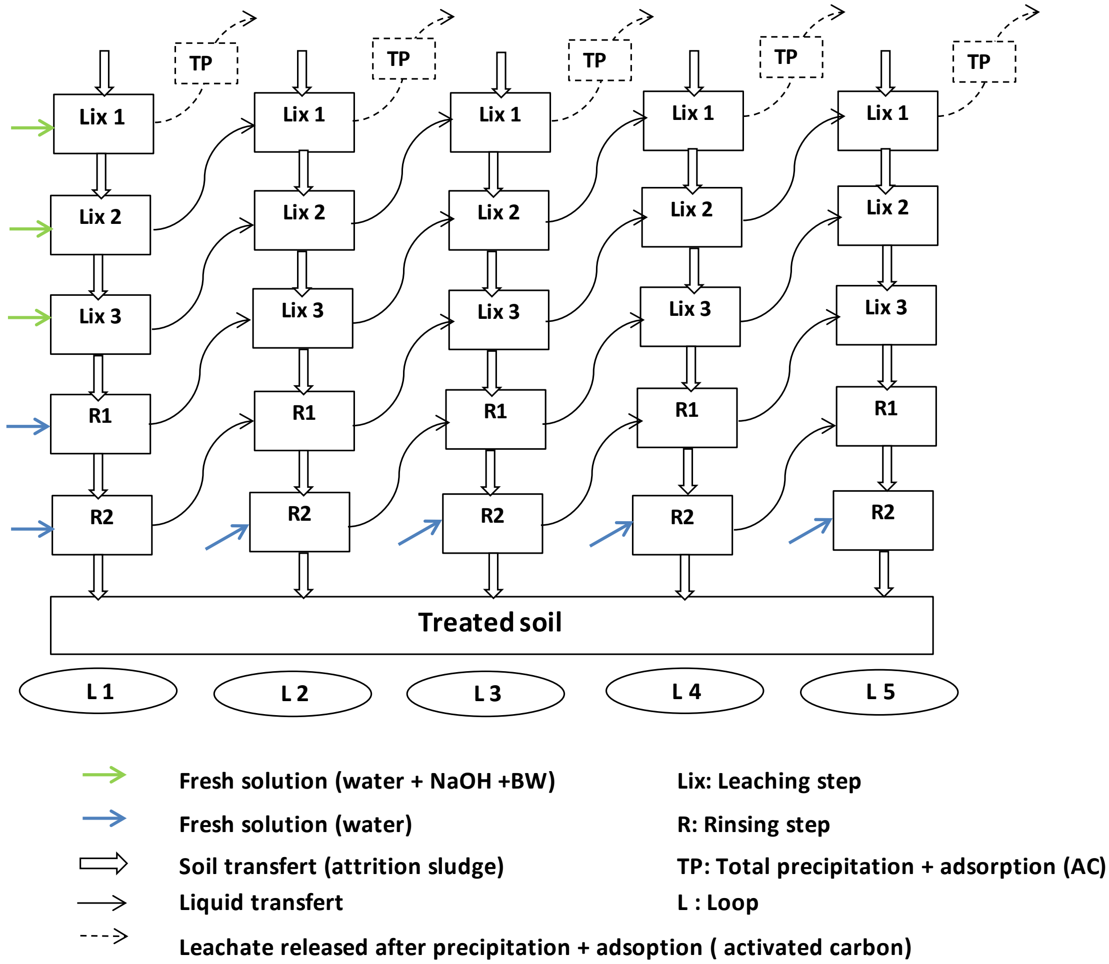
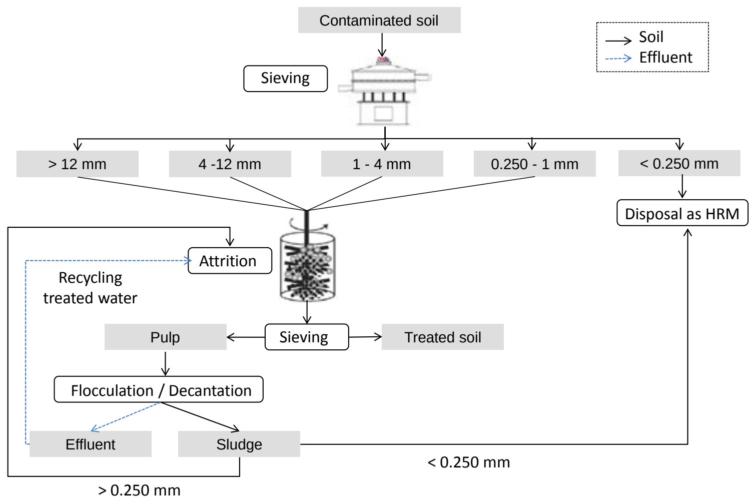
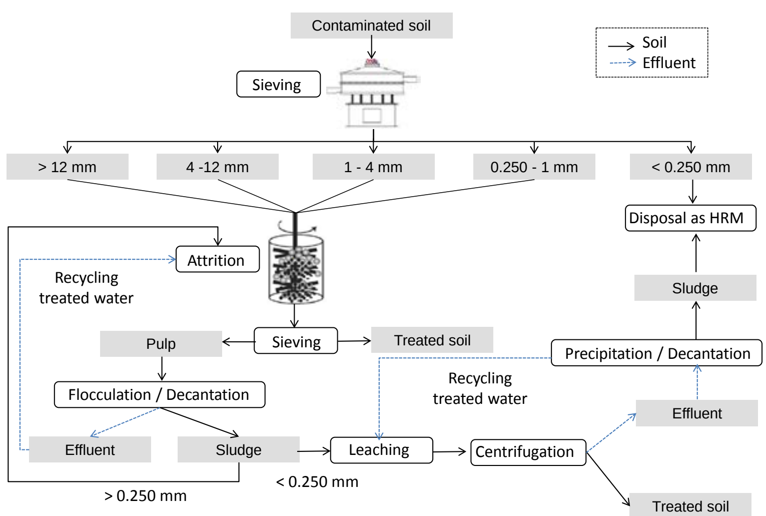

TP
Lix 1
Lix 2
Lix 3
R1
R2
Treated soil
L 1
L 2
L 3
L 4
L 5
Fresh solution (water + NaOH +BW)
Fresh solution (water)
Soil transfert (attrition sludge)
Liquid transfert
Leachate released after precipitation + adsoption (activated carbon)
Lix: Leaching step
R: Rinsing step
TP: Total precipitation + adsorption (AC)
L : Loop

Fig. 2



Contaminated soil
Soil
Effluent
Sieving
> 12 mm
4 -12 mm
1 - 4 mm
0.250 - 1 mm
< 0.250 mm
Disposal as HRM
Attrition
Recycling treated water
Pulp
Sieving
Treated soil
Flocculation / Decantation
Effluent
Sludge
< 0.250 mm
> 0.250 mm

a.



Contaminated soil
Soil
Effluent
Sieving
> 12 mm
4 -12 mm
1 - 4 mm
0.250 - 1 mm
< 0.250 mm
Disposal as HRM
Attrition
Recycling treated water
Sludge
Pulp
Sieving
Treated soil
Precipitation / Decantation
Recycling treated water
Flocculation / Decantation
Effluent
Effluent
Sludge
Leaching
Centrifugation
< 0.250 mm
> 0.250 mm
Treated soil

b.

Fig 3.

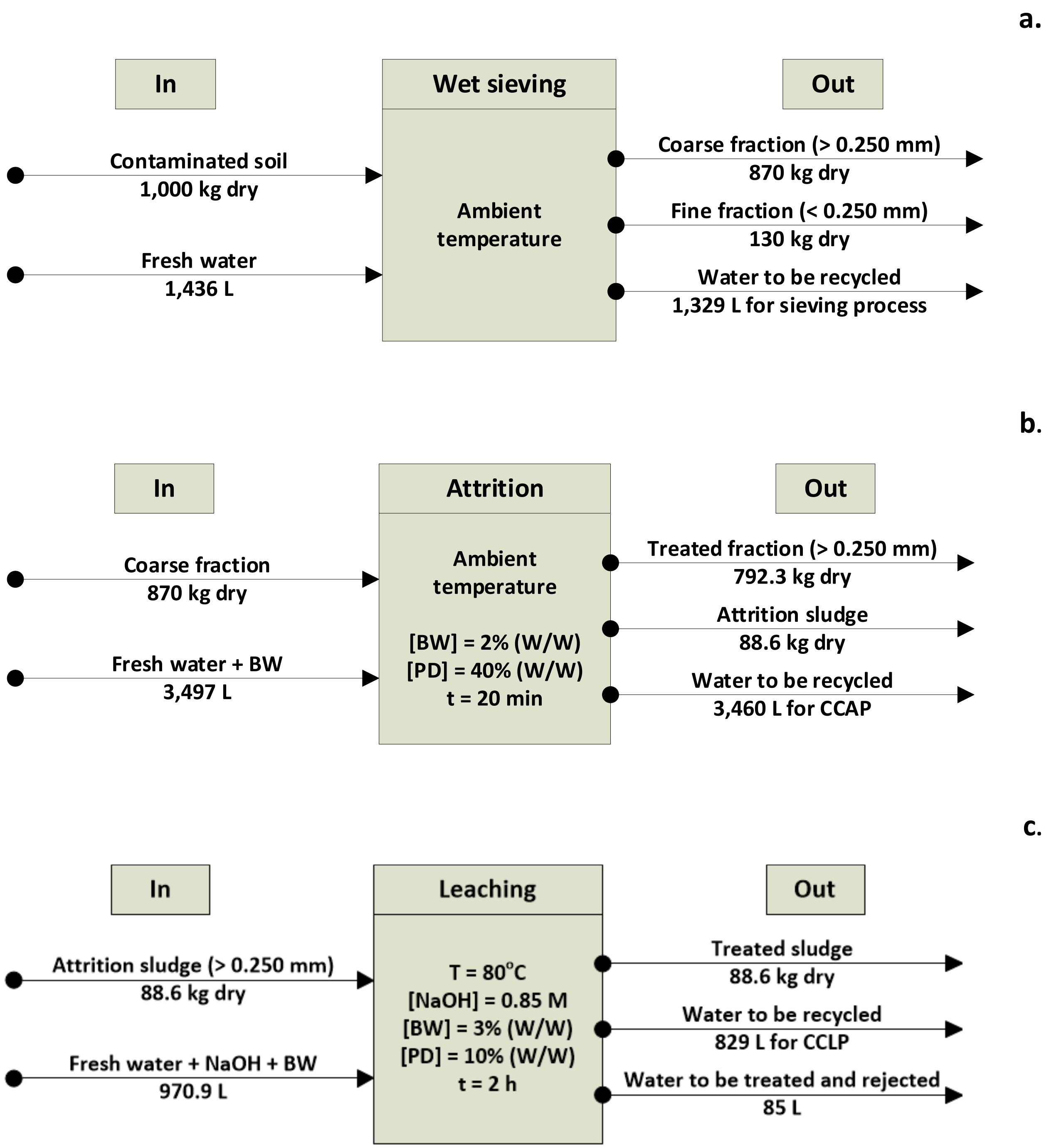
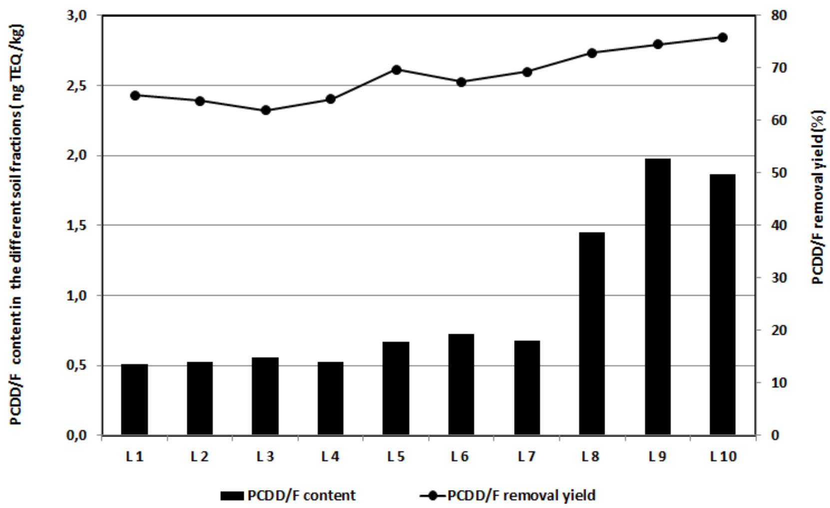
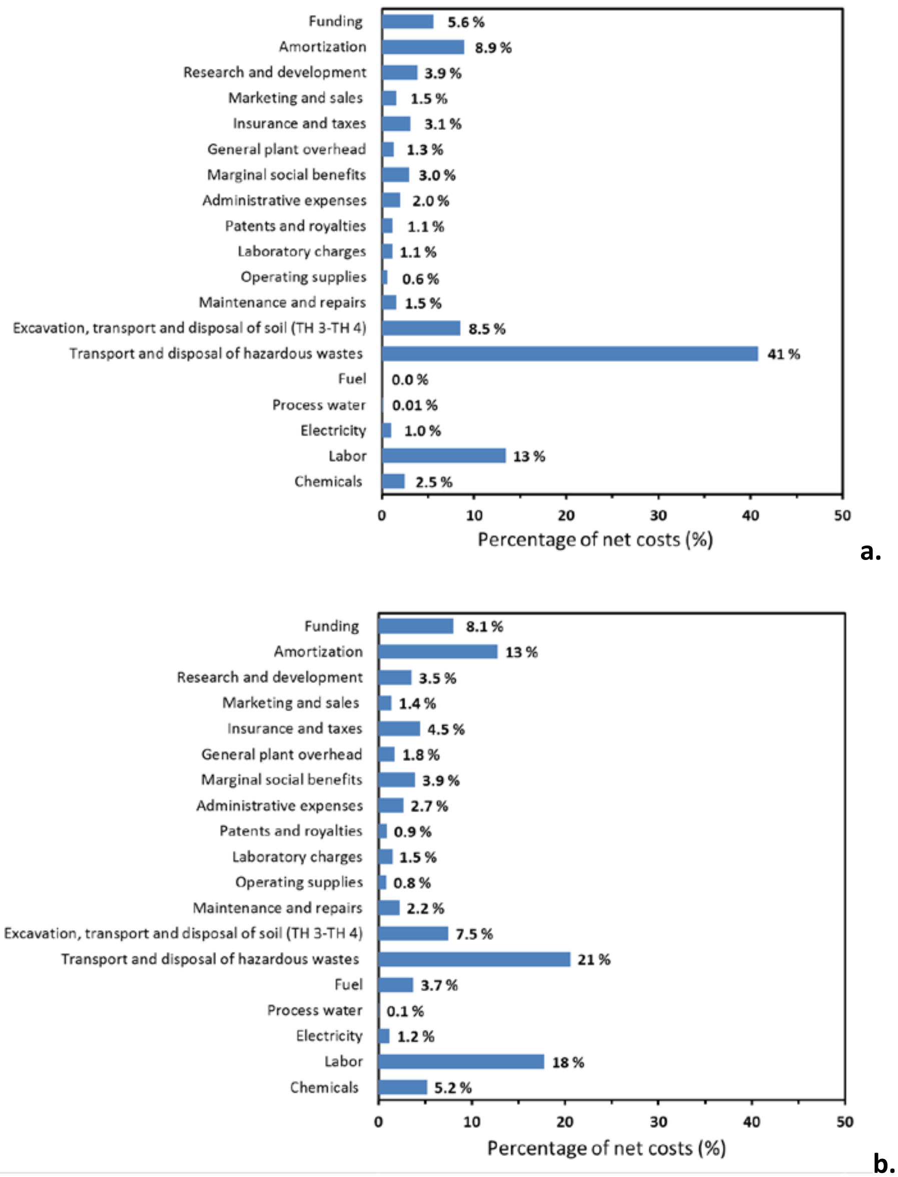

a.
In
Wet sieving
Out
Contaminated soil
1,000 kg dry
Fresh water
1,436 L
Ambient temperature
Coarse fraction (> 0.250 mm)
870 kg dry
Fine fraction (< 0.250 mm)
130 kg dry
Water to be recycled
1,329 L for sieving process
b.
In
Attrition
Out
Coarse fraction
870 kg dry
Fresh water + BW
3,497 L
Ambient temperature
[BW] = 2% (W/W)
[PD] = 40% (W/W)
t = 20 min
Treated fraction (> 0.250 mm)
792.3 kg dry
Attrition sludge
88.6 kg dry
Water to be recycled
3,460 L for CCAP
c.
In
Leaching
Out
Attrition sludge (> 0.250 mm)
88.6 kg dry
Fresh water + NaOH + BW
970.9 L
T = 80°C
[NaOH] = 0.85 M
[BW] = 3% (W/W)
[PD] = 10% (W/W)
t = 2 h
Treated sludge
88.6 kg dry
Water to be recycled
829 L for CCLP
Water to be treated and rejected
85 L

Fig. 4



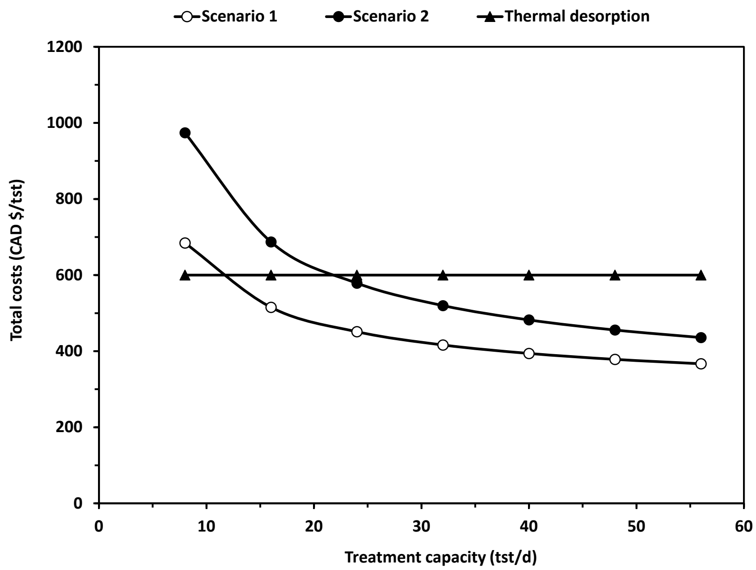
PCDD/F content in the different soil fractions (ng TEQ/kg)
3,0
2,5
2,0
1,5
1,0
0,5
0,0
L 1
L 2
L 3
L 4
L 5
L 6
L 7
L 8
L 9
L 10
80
70
60
50
40
30
20
10
0
PCDD/F removal yield (%)
PCDD/F content
PCDD/F removal yield

Fig. 5



Funding
Amortization
Research and development
Marketing and sales
Insurance and taxes
General plant overhead
Marginal social benefits
Administrative expenses
Patents and royalties
Laboratory charges
Operating supplies
Maintenance and repairs
Excavation, transport and disposal of soil (TH 3-TH 4)
Transport and disposal of hazardous wastes
Fuel
Process water
Electricity
Labor
Chemicals
5.6 %
8.9 %
3.9 %
1.5 %
3.1 %
1.3 %
3.0 %
2.0 %
1.1 %
1.1 %
0.6 %
1.5 %
8.5 %
41 %
0.0 %
0.01 %
1.0 %
13 %
2.5 %
0
10
20
30
40
50
Percentage of net costs (%)
a.
Funding
Amortization
Research and development
Marketing and sales
Insurance and taxes
General plant overhead
Marginal social benefits
Administrative expenses
Patents and royalties
Laboratory charges
Operating supplies
Maintenance and repairs
Excavation, transport and disposal of soil (TH 3-TH 4)
Transport and disposal of hazardous wastes
Fuel
Process water
Electricity
Labor
Chemicals
8.1 %
13 %
3.5 %
1.4 %
4.5 %
1.8 %
3.9 %
2.7 %
0.9 %
1.5 %
0.8 %
2.2 %
7.5 %
21 %
3.7 %
0.1 %
1.2 %
18 %
5.2 %
0
10
20
30
40
50
Percentage of net costs (%)
b.

Fig. 6



Scenario 1: attrition (> 0.125 mm) and disposal of the fine fractions (< 0.125 mm) and attrition sludge (< 0.125 mm); Scenario 2: attrition (> 0.125 mm) with alkaline leaching of attrition sludge (< 0.125 mm) and disposal of the fine fractions (< 0.125 mm).

Fig. 7

Table 1 **Basic operating parameters, market parameters and capitalization parameters of the techno-economic model**

Parameters	Values	Units
Basic operating parameters		
Operating period	350	d/y
Processing capacity of a plant	24	tst/d
Daily operation period	24	h/d
Operating time of the equipment	60	min/h
Operational efficiency factor	90	%
Factor of safety (for equipment)	20	%
Market parameters		
Annual inflation rate	2.0	%/yr
Annual interest rate	5.0	%/yr
Annual discount rate	6.0	%/yr
Income tax	30	% of gross income
Exchange rate	1.30	$US/$CAD
Chemical Engineering Plant Cost Index	567.5	dec-17
Capitalization parameters		
Amortization period	10	yr
Lifetime of equipment	10	yr
Direct (dir.) costs		
Equipment		
Insulation installation equipment	19	%
Instrumentation and control	3	%
Piping and pipeline systems	7	%
Electrical system	8	%
Building process and services	18	%
Landscaping	3	%
Facilities and services	14	%
Land acquisition	0	%
Taxes on equipment	0	%
Indirect (indir.) costs		
Engineering and supervision	32	%
Construction spending	10	%
Construction management fees	9	% cap. (dir. + indir.)
Contingent fees	26	% cap. (dir. + indir.)
Working capital	15	% fixed capital costs

Table 2 Unit direct and indirect costs (CAD $/t) of parameters related to the treatment of soil contaminated by As, Cr, Cu, PCP and PCDD/F

Parameters	Values	Units
Direct operating costs		
Chemicals		
Sulfuric acid	0.08	CAD $/kg
Sodium hydroxide	0.35	CAD $/kg
Cationic polymer	7.00	CAD $/kg
Surfactant (BW)	1.00	CAD $/kg
Activated carbon	1.00	CAD $/kg
Labor		
Unit cost	25	CAD $/h
Supervision	20	% (labor cost)
Utilities		
Unit cost of electricity	0.07	CAD $/kWh
Unit cost of water process	0.50	CAD $/m^3
Unit cost of fuel	3.50	CAD $/M Btu
Solids and concentrates management		
Loading cost	2.5	CAD $/tst
Transportation cost	0.15	CAD $/tst/km
Transport distance		
Regular waste	50	km
Organic hazardous wastes	50	km
Metallic hazardous wastes	50	km
Mixed hazardous wastes	500	km
Soil (TH 3-TH 4)*	25	km
Unit cost of landfill or treatment		
Regular waste	75	CAD $/tst
Organic hazardous wastes	500	CAD $/tst
Metallic hazardous wastes	300	CAD $/tst
Mixed hazardous wastes	600	CAD $/tst
Soil (TH 2-TH 3)	37.5	CAD $/tst
Soil (TH 3-TH 4)	37.5	CAD $/tst
Maintenance and repairs	2.00	% fixed capital costs/yr
Current materials	0.75	% fixed capital costs/yr
Laboratory charges	10	% operating labor
Patents and royalities	5.00	CAD $/tst
Indirect and General costs		
Administrative expenses	15	% operating labor + supervision
Marginal social benefits	22	% operating labor + supervision
General plant overhead	10	% operating labor + supervision
Insurance and taxes	4.0	% fixed capital costs/yr
Marketing and sales	2.0	% total costs
Research and development	5.0	% total costs

**TH: threshold defined by industrialized countries for both organic and inorganic contaminants depending on the expected use of the soil. - Scenario 1: attrition (> 0.125 mm) and disposal of the fine fractions (< 0.125 mm) and attrition sludge (< 0.125 mm); Scenario 2: attrition (> 0.125 mm) with alkaline leaching of attrition sludge (< 0.125 mm) and disposal of the fine fractions (< 0.125 mm).*

Table 3 **Soil characteristics and initial metal(loid)s, PCP and PCDD/F contents in the different soil fractions**

Soil characteristics	Soil proportion (%)	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	PCP (mg/kg)	PCDD/F (ng TEQ/kg)
Particle size						
x > 12 mm	15.9	10.5	28.3	47.9	0.30	1,450
4 < x < 12 mm	34.6	20.4	26.2	52.7	0.50	2,213
1 < x < 4 mm	12.9	32.7	38.8	88.4	1.30	5,345
0.250 < x < 1 mm	23.6	32.0	33.3	91.1	4.10	7,741
x < 0.250 mm	13.0	243	192	312	41.7	30,110
Entire soil	100	52.1	51.4	99.3	6.78	7,427
pH in water at 25ºC	7.11					
Organic carbon (%)	0.59					
Inorganic carbon (%)	0.48					
Dry matter (%)	94.0					

Table 4 **Mass proportions, initial and final concentrations of contaminants as well as removal yields (RY) obtained after treatment of the coarse fraction (> 0.250 mm) of soil by attrition, and after the treatment of attrition sludge (< 0.250 mm) by chemical leaching**

Fraction		**> 12 mm**	**4-12 mm**	**1-4 mm**	**0.250-1 mm**	**Attrition sludge (< 0.250 mm)**
Mass proportion	(%)	15.9	34.6	12.9	23.6	8.2
As	Before (mg/kg)	10.5	20.4	32.7	32.0	123
	After (mg/kg)	8.68 ± 0.89	13.5 ± 4.40	21.1	18.7 ± 0.07	49.2 ± 6
	RY (%)	17	34	35	42	60 ± 3
Cr	Before (mg/kg)	28.3	26.2	38.8	33.3	985
	After (mg/kg)	22.3 ± 3.64	25.5 ± 0.42	27.6	23 ± 0.57	963 ± 69
	RY (%)	13	3	29	31	2.2 ± 0.9
Cu	Before (mg/kg)	47.9	52.7	88.4	91.1	254
	After (mg/kg)	37.1 ± 0.92	42.1 ± 3.25	54.7	58.6 ± 1.90	196 ± 0.07
	RY (%)	23	20	38	36	23 ± 1
PCDD/F	Before (ng TEQ/kg)	1,450	2,213	5,345	7,741	12,780
	After (ng TEQ/kg)	528 ± 18.0	692 ± 27	1,452	1,923 ± 78	3,321 ± 222
	RY (%)	64	69	73	75	76 ± 2

Table 5 **Percentage of the ratio outputs/inputs calculated for the different stages of the process (sieving, attrition and alkaline leaching)**

Mass and volume balance	**Sieving**		**Attrition**		**Leaching**	
	Volume (L)	**Mass (dry kg)**	**Volume (L)**	**Mass (dry kg)**	**Volume (L)**	**Mass (dry kg)**
Inputs	541	1,000	476	877	2,915	2,048
Outputs	541	1,000	476	881	2,915	2,052
Out/In (%)	100.0	100.0	100.0	100.4	100.0	100.2

Table 6 **Nets costs (CAD$/t) related to the treatment of soil contaminated by As, Cr, Cu, PCP and PCDD/F**

Soil sample	Scenario 1	Scenario 2	Units
Direct operating costs			
Chemicals			
Sulfuric acid	0.00	1.98	CAD$/kg
Sodium hydroxide	0.00	9.70	CAD$/kg
Cationic polymer (CMX123)	0.50	0.50	CAD$/kg
Surfactant (BW)	10.81	13.3	CAD$/kg
Activated carbon	0.00	4.60	CAD$/kg
Labor			
Operating labor	50.50	85.62	CAD$/h
Operating Supervision	10.10	17.12	% (labor cost)
Utilities			
Electricity	4.43	6.9	CAD$/kWh
Process water	0.05	0.44	CAD$/m3
Fuel	0.00	21.58	CAD$/M Btu
Transport of mixed hazardous wastes (Récupère-sol)	13.52	13.61	CAD$/tst/km
Excavation and transport of soil (TH 3- TH 4) ($120/t)	5.50	6.21	CAD$/tst/km
Disposal of mixed hazardous wastes	170.77	105.39	CAD$/tst
Disposal of soil (TH 3- TH 4)	33.01	37.23	CAD$/tst
Maintenance and repairs	6.99	12.89	% fixed capital costs/y
Operating supplies	2.62	4.83	% fixed capital costs/y
Laboratory charges	5.05	8.56	% operating labor
Patents and royalties	5.00	5.00	CAD$/tst
Total direct operating costs	**318.86**	**355.47**	**CAD$/tst**
Indirect and General costs			
Administrative expenses	9.09	15.41	% operating labor + supervision
Marginal social benefits	13.33	22.60	% operating labor + supervision
General plant overhead	6.06	10.27	% operating labor + supervision
Insurance and taxes	13.98	25.78	% fixed capital costs/y
Marketing and sales	6.96	8.14	% total costs
Research and development	17.40	20.35	% total costs
Amortization	40.19	74.12	CAD$/tst
Funding	25.28	46.62	CAD$/tst
Total indirect and capital costs	**132.29**	**223.30**	**CAD$/tst**
Net costs	**451.15**	**578.76**	**CAD$/tst**
Thermal desorption cost	**600.00**	**600.00**	**CAD$/tst**
Profit	**148.85**	**21.24**	**CAD$/tst**

Scenario 1 (attrition (> 0.125 mm) and disposal of the fine fractions (< 0.125 mm) and attrition sludge (< 0.125 mm); Scenario 2 (attrition (> 0.125 mm) with alkaline leaching of attrition sludge (< 0.125 mm) and disposal of the fine fractions (< 0.125 mm).

Supplementary Table 1 Thresholds (TH) defined for decision making regarding contaminated soil fractions

Contaminants	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	PCP (mg/kg)	PCDD/F (ng TEQ/kg)
TH 1	6	85	40	0.1	-
TH 2	30	250	100	0.5	15
TH 3	50	800	500	5	750
TH 4	250	4,000	2,500	74	5,000