1	(Water, Air & Soil Pollution)
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3	Influence of soil parameters on the efficiency of the attrition process to remove metals,
4	PCP, dioxins and furans from contaminated soils
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35	0.441 2017
30	October 2017

1 Abstract

2 The objective of this study was to evaluate the influence of the soil parameters (particle size, initial contamination 3 level, etc.) on the performances of an attrition process to remove As, Cr, Cu, pentachlorophenol (PCP) and dioxins 4 and furans (PCDD/F). Five different contaminated soils were wet-sieved to isolate five soil fractions (< 0.250, 5 0.250-1, 1-4, 4-12 and > 12 mm). Five attrition steps of 20 min each, carried out in the presence of a biodegradable surfactant ([BW] = 2%, w.w⁻¹) at room temperature with a pulp density fixed at 40% (w.w⁻¹), were applied to the 6 7 coarse soil fractions (> 0.250 mm) of different soils. The results showed good performances of the attrition process 8 to simultaneously remove PCP and PCDD/F from contaminated soil fractions initially containing between 1.1 and 13 mg of PCP.kg⁻¹ (dry basis) and between 1,795 and 5,720 ng TEQ of PCDD/F.kg⁻¹. It appeared that the amounts of 9 contaminants removed were significantly correlated (*p*-value < 0.05, $R^2 = 0.96$) with the initial amounts of PCP and 10 11 PCDD/F, regardless of the particle size of the soils studied. The nature of the soil (granulometric distribution, pH, 12 total organic carbon (TOC) (organic matter) and diverse industrial origin) slightly and negatively influenced the efficiency of organic contaminants removals using attrition. However, the attrition treatment allowed an efficient 13 removal of both PCP and PCDD/F from the coarse fraction of contaminated soil; despite the nature of the soil. 14

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16 **Keywords**: Soil remediation; Pentachlorophenol; Dioxins and furans; Attrition process; Initial contaminant content;

17 Particle size

1 **1 Introduction**

2 The contamination of soils by inorganic (As, Cd, Cr, Cu, Ni, Pb, Se, Zn, etc.) and organic compounds (polycyclic 3 aromatic hydrocarbons (PAHs), pentachlorophenol (PCP), dioxins and furans (PCDD/F), etc.) is strongly linked to 4 the industrial activities (Coronas et al. 2016; Han et al. 2002). In the United States, among the 1,585 sites 5 enumerated on the priority list, approximately 313 sites have a serious problem of contamination by PCP (ATSDR 6 2001). The pentachlorophenol is listed among the 126 priority pollutants by the Environmental Protection Agency of 7 the United States (US EPA) and the European Union (UE 2000; US Congress 1995; US EPA 2003). The PCDD/F, 8 which are impurities and by-products related to the degradation of PCP, represent a family of 210 compounds 9 including 75 congeners of polychlorinated dibenzo-p-dioxin (PCDD) and 135 congeners of polychlorinated 10 dibenzofurans (PCDF). Among these 210 compounds, 17 congeners, substituted in position 2, 3, 7, and 8, are known 11 as toxic, non-polar and highly hydrophobic compounds. These compounds are resistant to most of biological, 12 chemical and physical activities. Indeed, due to their stability and their high hydrophobicity, PCDD/F and PCP easily accumulate and persist in the living organisms and in the environment (Augustijin-Becker et al. 1994; 13 14 Government of Canada 1990; INSERM 2000; Tissier et al. 2005). Both PCP and PCDD/F are known to be toxic for 15 humans and fauna and to be potentially carcinogenic compounds (UK Environment Agency 2009; US EPA 2003; 16 World Health Organization 2003). Their adsorption on soil depends on their hydrophobic interactions with organic 17 matter (Banerji et al. 1993; INSERM 2000). Trivalent chromium is known to be an essential element in protein, lipid 18 and glucidic metabolism of mammals (Pantsar-Kallio et al. 2001). Deficiencies of Cr (III) induce among other 19 effects an increase in circulating insulin concentrations, a decrease of sperm count and fertility, and shortening life 20 expectancy (Pichard et al. 2005). Global and national organizations have identified some As compounds as 21 hazardous substances to human health (ATSDR 2007). For many years, Cu is known to be essential for many 22 metabolic processes due to its presence in proteins and enzymes. However, in excessive amounts, Cu can cause 23 damages to the liver and kidneys (USEPA 2006).

There are numerous technologies developed for the treatment of soils contaminated by a combination of metals and organic compounds. Among these methods, physical separation methods are usually chosen because of their low cost and simplicity (Mercier et al. 2001). Attrition was found to be an efficient technique (INRS-Georessources & CRM 1997). This technique can be used as a physical decontamination process alone (Bayley et al. 2005; Bisone et

al. 2013; Guemiza et al. 2016; Metahni et al. 2016; Stražišar and Sešelj 1999) as well as a step prior to other 1 2 decontamination processes such as gravity concentration and froth flotation (Bisone et al. 2012; Jobin et al. 2015, 2016; Veetil et al. 2014; Williford et al. 1999). Attrition scrubbing allows friction between contaminated particles; 3 enhancing the removal of the thick films around soil particles and/or the detachment of fine particles usually 4 5 contaminated from the surface of the coarse particles (uncontaminated). Therefore, this process allows the release of 6 contaminants from the coarse particles and favors the concentration of the contaminants in a smaller fraction of fine 7 particles (Bayley et al. 2005; Dermont et al. 2008; Marino et al. 1997; Petavy et al. 2009). According to several 8 studies, some parameters such as the pulp density, the temperature, the particle size, the nature and level of 9 contamination influence the efficiency of attrition scrubbing.

10 **1.1 Pulp density**

11 Bayley et al. (2005) showed that the efficiency of an attrition scrubber on the removal of both organic (Bees Wax 12 and mineral oil Catenex S341) and inorganic compounds is directly proportional to the increase of the pulp density 13 from 20% to 50%. This relationship may be due to the higher probability of collision between soil particles when the 14 solid/liquid ratio is higher. Indeed, the collision of particles enhances the removal of contaminants from the particle 15 surfaces. A study carried out by Petavy et al. (2009) showed that 70% dryness rate was the best rate for the best 16 treatment of metal contaminated sediments by an attrition process at laboratory scale. However, Feng et al. (2001) 17 showed that the efficiency of an attrition process to remove diesel oil from soil contaminated increased when the 18 solid concentration was increased from 70 to 80% and decreased when increasing the solid concentration of the 19 slurry to 85%. This decrease of performances may be due to the fact that the slurry becomes too dense, which 20 decreases the inter-particle motion and the scrubbing action. Recently, Guemiza et al. (2016) used an experimental design methodology to evaluate the effect of pulp density in the range of 20 to 40% (w.w⁻¹) on the removal of PCP 21 22 and PCDD/F from the 1-4 mm fraction of a contaminated soil using attrition. Their results showed that a high pulp 23 density is more efficient for the removal of both contaminants. Indeed, when the pulp density was fixed at 40% 24 (w.w⁻¹), up to 67% of PCP and 62% of PCDD/F were removed from the 1-4 mm soil fraction.

25 **1.2 Temperature and stirring rate**

According to Bayley et al. (2005), the increase of the temperature led to an increase of the removal of organic contaminant (mineral oil) from soils during attrition process (PD = 20%, 1,200 rpm, T = 20-55°C). A study carried 1 out by Guemiza et al. (2016) indicated that increasing the temperature of the solution of attrition between 25 and 2 75°C led to a significant increase of the removal of PCDD/F from the 1-4 mm soil fraction. In contrast, the temperature had no significant effect on the removal of PCP from contaminated soil during the attrition process. 3 These results are not comparable with those obtained by Tse and Lo (2002) that showed that the increase of the 4 5 temperature allows an increase of the solubility of PCP. Some researchers presumed that the increase of the 6 temperature leads to a decrease of the potential energy that keeps the contaminant bonded to the surface of the soil 7 particles, increasing the removal of contaminants from the soil (Bayley et al. 2005; Thorvaldsen and Wakefield, 8 1999).

9 Another parameter that could influence the performance of an attrition process is the stirring speed. A study carried 10 out at room temperature showed that three attrition steps of 20 min each performed at 1,700 rpm with a pulp density fixed at 40% (w/w) allowed the removal of 24 - 42% of As, 0 - 13% of Cr, 23 - 46% of Cu, 0 - 85% of PCP and 17 -11 12 64% of PCDD/F from the coarse particles (> 0.125 mm) of contaminated soils (Metahni et al. 2016). The attrition 13 tests carried out by Bayley et al. (2005) with an aqueous suspension of contaminated soil showed that the speed of the impeller had a significant effect on the removal of contaminants. Their results suggested that an impeller speed 14 15 between 1,200 and 1,800 rpm was recommended to product the energy required to the scrubbing (Bayley et al. 16 2005). According to these authors increasing the impeller speed and therefore, the power input to the soil solution 17 enhanced the performance of contaminant removals from contaminated solid matrix.

18 **1.3** Surfactant concentration

19 In order to enhance the extraction of organic contaminants such as PAHs, PCP and PCDD/F from contaminated 20 soils, several studies suggested the addition of a surfactant to the attrition process (Olea et al. 2003; Paterson et al. 21 1999; Veetil et al. 2014). Indeed, due to their amphoteric proprieties, these compounds allowed the formation of 22 micelles in solution, enhancing the removal of highly hydrophobic compounds. Bisone et al. (2013) removed more 23 than 90% of PAHs from soils using attrition in the presence of a surfactant ([Cocamidopropyl Hydroxysultaine, $CAS = 0.2 \text{ g.L}^{-1}$. Metahni et al. (2016) found that three attrition steps (PD = 40%, T = 25°C, 1-4 mm soil fraction) 24 allowed the removal of 60% and 97% of PCP with and without the addition of surfactant (cocamidopropylbetaine, 25 BW) at 1% (w.w⁻¹), respectively. Recently, Guemiza et al. (2016) showed that the concentration of the surfactant 26 BW $(0 - 3\%, w.w^{-1})$ was the main parameter influencing the removals of both PCP and PCDD/F from the 1-4 mm 27

soil fraction by attrition (PD = 40%, T = 25°C, t = 20 min, 5 attrition steps), using an experimental design
 methodology.

3 1.4 Particle size

4 The particle size has an important role in the distribution of both organic and inorganic contaminants in soils. For 5 example, significant amounts of contaminants can be retained by clays due to their large specific surface areas (up to 6 800 m²/g), as well as, their high cation exchange capacity (CEC). A study carried out by Stražišar and Sešelj (1999) 7 showed that it is easier to remove inorganic contaminant (Pb) from coarse fractions than from fine fractions when 8 using attrition scrubbing. Furthermore, the coarse fractions are generally less contaminated (Stražišar and Sešelj, 9 1999). Feng et al. (2001) observed that the effectiveness of attrition (PD = 80%, T = 25°C, t = 30 min, 600 rpm) to 10 remove organic contaminant (Diesel) from soil increased with the increase of the size particles ranging from 0.1 to 11 0.5 mm. Recently, Jobin et al. (2015) showed that attrition did not significantly reduce the concentrations of 12 inorganic contaminants from the 0.250-1 mm soil fraction of a soil contaminated by incineration residues, whereas 13 the concentrations of arsenic, copper and zinc were significantly reduced after treatment by attrition of the 1-2 mm 14 and 2-4 mm soil fractions. According to these authors, the effect of attrition, for metals, was positively correlated with the size of soil particles (Jobin et al. 2015). All of these studies showed that soils having a low porosity such as 15 16 sands will be better suited to attrition scrubbing than soils with a high porosity.

17 1.5 Type of soil

18 Several parameters influencing the fixation and the behavior of contaminants in soil including its moisture content 19 and the type of contaminants present in the soil, can also impact the performance of an attrition process (Darling et 20 al. 2003; Khodadoust et al. 2005; Ma et al. 2007). Similarly, the pH and the reducing conditions of the soil have a 21 direct influence on the mobility and the availability of contaminants present in soils (Charlatchka et al. 2000; Lee et 22 al. 1998; Subramanian 2007). In addition, the presence of organic matter (humic and fulvic acids), metallic oxides 23 (iron, aluminum or manganese oxides) can immobilize the contaminants into the soil, especially inorganic contaminants (Banerji et al. 1993). According to several authors, As, Cr, Cu, PCP, and PCDD/F are strongly fixed to 24 25 the organic matter or/and to metal oxides present in the soil, indicating that the efficiency of the decontamination 26 process might be influenced by the nature of the soil (amount of organic matter or metal oxides) (Banerji et al. 1993; 27 Revnier et al. 2013; UK Environment Agency 2009). According to the study carried out Metahni et al. (2016), it appeared that the performance of an attrition process to remove PCDD/F from the coarse soil fractions (> 0.125 mm)
was influenced by the nature of the soil (granulometric distribution, pH, total organic carbon (TOC) (organic
matter), total inorganic carbon (TIC) and diverse industrial origin), the type and the initial level of contaminants
present in soils.

5 The objective of this study was to evaluate the influence of several parameters including the type and the level of 6 contamination as well as the nature of the soil (granulometric distribution, TOC (organic matter), TIC and diverse 7 industrial origin) on the performance of an attrition process to remove As, Cr, Cu, PCP and PCDD/F from 8 contaminated soils.

9 2 Material and Methods

10 2.1 Soil sampling and characterization

11 Five soil samples (S1, S2, S3, S4 and S5) contaminated by As, Cr, Cu, PCP and PCDD/F were collected from the 12 same area. These soils were chosen for their different levels of contaminants (low, moderate and high), especially 13 for PCDD/F to evaluate the influence of the level of contamination on the performance of the attrition process developed (Guemiza et al. 2016). Other contaminated soils (F1, F2 and F3) emerging from different industrial areas 14 were collected to determine the influence of the nature of the soil (granulometric distribution, pH, TOC, TIC and 15 16 diverse industrial origin) on the removal of both organic and inorganic contaminants from soils using an attrition 17 process. All these soils were sampled at a depth between 0 and 15 cm on different industrial sites and preserved in High Density Polyethylene (HDPE) containers. Soils were then wet-sieved using a mechanical 76-cm diameter 18 SwecoTM to isolate five soil fractions (< 0.250 mm, 0.250-1 mm, 1-4 mm, 4-12 mm and >12 mm). Each soil fraction 19 20 was collected separately and dried at 60°C. Then, the particle size distribution was analyzed for each soil (S1, S2, 21 S3, S4, S5, F1, F2 and F3). Initial concentrations of As, Cr, Cu, PCP and PCDD/F were determined in all the 22 fractions (> 12 mm; 4-12 mm; 1-4 mm and 0.250-1 mm) of soils S1, S2, S3, S4 and S5 sampled on the same site (to evaluate the influence of the initial concentration and the particle size on the performance of the attrition process 23 developed) and only in the 1-4 mm fraction of the soils F1, F2, and F3 (to evaluate the influence of the nature of the 24 25 soil on the performance of the attrition process developed).

1 2.2 Attrition decontamination process

All attrition experiments were performed at ambient temperature ($T = 25^{\circ}C$) with a pulp density fixed at 40% (w/w), 2 and a concentration of amphoteric surfactant fixed at 2% (Cocamidopropylbetaine: BW, w.w⁻¹) (Guemiza et al. 3 4 2016). Attrition experiments were performed using a stainless reactor with a capacity of 10 L equipped with internal 5 baffles in triplicate. For each experiments, 2 kg of coarse soil fraction (>12 mm, 4-12 mm, 1-4 mm and 0.250-1 mm) 6 were mixed with tap water during 20 min using a mechanical stirrer (Light EV1 P25, AXFLOW, New York, NY, 7 USA) and a stainless steel axial propeller (6 cm diameter). Agitation was set at 1,700 rotations per minute (rpm). 8 After each attrition step, the solid/liquid separation was carried out by sieving onto different sieves (6 mm for the > 9 12 mm soil fraction, 2 mm for the 4-12 mm soil fraction, 0.5 mm for the 1-4 mm soil fraction and 0.250 mm for the 10 0.250-1 mm soil fraction). The attrited soil (coarser fraction retained by the sieve) was then washed with 4 L of tap water before to be reintroduced to the attrition process until a total of 5 attrition steps were achieved. At the end of 11 12 the attrition process composed of 5 attrition steps, the attrited soil was placed in a 20-L High-Density Polyethylene 13 (HDPE) container in order to be washed with 4 L of tap water using a Karcher (Karcher, electric sprayer, 140 kg/cm², Quebec, QC, Canada) before to be sieved using the appropriate sieve. Then, 2 L of tap water were sprayed 14 15 on the attrited soil fraction in order to remove most of the foam. Figure 1 presents the detailed flow sheet of the 16 attrition process applied to the coarse soil fractions (> 0.250 mm). After the solid/liquid separation, the attrited soil 17 fractions were collected and dried at 60°C in an oven and the residual concentrations of PCP, PCDD/F, and metals present in the dry treated soil fractions were then measured. 18

19 2.3 Influence of soil parameters on the efficiency of the attrition process

20 2.3.1 Influence of initial contaminant contents

Five lots of contaminated soils (S1, S2, S3, S4, and S5), having different initial concentrations of contaminants, were sampled in five different locations in the same contaminated site to study the robustness of the attrition process and especially the effect of the initial contaminant contents on the performances of As, Cr, Cu, PCP and PCDD/F removals from contaminated soils. The particle size distribution was performed using four different sieves (12, 4, 1 and 0.250 mm). Attrition experiments were performed in triplicate on the 1-4 mm soil fraction of S1, S2, S3, S4 and S5 using operational conditions described in Section 2.2 and defined by Guemiza et al. (2016).

1 2.3.2 Influence of particle size

To study the effect of the soil particles size on the efficiency of the optimized attrition process, several attrition
assays were performed on all the coarse fractions (0.250-1 mm, 1-4 mm, 4-12 mm et >12 mm) of the soils S1, S2,
S3, S4 and S5. All attrition assays were conducted in triplicate using operational conditions described in Section 2.2.

5 2.3.3 Effect of the nature of soils

The objective of this part was to evaluate the influence of the nature of the soil on the efficiency of the optimized attrition process. Therefore, soils from various locations (F1, F2 and F3) contaminated by As, Cr, Cu, PCP and PCDD/F, sampled in three different industrial areas, were used for these experiments. The particle size distribution of each soil was performed using four sieves (12, 4, 1 and 0.250 mm). Attrition assays were conducted in triplicate on the 1-4 mm fraction of these soils using operational conditions described in Section 2.2. A characterization of the 1-4 mm fraction of each soil sample was performed and the following parameters were followed: total organic carbon, total inorganic carbon, pH and the granulometric distribution.

13 2.4 Analytical methods

The pH level was determined according to the method described by the Quebec Expertise Center for Environmental Analysis (CEAEQ) (MA. 100 - S.T 1.0 and MA. 100 - pH 1.1). Total carbon (TC), organic carbon (OC) and inorganic carbon were analyzed using a CHNS Leco analyser (LECO TruSpec® Micro CHNS 932, Michigan, USA). The particle size distribution of the fine fraction of soils was determined by the laser particle sizer (Partica Laser Scattering LA-950V2 – Laser Particle Size Analyzer. Japan (Tokyo), ATS Burlington, Ontario).

After mineralization of the soil according to the official method of the Quebec government (MA. 205-Mét/P 1.0), metal and metalloid contents were measured by ICP-AES (Varian, Mississauga, ON, Canada) in our laboratories. The analytical results were controlled with certified standard solutions (Multi-elements Standard 900-Q30-100, SCP Science, Lasalle, QC, Canada) and certified soil samples (SQC 001 – Lot 011233). A solution of yttrium was used as an internal standard to ensure that there is no deviation during the analysis.

PCP contents present in soils before and after attrition were determined in our laboratories (INRS-ETE). PCP was extracted from soil samples (5-40 g depending on the particle size) with methylene chloride (300 mL) using a Soxhlet extraction. Then, a solution of sodium hydroxide at 20 g.L⁻¹ was used to transfer the PCP in an aqueous phase and a liquid/liquid extraction step was performed. Then, a derivatization step of PCP was performed using anhydrous acetate and a solution of potassium carbonate (75%, v/v). After one night of vigorous mixing, a liquid/liquid extraction step was done after the addition of methylene chloride. Phenanthren- d_{10} (internal standard) and ¹³C-pentachlorophenol (recovery standard) were added during the preparation of the samples. PCP analysis was performed 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 the CEAEQ method MA. 400 – Phe. 1.0 (CEAEQ 2013).

7 The analysis of the 17 toxic congeners of PCDD/F was performed in our laboratories according to the CEAEQ 8 method MA. 400 – D.F. 1.1 (CEAEQ 2015). After, a Soxhlet extraction of PCDD/F from soil using toluene, 9 purification and concentration steps were performed using a multilayer silica column followed by an alumina 10 column. After evaporation, an internal standard (¹³C-labelled analogues purchased at Wellington laboratories) was 11 added and the sample was analyzed using a GC-MS (Thermo Scientific, model Trace 1310 Gas Chromatograph 12 coupled with mass spectrometer detector ISQ, column type ZB Semi-volatile, 60 m × 0.25 mm × 0.25 μm).

13 2.4.1 Statistical analysis

The relationship between the amount of organic contaminants removed, expressed in g (dependent variable *y*) and the initial amount of contaminant, expressed in g (independent variable *x*) was established as described by the Equation 1 (Cornillon and Matzner-Lober 2006).

17

$$18 \qquad y = ax + b \tag{1}$$

19

Where, y represents the amounts of contaminants removed (g), x represents the initial amounts of contaminant present in the soil (g) and a and b represent the regression coefficients which are estimated using least square approach minimizing the sum of square errors method.

The significance of the regression coefficients was tested at a confidence level of 5%. The implementation of the regression model was made using the R software (Team, 2014). This software gives the estimate value of the regression coefficients and the critical probability (*p*-value) of significance. In order to compare the contaminant removal yields obtained at the end of the attrition process, statistical comparisons were made by Student's test, attrition experiments were made in triplicates (n = 3). The mean values and standard deviation were calculated for each batch of experiments carried out on the same 1-4 mm fraction of soil sample. A calculated *t*-value was determined according to the Equations 2 and 3:

5

6
$$t(n_A + n_B - 2) = \frac{\bar{Y}_a - \bar{Y}_b}{Sp\sqrt{\frac{1}{n_a} + \frac{1}{n_b}}}$$
 (2)

7

8 Where $t(n_A + n_B - 2)$ represented the calculated *t*-value, n_A and n_B represented the number of assays for each 9 series $(n_A = n_B = 3)$; \bar{Y}_a and \bar{Y}_b represented the mean value of each series and S_p^2 is calculated according to the 10 following equation:

11

12
$$S_p^2 = \frac{\sum_{i=1}^{n_A} (Y_{a,z} - \bar{Y}_a)^2 + \sum_{i=1}^{n_B} (Y_{b,z} - \bar{Y}_b)^2}{n_A + n_B - 2}$$
 (3)

13

14 Where $Y_{i,z}$ represented the removal yields obtained for the assays of the serie i.

15 A *t*-Student table was used to determine the threshold defined for a certain confidence level ($\alpha = 0.5\%$) and degree 16 of freedom (df = 4). To conclude an hypothesis was used that, if the *t*-values exceed the threshold value, $t(\alpha = 0.5\%)$. 17 df = 4) = 2.776, the mean values of removal yields are considered as significantly different.

18 **3** Results and Discussion

19 3.1 Influence of initial contaminant contents on the performance of the attrition process

One of the goal of this study was to examine the influence of the initial concentration of contaminants (soils S1, S2, S3, S4 and S5) on the performances of the attrition process on the 1-4 mm soil fraction. Some basic characteristics of the 1-4 mm soil fraction obtained for each soil studied and the initial concentration of both inorganic and organic contaminants are given in Table 1. According to these results, it can be noticed that the 1-4 mm soil fraction

represented approximately 28-30% for all the soil samples studied. The pH measured for all the soil fractions 1 2 studied was neutral, with values varying between 7.02 and 7.21. The total and organic carbon concentrations of all the soil fractions studied were relatively low, varying between 0.48 - 0.75% and 0.35 - 0.62%, respectively. The 3 concentrations of As, Cr and Cu varied from 13 to 55 mg.kg⁻¹, from 27 to 64 mg.kg⁻¹ and from 45 to 160 mg kg⁻¹, 4 5 respectively. These results highlighted that the contamination of the 1-4 mm soil fraction by inorganic compounds 6 was very low. However, the concentrations of PCP and PCDD/F varied from one soil to another one. Indeed, the initial concentrations of PCP measured in the 1-4 mm soil fraction were estimated at 1.1, 2.9, 8.7, 5.1 and 13 mg.kg⁻ 7 8 ¹ for the soil S1, S2, S3, S4 and S5, respectively. These soil fractions also contained different concentrations of PCDD/F, the lowest concentration was estimated at 1,795 ng TEQ kg⁻¹ whereas the highest concentration was 9 estimated at 5,720 ng TEQ.kg⁻¹. Table 2 presents the toxicity equivalency factors and the associated TEQ obtained 10 for each dioxin and furan initially present in the 1-4 mm soil fraction of S1, S2, S3, S4 and S5. These results showed 11 12 that only 12 of the 17 congeners considered as toxic were present in the 1-4 mm soil fraction for all the soil samples (S1, S2, S3, S4 and S5). According to the values obtained, it appeared that the congeners 1,2,3,6,7,8 hexa-13 chlorinated (1,2,3,6,7,8 HCDD), 1,2,3,4,6,7,8 hepta-chlorinated (1,2,3,4,6,7,8 HPCDD) and octa-chlorinated 14 15 (OCDD) dioxins were present in high concentrations in the 1-4 mm soil fractions of S1, S2, S3, S4 and S5.

Five attrition steps of 20 min each, carried out in the presence of a biodegradable surfactant ([BW] = 2%, w.w⁻¹) at 16 room temperature with a pulp density fixed at 40% (w.w⁻¹) were applied to 2 kg of the 1-4 mm fraction of the 17 18 different soils studied. All the experiments were performed in triplicates. According to the results, the removal yields obtained for inorganic contaminants varied between 21 and 51% for As, between 19 and 50% for Cr and 19 20 between 19 and 54% for Cu (values not showed here). A study carried out by Williford et al. (1999) showed that a 21 pre-treatment by attrition allowed a comparable removal yield for Cr (27%). Recently, Metahni et al. (2016) 22 removed 24% of As, 13% of Cr and 32% of Cu from the coarse soil fractions (>0.125 mm) by an attrition process. 23 These low removal yields can be explained by the low initial contents of these metals in the 1-4 mm soil fraction and 24 by the fact that the attrition process was performed at pH = 7 and that the solubility of metals is not favorable at 25 neutral pH. However, the attrition process developed efficiently removed to remove As, Cr and Cu from the 1-4 mm 26 fraction of soils studied indeed, low residual concentrations of these contaminants have been measured in the treated soils. The removal yields obtained for organic contaminants initially present in the 1-4 mm soil fractions were 27 28 estimated at 50, 59, 67, 63 and 64% for PCP and 50, 54, 62, 52 and 55% for PCDD/F for the soils S1, S2, S3, S4

1 and S5, respectively. A study carried out by Metahni et al. (2016) showed that an attrition process allowed the 2 removal of 0 - 85% of PCP and 17 - 64% of PCDD/F from the coarse fractions (>0.125 mm) of different soils, depending of the characteristics of the soil studied. These results highlighted that good removal yields were 3 obtained, regardless the initial concentrations of PCP or PCDD/F initially present in the soil fraction, except for S1. 4 5 The lowest removal yields obtained for the soil S1 (50% for PCP and 50% for PCDD/F) can be explained by the 6 lowest initial concentrations of these contaminants in the 1-4 mm soil fraction compared to the other soils studied. 7 These results prove that the attrition process developed is efficient to remove organic contaminants from the 1-4 mm 8 fraction of soil with the experimental conditions tested.

9 A statistical analysis was performed to determine if the initial concentration of both PCP and PCDD/F can influence 10 the performance of the attrition process to remove these contaminants from the 1-4 mm soil fractions. Figure 2 presents the relationship observed between the amounts of contaminant removed and the initial amounts of 11 contaminant in the 1-4 mm soil fractions of S1, S2, S3, S4 and S5 (It is important to note that it represents quantity 12 13 of contaminants, PCP (mg) and PCDD/F (ng)). According to these results, a linear correlation was observed between the amounts of contaminants removed and the initial amounts of PCP ($R^2 = 0.999$) and PCDD/F ($R^2 = 0.994$). These 14 15 results also highlighted that the performance of the attrition process did not attain an asymptote when the initial 16 amounts of PCP increased from 2.2 to 26 mg and the initial amounts of PCDD/F increased from 3,590 to 11,440 ng 17 for the treatment of 2 kg of the 1-4 mm soil fraction of soils coming from various sampling point on the same site (S1 to S5). 18

According to the statistical analysis performed on these results, it appeared that this linear correlation was significant for both PCP and PCDD/F removals from the 1-4 mm soil fractions (*p*-value = 1.21×10^{-8} for PCP and 5.43 $\times 10^{-7}$ for PCDD/F). When correlated the amounts of both PCP and PCDD/F removed (y) with the initial amounts (x) under the relation y = a x + b, results obtained by the used software have neglect the value at the origin (b). The correlation obtained between the amounts of PCP and PCDD/F removed (y) and the initial quantities (x) are shown in the Equation 4 for PCP and the Equation 5 for PCDD/F.

26	Amount of PCP removed =0.703 x initial amount of PCP	(4)
27	Amount of PCDD/F removed =0.647 x initial amount of PCDD/F	(5)

1 According to these Equations 4 and 5, it appeared that the linear coefficient (a) was lower than 1 for both PCP (a =2 (0.703) and PCDD/F (a = 0.647) regressions, indicating that a small loss of efficiency of the attrition process to remove these contaminants from the 1-4 mm soil fraction was observed when increasing the initial concentration of 3 4 these organic compounds. These Equations can be used to estimate the amounts of organic contaminants that can be 5 removed from the 1-4 mm soil fraction by attrition process in the domain of initial concentrations studied.

6 3.2

Influence of the particle size on the performance of the attrition process

7 The second aim of this study was to review the effect of the particle size on the performance of the attrition process 8 to remove As, Cr, Cu, PCP and PCDD/F from contaminated soils (S1, S2, S3, S4 and S5). Table 3 presents the mass 9 proportions of the different coarse soil fractions (> 12, 4-12, 1-4 and 0.250-1mm), the initial concentration of both 10 inorganic and organic contaminants and the removal yields obtained after the attrition process for each soil studied. 11 According to these results, the coarse fractions represent approximately 21-23% (4-12 mm fraction), 28-30% (1-4 12 mm fraction) and 19-22% (0.125-1 mm) for all the samples studied whereas the coarsest fraction (> 12 mm) represents about 10-16% of the entire soils S1, S2, S3, S4 and S5. The concentrations of As, Cr and Cu varied 13 between 5.2 and 55 mg.kg⁻¹, between 7.4 and 64 mg.kg⁻¹ and between 37 and 160 mg.kg⁻¹, respectively. It can be 14 15 noticed that the contamination of the coarse fractions by inorganic contaminants was very low for all the samples 16 studied. The removal yields obtained at the end of the attrition process for these inorganic contaminants varied from 17 27 to 71% for As, from 15 and 57% for Cr and from 19 and 66% for Cu; indicating that the attrition process was quite efficient to remove these contaminants, even if they were not problematic. Recently, Metahni et al. (2016) 18 removed 24 - 42% of As, 0 - 13% of Cr and 23 - 46% of Cu by an attrition process applied on the coarse fractions 19 20 (>0.125 mm) of four different soils. The present study showed that the performances of attrition process studied to remove As, Cr and Cu from the coarse soil fractions was be improved regardless the particle size compared to the 21 previous study (Metahni et al. 2016). 22

The initial concentrations of both organic contaminants varied between 0.9 and 26 mg.kg⁻¹ for the PCP and between 23 612 ng TEQ.kg⁻¹ and 8,970 ng TEQ.kg⁻¹ for the PCDD/F from one fraction to another one of the different soils 24 studied as showed in Table 3. Moreover, our results highlighted an important variation of the removal yields 25 26 obtained for the different organic contaminants initially present in the coarse fractions of each soil, varying from 18 27 to 89% for PCP and from 20 to 76% for PCDD/F. A study carried out by Metahni et al. (2016) showed that an attrition process applied on the coarse soil fractions (>0.125 mm) allowed quite similar removal yields, 85% for PCP
and 64% for PCDD/F. These important differences of metals, PCP and PCDD/F removal yields can be explained by
the difference of the particle size and/or by the high heterogeneity of the contaminant contents initially present in the
different fractions of soils studied.

According to the results obtained in the Section 3.1, a linear correlation was found between the amounts of 5 6 contaminants removed and the initial amount of PCP and PCDD/F in the 1-4 mm soil fractions of S1, S2, S3, S4 and 7 S5. Another statistical analysis was performed to determine if the particle size can influence the performance of the 8 attrition process to remove PCP and PCDD/F from the coarse soils fractions. Figure 3 presents the relationship 9 observed between the amounts of PCP (Fig. 3a) and PCDD/F (Fig. 3b) removed and the initial amounts of 10 contaminant present in the coarse fractions (It is important to note that it represents > 0.250 mm, 1-0.250 mm, 1-4 mm, 4-12 mm and >12 mm pooled on the same figure, that it represents quantity of contaminants, PCP (mg) and 11 PCDD/F (ng)).) of the soil samples S1, S2, S3, S4 and S5. The results showed that, regardless of the particle size of 12 13 the soil studied, the performance of the attrition process did not attain an asymptote when the initial amounts of PCP and PCDD/F increased from 1.80 to 51.8 mg and from 1,225 to 17,940 ng, respectively, for the treatment of 2 kg of 14 15 soil using an attrition process. Figure 3 presents the correlation coefficients, the *p*-values and the Equations obtained 16 for PCP (Fig. 3a) and for PCDD/F (Fig. 3b) obtained when correlating the amounts of both PCP and PCDD/F 17 removed with the amounts initially present in the coarse soil fraction (> 0.250 mm) of S1, S2, S3, S4 and S5. 18 According to the results, the amounts of contaminants removed were significantly correlated with the initial amounts of PCP and PCDD/F (*p*-value < 0.05 and $R^2 = 0.96$), regardless the particle size of the soil samples studied. In 19 20 contrast, several studies showed that the effect of attrition was positively correlated with the size of soil particles for 21 remove diesel (Feng et al. 2001) and inorganic contaminants (Jobin et al. 2015; Stražišar and Sešelj, 1999). But, 22 PCP and PCDD/F have different nature than other organic contaminants and inorganic contaminants so there are no 23 comparable. According to Petavy et al. (2009) the percentage of fine particles (attrition sludge) created by the 24 friction force will be higher with the coarse fractions.

According to the Equations obtained, it appeared that the linear coefficient (a) was lower than 1 for both PCP (a = 0.77) and PCDD/F (a = 0.65) regressions; indicating that a small loss of efficiency of the attrition process to remove these contaminants from the coarse soils fractions (>0.250 mm) of the soil samples studied was observed when the initial concentration of these organic compounds increased from 0.9 to 26 mg PCP.kg⁻¹ and from 612 to 8,970 ng

TEQ.kg⁻¹. These Equations can be used to estimate the amounts of organic contaminants that can be remove from
the coarse soil samples fractions (>0.250 mm) studied using this attrition process in the domain of initial
concentrations studied for the soil samples studied.

4 3.3 Influence of the nature of soils on the performance of the attrition process

5 In this part, the influence of the nature of the soil (granulometric distribution, TOC, TIC, pH and diverse industrial 6 origin) on the efficiency of the optimized attrition process has been evaluated. Three soils (F1, F2 and F3) 7 contaminated by As, Cr, Cu, PCP and PCDD/F, sampled in three different industrial areas were used. Figure 4 8 presents the mass proportion of the coarse soil fractions for each soil studied. The results showed important 9 differences between the mass proportions of the different soil fractions from one soil to another one, which varied 10 from 15.4 to 33.7%, from 13.7 to 31.0%, from 4.60 to 22.5% and from 23.3 to 34.0% for the >12, 4-12, 0.250-1 and <0.250 mm soil fractions, respectively. According to these results, the soil texture of the different soils studied was 11 12 quite different. Indeed, the soils F1 and F3 were mainly composed of coarse particles (>4 mm) whereas the soil F2 13 contained huge amounts of fine particles (<1 mm). In the case of the 1-4 mm soil fraction, the mass proportion was quite similar for all the soils studied, with values estimated at 14.5, 12.5 and 15.0 for F1, F2 and F3, respectively. 14 15 Table 4 presents some basic characteristics of the 1-4 mm soil fraction measured for each soil studied and the initial 16 concentration of inorganic contaminants. The initial concentrations of both organic contaminants, PCP and PCDD/F, 17 are presented in Figure 5. The pH measured for the 1-4 mm fraction of the soils F1, F2 and F3 estimated at 7.42, 7.65 and 7.37, respectively, indicating that the initial pH of the different soils studied was quite similar. The total 18 19 carbon measured varied between 0.87 and 7.36% and the inorganic carbon varied between 0.51 and 6.71%. The 20 concentration of organic carbon was relatively low, with values estimated at 0.65%, 0.63% and 0.36% for the soil F1, F2 and F3 (fraction 1-4 mm), respectively, compared with the content of carbon matter (5-7%) of soil which 21 used by Petavy et al. (2009). These results highlighted that the 1-4 mm fraction of the different soils studied present 22 23 very low concentration of both inorganic and organic contaminants. The concentrations of inorganic contaminants varied between 7.7 and 10 mg As.kg⁻¹, between 23 and 57 mg Cr.kg⁻¹ and between 35 and 51 mg Cu.kg⁻¹ for the 24 25 different soils studied. However, it was possible to notice that the 1-4 mm fraction of the different soils studied contained similar concentrations of PCP, ranging from 0.32 and 0.49 mg.kg⁻¹ and similar concentrations with an 26 average of 630 ng TEQ.kg⁻¹ for PCDD/F except for F1 which was 876 ng TEQ.kg⁻¹ for PCDD/F. 27

Five attrition steps of 20 min each were applied, in triplicates, to the 1-4 mm fraction of the soils F1, F2 and F3 1 $([BW] = 2\% (w.w^{-1}), PD = 40\% (w.w^{-1}), T = 25^{\circ}C)$. This attrition process allowed the removals of a small 2 percentage of inorganic contaminants, with removal yields varying from 18 to 29% for As, from 10 to 33% for Cr 3 4 and from 14 to 28% for Cu. These low removal yields can be explained by the low initial contents of these metals in 5 the 1-4 mm soil fraction (Table 4). In addition, the attrition process was performed at neutral pH. Indeed, the pH 6 measured in the solution (aqueous phase) after the attrition process varied between 7.3 and 7.5 for all soils studied. 7 These values indicated that the pH of the solution after attrition was similar for the different soils studied, which 8 seemed logical as the pH of the soil samples were similar and near 7.5. Around this pH, the solubility of both metals 9 and organic contaminants is very low, which indicated that the main reasons related to the removals observed for 10 both organic and inorganic contaminants were not due to the pH of the solution. However, the removal of the fine particles, highly contaminated from the coarse particles due to the friction introduced by attrition is the main 11 12 mechanism implicated into the removal of both organic and inorganic contaminants from the soil fractions studied. 13 Figure 5 presents the initial and final concentrations of PCP (Fig. 5a.) and PCDD/F (Fig. 5b.) measured in the 1-4 14 mm soil fraction of the different soils and the associated removal yields. According to these results, the removal 15 yields obtained for the organic contaminants initially present in the 1-4 mm soil fractions were estimated at $65 \pm 1\%$, $75 \pm 6\%$ and $72 \pm 5\%$ for PCP and $72 \pm 3\%$, $70 \pm 8\%$ and $61 \pm 7\%$ for PCDD/F for the soils F1, F2 and F3, 16 17 respectively.

Removed quantity of organic contaminants from the 1-4 mm fraction of soils F1, F2 and F3 were calculated (values not showed here) using these removal yields obtained at the end of the attrition process. Also, these removed quantities were estimated using the Equation 4 for PCP and the Equation 5 for PCDD/F deducted in section 3.1. The ratio calculated removed quantity/estimated removed quantity were 109, 117 and 112% for PCP and 125, 122 and 108% for PCDD/F for soils F1, F2 and F3, respectively. Results showed that there is still 10 to 25% which is not explained by the equations moderating the attrition removal. But the equations overvalued the results.

According to the equality of means test (Student t-test), the t-values were estimated at 2.88 for PCP and 0.42 for PCDD/F (comparison of soils F1 and F2), 2.14 for PCP and 2.58 for PCDD/F (comparison of soils F1 and F3) and 4.25 for PCP and 1.55 for PCDD/F (comparison of soils F2 and F3). The comparison of these t-values and the theorical t-value defined for a confidence interval of 95% and a degree of freedom of 4 (theorical t-value = 2.776) revealed that the calculated t-values were higher than the theorical value for the removal of PCP except for the soils F1 and F3 (t = 2.14). However, the calculated t-values were lower than the theorical value for the removal of PCDD/F from all soils studied. These results indicated that the removal yields of PCP obtained were significantly different; except for the removal of PCP from the soils F1 and F3 which were significantly the same. But, for the removal of PCDD/F results indicated that the removal yields obtained were significantly the same. Therefore, it appeared that the attrition process developed was efficient to remove of PCP and PCDD/F from the 1-4 mm fraction of all soil samples.

7 The proportion of attrition sludge recovered following the attrition process of the soils F1, F2 and F3 were around 8 24, 26 and 19%, respectively. Figure 6 presents the particle diameter of the attrition sludge versus percentage of 9 cumulative particles smaller than 500 µm for the 1-4 mm fraction of the soils F1, F2 and F3. Figure 6 shows that the 10 particle size distribution of the attrition sludge recovered at the end of the attrition of 1-4 mm fraction of the soils 11 F1, F2 and F3 was similar. According to this Figure 6, 50% of the attrition sludge particles have sizes lower than 17, 25 and 20 µm for the soils F1, F2 and F3. According to Petavy et al. (2009) and the general knowledge on attrition 12 13 (Wills, 1992), attrition induces the removal of the fine particles which are easily removed from the coarse particles 14 and transferred to the attrition sludge, allowing the removal of the organic matter present on the surface of these fine 15 particles from soil. A plausible hypothesis is that organic contaminants are adsorbed on this organic matter (Bates et 16 al. 2000; Subramanian, 2007). In our study, the organic matter contents (OC) were around 0.64% for soils F1 and F2 17 (1-4 mm fraction) whereas for the soil F3 was only 0.36% which can explain that the removal yields of PCDD/F 18 from the soils F1 and F2 were quite higher than the removal yield from the soil F3 (1-4 mm fraction). Indeed, as 19 hydrophobic compounds such as PCDD/F have a huge affinity to organic matter, the removal of fine particles which 20 contained huge amounts of organic carbon can explained the better PCDD/F removal performances observed for the 21 soils F2 and F3 than for the soil F1. However, for the PCP initially present in high concentrations (few mg/kg) 22 compared to PCDD/F (few µg/kg), the removal yields of this contaminant from soils F1, F2 and F3 seemed to not be 23 proportional to soils organic matter contents. In fact, despite the fact that the organic matter contents of soils F2 and 24 F3 were different (0.64% and 0.36%, respectively), the removal yields of PCP were similar with an overage of 73%. 25 These results can be explained by the low soils organic matter contents.

According to several authors, organic contaminants are strongly fixed to the organic matter present in the soil, indicating that the efficiency of the decontamination process might be influenced by this component of the soil (Banerji et al. 1993; UK Environment Agency 2009). However, additional experiments should be performed on
 contaminated soil having different amounts of organic carbon to confirm or infirm this hypothesis.

3 4 Conclusion

4 This research evaluated the performances and the robustness of an attrition process to remove As, Cr, Cu, PCP and 5 PCDD/F from the coarse particles (> 0.250 mm) of contaminated soils. Soils studied were sieved to isolate five soils 6 fractions (< 0.250, 0.250-1, 1-4, 4-12 and >12 mm). Five attrition steps of 20 min each, carried out in the presence of a biodegradable surfactant ([BW] = 2%, w.w⁻¹) at room temperature with a pulp density fixed at 40% (w.w⁻¹) 7 8 were applied to the coarse fractions (>0.250 mm) of the different soils studied. The first series of assays was 9 performed on the 1-4 mm fraction of five soil samples, located on the same contaminated site, having different 10 initial concentrations of contaminants to examine the influence of the initial concentration of contaminants on the performances of the attrition process. The results showed satisfactory performances of the attrition process to treat 2 11 kg of soil samples when the initial amounts of PCP and PCDD/F increased from 2.2 to 26 mg and from 3,590 to 12 11,440 ng TEQ, respectively. A statistical analysis was used to evaluate the influence of the particle size on the 13 14 performance of this attrition process. The results showed that the amounts of organic contaminants removed were significantly correlated with the initial amounts of PCP and PCDD/F (*p*-value < 0.05, $R^2 = 0.96$), regardless the 15 16 particle size of the soil samples studied. According to these results, the initial level of contaminants and the particle 17 size seemed to not significantly influence the performances of the attrition process to remove inorganic and organic 18 contaminants from the coarse particles of contaminated soils (degree of confidence = 95%). The last series of assays 19 of attrition was performed on the 1-4 mm fraction using soils contaminated by As, Cr, Cu, PCP and PCDD/F, 20 sampled on three different industrial areas. According to our results, the attrition process allowed an important 21 removal of PCP and PCDD/F from the 1-4 mm soil faction, despite the nature (granulometric distribution, TOC, 22 TIC and diverse industrial origin) of the soil. The removal yields obtained varied between 65 and 75% for PCP and 23 between 61 and 72% for PCDD/F. Therefore, the nature of the soil does not seem to influence the performances of 24 the attrition process to remove organic contaminants from the 1-4 mm fraction of contaminated soils. These results 25 are notably encouraging, and this process can be considered for an industrial application.

26

1 Acknowledgements

- 2 Sincere thanks are extended to the Natural Sciences and Engineering Research Council of Canada (grant number
- 3 RDCPJ 463019-14) for their financial contribution to this study.

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1 Figure Caption List

2	Fig. 1	Flowsheet of the attrition process developed for the decontamination of the coarse particles
3		(> 0.250 mm) of soils contaminated by As, Cr, Cu, PCP and PCDD/F
4	Fig. 2	Correlation between the amounts of contaminant removed and the initial amounts of PCP
5		(a) and PCDD/F (b) present in the 1-4 mm soil fraction for the soils S1, S2, S3, S4 and S5 (1-4
6		mm soil fraction, 5 attrition steps of 20 min each, T = 25° C, PD = 40% (w.w ⁻¹), [BW] = 2%
7		(w.w ⁻¹))
8	Fig.3	Correlation between the amounts of contaminant removed from the coarse fractions (> 0.250
9		mm) and the initial amounts of PCP (a) and PCDD/F (b) for the soils S1, S2, S3, S4 and S5 (5
10		attrition steps of 20 min each, $T = 25^{\circ}C$, $PD = 40\%$ (w.w ⁻¹), $[BW] = 2\%$ (w.w ⁻¹))
11	Fig.4	The mass proportion of the coarse fractions (>12, 4-12, 1-4, 0.250-1 and <0.250 mm) of the
12		soils F1, F2 and F3
13	Fig.5	The initial and final concentrations of PCP (a) and PCDD/F (b) measured in the 1-4 mm soil
14		fraction of the soils F1, F2 and F3 and the associated removal yields
15	Fig.6	The particle size distribution determined by laser granulomere of the attrition sludge of the
16		1-4 mm fraction of soils F1, F2 and F3



Fig. 1. Flowsheet of the attrition process developed for the decontamination of the coarse particles (> 0.250 mm) of soils contaminated by As, Cr, Cu, PCP and PCDD/F *(Where AT: attrition step, R: Rinsing step and S: sieving step)*











Fig. 4.



Fig. 5.



Fig. 6.

Soil sample	S1	S2	S 3	S4	S 5
Characteristics (fraction 1-4 mm)					
Mass percentage (%, w.w ⁻¹)	30	29	28	29	30
pH in water at 25 °C	7.02	7.06	7.15	7.10	7.21
Total carbon content (%)	0.48	0.51	0.74	0.50	0.75
Organic carbon content (%)	0.35	0.42	0.62	0.45	0.57
Inorganic carbon content (%)	0.12	0.09	0.12	0.05	0.18
Contaminants (mg.kg ⁻¹) (fraction 1-4 mm)					
As (mg.kg ⁻¹)	14	14	55	13	21
Cr (mg.kg ⁻¹)	27	30	64	28	31
$Cu (mg.kg^{-1})$	45	46	160	47	61
PCP (mg.kg ⁻¹)	1.1	2.9	8.7	5.1	13
PCDD/F (ng TEQ.kg ⁻¹)	1,795	2,795	5,720	3,130	3,805

Table 1Soil parameters and initial metals, PCP and PCDD/F contents in the 1-4 mmfraction of the soils S1, S2, S3, S4 and S5

	Toxicity	Associate	ГО, 1988)			
Dioxin or Iuran	Equivalency Factor (TEF)	S 1	S2	S3	S4	S5
2,3,7,8 TCDF	0.1	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
2,3,7,8 TCDD	1.0	< 1.6	< 1.6	< 1.6	< 1.6	< 1.6
1,2,3,7,8 PCDF	0.05	< 0.06	< 0.06	< 0.06	< 0.06	< 0.06
2,3,4,7,8 PCDF	0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
1,2,3,7,8 PCDD	0.5	140	141	142	162	177
1,2,3,4,7,8 HCDF	0.1	42.9	70.7	83.0	82.4	106
1,2,3,6,7,8 HCDF	0.1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
1,2,3,7,8,9 HCDF	0.1	65.2	86.2	84.3	92.8	107
1,2,3,4,7,8 HCDD	0.1	56.5	63.7	180	67.9	131
1,2,3,6,7,8 HCDD	0.1	330	408	615	453	639
1,2,3,7,8,9 HCDD	0.1	117	150	159	182	283
2,3,4,6,7,8 HCDF	0.1	28.5	41.0	255	48.2	50.6
1,2,3,4,6,7,8 HPCDF	0.01	158	262	276	227	247
1,2,3,4,6,7,8 HPCDD	0.01	471	627	1,675	763	916
1,2,3,4,7,8,9 HPCDF	0.01	21.5	34.2	48.8	32.6	46.1
OCDD	0.001	290	778	1,800	859	927
OCDF	0.001	74.6	133	404	163	175
Total		1,795	2,795	5,720	3,130	3,805

Table 2Toxicity equivalency factor and associated TEQ for each dioxin or furan initially
present in the 1-4 mm soil fraction of S1, S2, S3, S4 and S5 samples

	N (*	As		Cr		Cu		РСР		PCDD/F	
Soil sample	Mass proportion	Before	RY	Before	RY	Before	RY	Before	RY	Before	RY
	(%)	(mg.kg ⁻¹)	(%)	(mg.kg ⁻¹)	(%)	(mg.kg ⁻¹)	(%)	(mg.kg ⁻¹)	(%)	(ng.kg ⁻¹)	(%)
S1											
> 12 mm	12	5.7	65 ± 3	7.4	50 ± 5	47	37 ± 6	0.9	64 ± 8	612	40 ± 9
4-12 mm	22	7.9	43 ± 2	18	32 ± 6	57	38 ± 8	4.0	34 ± 4	1,130	39 ± 2
1-4 mm	30	14	30 ± 10	27	42 ± 9	45	30 ± 10	1.1	50 ± 4	1,795	50 ± 1
0.250-1 mm	21	16	45 ± 6	24	40 ± 10	70	33 ± 9	7.6	89 ± 0	5,025	65 ± 2
S2											
> 12 mm	11	8.7	57 ± 1	13	31 ± 2	53	45 ± 3	1.9	31 ± 6	713	20 ± 8
4-12 mm	23	7.5	67 ± 3	25	29 ± 7	62	58 ± 4	4.2	18 ± 2	1,180	39 ± 1
1-4 mm	29	14	51 ± 4	30	35 ± 4	46	46 ± 2	2.9	59 ± 7	2,795	54 ± 1
0.250-1 mm	22	15	56 ± 8	23	44 ± 6	73	40 ± 10	3.4	51 ± 4	5,200	61 ± 2
S 3											
> 12 mm	16	18	61 ± 1	28	54 ± 2	37	66 ± 1	1.3	28 ± 1	1,800	40 ± 5
4-12 mm	23	33	42 ± 3	39	35 ± 5	71	40 ± 5	2.6	46 ± 3	3,080	50 ± 2
1-4 mm	28	55	42 ± 6	64	50 ± 4	160	54 ± 3	8.7	67 ± 1	5,720	62 ± 2
0.250-1 mm	23	53	66 ± 1	41	57 ± 4	116	58 ± 2	15	88 ± 1	6,195	58 ± 1
S4											
> 12 mm	10	5.2	42 ± 4	11	45 ± 3	46	44 ± 4	2.8	62 ± 8	1,220	33 ± 3
4-12 mm	23	10	71 ± 2	19	33 ± 4	64	41 ± 2	6.2	40 ± 9	1,690	53 ± 1
1-4 mm	29	13	21 ± 9	28	15 ± 9	47	46 ± 5	5.1	63 ± 2	3,130	52 ± 0
0.250-1 mm	19	16	50 ± 3	24	33 ± 6	73	35 ± 4	2.8	54 ± 5	8,970	76 ± 1
S5											
> 12 mm	10	13	36 ± 6	8.9	53 ± 5	42	32 ± 8	5.3	81 ± 2	970	24 ± 7
4-12 mm	21	11	67 ± 4	26	34 ± 6	59	50 ± 4	15	55 ± 3	2,970	46 ± 5
1-4 mm	30	21	45 ± 5	31	19 ± 7	61	19 ± 9	13	64 ± 1	3,805	55 ± 0
0.250-1 mm	22	19	50 ± 5	26	56 ± 3	84	41 ± 4	26	83 ± 1	6,575	54 ± 1

Table 3Mass proportions, initial contaminant contents measured in the coarse fractions of the soils S1, S2, S3, S4 and S5 and the
removal yields (RY) obtained after the attrition process

Table 4Soil parameters and initial inorganic contaminant . contents present in the 1-4 mmfraction of the soils F1, F2 and F3

Soil sample	F1	F2	F3
Characteristics (fraction 1-4 mm)			
pH in water at 25 °C	7.42	7.65	7.37
Total carbon (%)	7.36	2.17	0.87
Organic carbon (%)	0.65	0.63	0.36
Inorganic carbon (%)	6.71	1.54	0.51
Contaminants (mg.kg ⁻¹) (fraction 1-4 mm)			
As	7.7	5.4	10
Cr	23	23	57
Cu	35	40	51

Diovin or furen congener	Toxicity	Associated TI	Associated TEQ (ng TEQ,kg ⁻¹) (NATO, 1988)			
Dioxin of fur an congener	Equivalency Factor (TEF)	F1	F2	F3		
2,3,7,8 TCDF	0.1	< 0.05	< 0.05	< 0.05		
2,3,7,8 TCDD	1.0	< 1.6	< 1.6	< 1.6		
1,2,3,7,8 PCDF	0.05	< 0.06	< 0.06	< 0.06		
2,3,4,7,8 PCDF	0.5	< 0.5	< 0.5	< 0.5		
1,2,3,7,8 PCDD	0.5	82.0	67.0	113		
1,2,3,4,7,8 HCDF	0.1	72.4	70.5	37.7		
1,2,3,6,7,8 HCDF	0.1	31.4	36.0	25.1		
1,2,3,7,8,9 HCDF	0.1	70	53.0	< 0.3		
1,2,3,4,7,8 HCDD	0.1	< 0.2	< 0.2	36.1		
1,2,3,6,7,8 HCDD	0.1	98.6	71.8	36.1		
1,2,3,7,8,9 HCDD	0.1	29.9	22.1	28.9		
2,3,4,6,7,8 HCDF	0.1	< 0.25	< 0.25	< 0.25		
1,2,3,4,6,7,8 HPCDF	0.01	43.4	36.8	17.4		
1,2,3,4,6,7,8 HPCDD	0.01	202	92.4	124		
1,2,3,4,7,8,9 HPCDF	0.01	42.0	2.87	2.33		
OCDD	0.001	187	145	176		
OCDF	0.001	17.8	45.2	15.0		
Total		876	642	612		

Table 5Toxicity equivalency factor and associated TEQ for each dioxin or furan initially
present in the 1-4 mm soil fraction of F1, F2 and F3 samples