Influence of particle size distribution, organic carbon, pH and chlorides on washing of mercury contaminated soil

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HIGHLIGHTS

- Neither pH adjustment nor chloride introduction facilitated Hg removal from soil.
- No correlation was found between Hg and total/dissolved soil organic matter.
- Hg was firmly bound to soil making washing insufficient for soil clean-up.

ARTICLE INFO

ABSTRACT

Feasibility of soil washing to remediate Hg contaminated soil was studied. Dry sieving was performed to evaluate Hg distribution in soil particle size fractions. The influence of dissolved organic matter and chlorides on Hg dissolution was assessed by batch leaching tests. Mercury mobilization in the pH range of 3–11 was studied by pH-static titration. Results showed infeasibility of physical separation via dry sieving, as the least contaminated fraction exceeded the Swedish generic guideline value for Hg in soils. Soluble Hg did not correlate with dissolved organic carbon in the water leachate. The highest Hg dissolution was achieved at pH 5 and 11, reaching up to 0.3% of the total Hg. The pH adjustment was therefore not sufficient for the Hg removal to acceptable levels. Chlorides did not facilitate Hg mobilization under acidic pH either. Mercury was firmly bound in the studied soil thus soil washing might be insufficient method to treat the studied soil.

1. Introduction

Accumulation of Hg in soil originating from both natural and anthropogenic sources poses a major hazard for soil ecosystem and biosphere (Frohne et al., 2012). Due to the high risks to the human health and the environment (Clarkson et al., 2003; Canuel et al., 2009), Hg is among the priority contaminants to be remediated at the global level (USEPA, 2007).

Methods such as stabilization/solidification, vitrification, electro-remediation, soil washing, thermal desorption, immobilization, phytostabilisation and phytoextraction have been tested to treat Hg contaminated soils (Wang et al., 2012). Soil washing is one of the widely used techniques for Hg contaminated soil allowing for the reduced soil volume to be further treated or disposed of (Abumaizar and Smith, 1999; Dermont et al., 2008; Sierra et al., 2010). It is a physical separation process that utilizes water to concentrate contaminants into a smaller soil volume by means of particle size separation, specific-gravity separation, attrition scrubbing, froth flotation or magnetic separators (Vik and Bardos, 2003; USEPA, 2007). The ability to separate fractions with low contamination from those having high contaminant concentrations varies for different soils due to different origin of contamination (e.g., mining related contaminants that occur in mineral phases versus soil contaminated with chemical spillage where contaminants occur as soluble salts and particle coatings). Chemical extraction where solvents such as acids or alkalis are applied can be used to assist physical separation (FRTR, 2001; Bollen and Biester, 2011). The soil remediation method is then called chemical extraction. Generally, acids and alkalis release Hg pollutants from soil by solubilizing Hg compounds or/and soil components that sorb Hg (Schuster, 1991; Dermont et al., 2008).

Properties of contaminated soil might differ substantially from those of natural soil. Soil characterization is therefore necessary to estimate the practical and economic feasibility of soil

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washing/chemical extraction for each remediation case (Mann, 1999; Sierra et al., 2010).

This study aimed at evaluating the potential of soil washing to remediate a Hg contaminated soil by determining the total and soluble Hg concentrations in different soil particle size fractions; Hg association with soil organic matter (SOM); and the influence of pH and chlorides on Hg mobilization in the contaminated soil.

2. Materials and methods

2.1. Sample preparation

Three soil samples (Table 1) were collected from Tidermans padding area about 10 km upstream of Göta River, Sweden. Mercury contamination in the soil resulted from waste disposal, chlor–alkali process and harbor activities (Sweco Viak, 2007). This soil was also polluted by other trace elements, i.e., zinc, lead and copper. Three samples were pooled into one composite sample of approximately 45 kg (wet weight). The composite sample was oven-dried for 48 h at 45 °C, manually disaggregated, homogenized and subsequently sub-divided by a Riffle splitter prior to the experiment.

2.2. Soil sieving

The prepared samples were dry sieved into particle-size fractions of (in mm) <0.063, 0.063–0.125 , 0.125–0.25, 0.25–0.5, 0.5–1, 1–2, 2–4, 4–6.3, 6.3–12.5, 12.5–25 and >25 through normalized sieves positioned in an analytical sieve shaker (AS 200 control Restch) for 10 min. Wet sieving was performed afterwards with a water supply (1.5 L min^{-1}) on the top of the uppermost sieve and a water collection at the bottom (ISO/TS 17892-4:2004). Size fractions below 4 mm from the dry sieving were used for further leaching tests and extractions.

2.3. Soil and solution analysis

2.3.1. Soil

Soil pH and electrical conductivity were measured in 1:2 v/v soil-distilled H$_2$O suspensions. Total Hg and other element concentrations were measured by accredited laboratory ALS Scandinavia to determine the Hg distribution in different particle size fractions (modified USEPA methods 200.7 (ICP-AES) and 200.8 (ICP-SFMS)). Samples were digested with 5 mL conc. HNO$_3$ + 0.5 mL 30% H$_2$O$_2$ in closed Teflon containers in a microwave digestion system prior to element analyses. Total organic carbon (TOC) was assessed to measure the organic content of bulk soil and particle size fractions with a TOC analyzer (TOC-VCPH/CPN Shimadzu). The samples were pre-treated with concentrated HCl to remove the inorganic carbon. The remaining carbon was oxidized at 900 °C and the formed CO$_2$ was analyzed by non-dispersive infrared absorbance.

2.3.2. Mercury solubility in distilled water

Water-soluble Hg in bulk soil and particle size fractions was assessed by a batch leaching test at liquid-to-solid ratio (L/S) 10 for

24 h ± 1 h to assess the release of Hg upon contact with distilled water (SS-EN 12457-4).

2.3.3. The pH-dependent dissolution of Hg

A pH-static leaching test (24 h ± 1 h, L/S 10) was performed to determine the Hg dissolution in the pH range of 3–11 by automatic titrator (TitroMess-2000). The pH adjustments were: 0.1 M HNO$_3$ for pH 3 and 5; 0.1 M NaOH for pH 7; and 1 M NaOH for pH 9 and 11.

The pH-static titration at pH 3 and 5 using 0.1 M HCl as the titrant was additionally performed to determine the influence of chlorides on Hg mobilization in comparison to nitrate.

2.3.4. Dissolution of Hg associated with SOM

Bulk soil was mixed either with 0.1 or 0.4 M NaOH solution (L/S 10) and rotated for 24 h ± 1 h to assess the impact of different alkali concentrations on the release of Hg associated with SOM (Wallschläger et al., 1998; Boszke et al., 2008).

2.3.5. Hydrochloric acid extraction

In order to evaluate the effect of chlorides on Hg mobilization, 0.01, 0.05 and 0.1 M HCl solutions were used to extract Hg from the bulk soil (L/S 10) using a batch leaching test for 24 h and 48 h.

Eluates from the leaching and titration tests were filtrated through 0.45 μm nitrocellulose membrane filter prior to further analyses. Samples were acidified with ultra-high purity HNO$_3$ before element measurement (modified USEPA methods 200.7 (ICP-AES) and 200.8 (ICP-SFMS)). Dissolved chloride and sulfate ions were determined by liquid chromatography (CSN EN ISO 10304-1 and CSN EN ISO 10304-2); dissolved organic carbon (DOC) was measured following CSN EN 1484 procedure. These analyses were performed by accredited laboratory ALS Scandinavia.

2.3.6. Thermo-desorption of Hg

Analysis of thermo-desorption of Hg was carried out in triplicate for three replicate bulk soil samples (<4 mm) (nine in total) after HCl titration at pH 3 to identify the changes in Hg speciation. The technique is based on the thermal decomposition of Hg compounds from solids at different temperatures and continuous determination of the released volatile Hg (Biester and Scholz, 1996).

2.3.7. Statistical evaluation and modeling

Element speciation was calculated using the geochemical equilibrium modeling software Visual MINTEQ v3.0, using default parameters (Gustafsson, 2012). Information about solid-solution distribution and solution speciation of Hg(II) was obtained assuming that sorption by solid-phase humic acids (HA) and fulvic acids (FA) was the predominant sorption mechanism. The input data used were based on the compositions of the leachates and included pH, temperature (20 °C), and total concentrations of major elements of Ca, K, Mg, Na, Cl, SO$_4$ and DOC. For Hg and some other metals (Cu, Zn and Pb), the geochemically active concentrations were assumed to constitute 50% of the total concentrations. Model conditions were set according to the following hypothesis: (1) 50% of TOC is active; TOC contains 50% FA and HA, i.e. the FA/HA ratio = 1; 75% of DOC is FA; (2) Al and Fe were not included in the modeling as they are expected not to compete with Hg for specific functional groups of SOM (Stumm and Morgan, 2012). Pearson's correlation coefficient (r) was assessed to evaluate linear relationships between two variables.

Table 1
Initial characteristics of Hg-contaminated soil (±SD, n = 3).

Soil properties	Unit	Value
pH	–	6.5
Electrical conductivity (EC)	mS cm^{-1}	2.3 ± 0.1
Total organic carbon (TOC) (n = 4)	%	8 ± 2
Dissolved organic carbon (DOC)	mg kg^{-1}	209 ± 14
Hg concentration (n = 6)	mg kg^{-1}	34 ± 14

3. Results

3.1. Mercury distribution in soil particle size fractions

The particle size distribution obtained by dry sieving (Table 2) indicates that the soil consisted of more than 98% of coarse-grained fraction (Jury and Horton, 2004). Mercury concentrations of size fractions below 4 mm ranged from 10 to 49 mg kg^{-1} and decreased with increasing particle sizes (Table 2). The finest soil fraction contained nearly five times more Hg than the coarsest analyzed fraction. Even the least contaminated fraction (2–4 mm) substantially exceeded the Swedish generic guideline value for Hg in soils with less sensitive use, i.e. industrial and commercial land use (2.5 mg kg^{-1}) (Swedish EPA, 2009). The concentrations of water-soluble Hg, i.e. the amount of soil Hg dissolved in distilled water with no pH modifications, ranged from 4 to 37 μg kg^{-1} in particle size fractions, corresponding to 0.03–0.2% of total Hg. No apparent relationship was observed between water-soluble Hg and the particle size fractions (Table 2).

Larger amount of fine fractions (up to 0.25–0.5 mm) were observed in wet sieving than in dry sieving (Fig. 1), implying that more fines were bound to coarser particles in dry sieving.

3.2. Total organic carbon (TOC) in solid samples

The TOC contents varied from 5 to 11% in the particle size fractions, but no clear differences were observed in relation to the particle sizes (Table 3). No correlation ($r = 0.4$) was observed between TOC contents and total Hg ($n = 21$).

3.3. Mercury solubility in water

The concentrations of water soluble Hg were low in all fractions (Table 2), indicating that Hg was firmly bound to the soil matrix and was hardly leached by water. The contents of DOC increased with decreasing particle sizes and ranged from 0.2% to 0.4% of TOC (Table 3). No correlation was observed between DOC and soluble Hg ($r = 0.1$), while strong correlations were shown between DOC and Cd ($r = 0.92$) and Zn ($r = 0.94$).

3.4. The pH-dependent Hg dissolution

The amounts of Hg desorbed from soil at different pH values varied, but the desorption pattern was similar in all particle size fractions (Fig. 2). The least Hg dissolution was achieved at pH 3 and 9 (0.2–2.9 μg kg^{-1}), while the dissolution peaks were observed at pH 5 and 11 (11–43 μg kg^{-1}) in all fractions (Fig. 2).

A small increase in Hg dissolution was observed at pH 3 using HCl as the titrant compared to HNO$_3$. However, at pH 5, chloride ions seemed to have less impact on Hg mobilization than nitrate
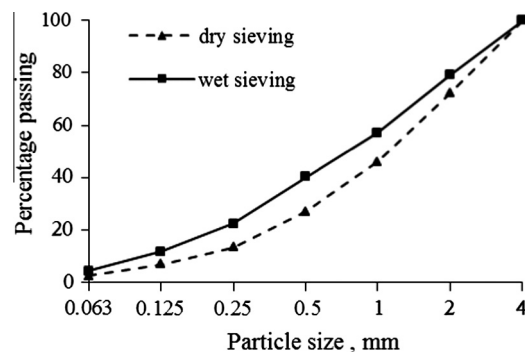

Fig. 1. Cumulative curves of particle size fractions (<4 mm) determined by dry and wet sieving of the studied soil.

ions. The content of DOC was slightly higher at pH 3 than pH 5 when HCl was the titrant while the concentration of dissolved Hg was higher at pH 5 using both titrants (Fig. 3).

3.5. Dissolution of Hg associated with SOM

Mercury desorption was largely enhanced by NaOH washing compared to distilled water (Table 4). But the dissolved Hg accounted for only a small percentage (up to 1.5%) of the total Hg and did not show any increase at higher NaOH concentration (Tables 2 and 4).

3.6. Thermo-desorption curves (TDCs) of Hg

Mercury in nine soil samples after HCl titration at pH 3 was classified as humic/matrix-bound and chloride species according to the TDCs (Fig. 4a and b). The TDCs of most of these peaks were quite symmetric and none of them was at the temperature below 100 °C, implying no occurrence of Hg(0) species.

3.7. Geochemical modeling and equilibrium calculations

According to the output of model exercise (Fig. 5), the majority of dissolved Hg was bound to chlorides when 0.1 M HCl was used, while dissolved organic matter (DOM) contributed as much as chlorides for dissolved Hg binding when 0.05 M HCl was used. In the absence or at very low concentration (0.01 M) of chlorides, DOM dominated the binding for dissolved Hg. The free form of Hg(II) was negligible. Elevated chloride concentrations accompanied by lower pH were supposed to enhance Hg mobilization according to the model simulation (Fig. 5). However, dissolved Hg concentration at acidic pH, as observed in this experiment, was substantially lower than predicted by the model (Fig. 6). A

Table 2
Distribution of particle size fractions and total Hg (Hg$_T$) in solid soil samples after dry sieving and soluble Hg (Hg$_S$) in leachates of the soil particle size fractions (±SD, $n = 3$).

| Texture	Particle size (mm)	Percentage passing	Hg$_T$ (mg kg^{-1})	Hg$_S$ (μg kg^{-1})
Clay/silt	<0.063	1.32	49 ± 2	16 ± 2
Sand	0.063–0.125	2.05	42 ± 2	27 ± 4
	0.125–0.25	3.08	30 ± 1	25 ± 1
	0.25–0.5	6.04	25 ± 6	37 ± 6
	0.5–1	8.42	20 ± 3	35 ± 3
	1–2	11.11	24 ± 5	16 ± 3
Gravel/stone	2–4	9.55	10 ± 1	4 ± 2
	4–6.3	14.65	8 ± 1	–
	6.3–12.5	15.51	6 ± 2	–
	12.5–25	14.06	3 ± 1	–
	>25	14.21	3 ± 3	–

"–" not measured.

Table 3
Characteristics of fractionated and bulk soil (EC: electrical conductivity; TOC: total organic carbon; DOC: dissolved organic carbon, ±SD, n = 3).

Particle size (mm)	EC (mS cm^{-1})	pH (1:2 H$_2$O)	TOC (%)	DOC (mg kg^{-1})	DOC/TOC (%)
<0.063	2.5 ± 0.1	6.56–6.64	8.9 ± 0.1	391 ± 11	0.44
0.063–0.125	2.6 ± 0.1	6.55–6.65	8.7 ± 0.2	336 ± 2	0.39
0.125–0.25	2.5 ± 0.1	6.47–6.53	9.2 ± 0.5	256 ± 2	0.28
0.25–0.5	2.4 ± 0.1	6.43–6.57	8.9 ± 0.9	213 ± 2	0.24
0.5–1	2.2 ± 0.0	6.46–6.54	10.9 ± 0.7	190 ± 5	0.17
1–2	1.8 ± 0.1	6.58–6.62	8.5 ± 2.1	172 ± 9	0.20
2–4	1.3 ± 0.1	6.69–6.71	4.9 ± 1.5	145 ± 5	0.29
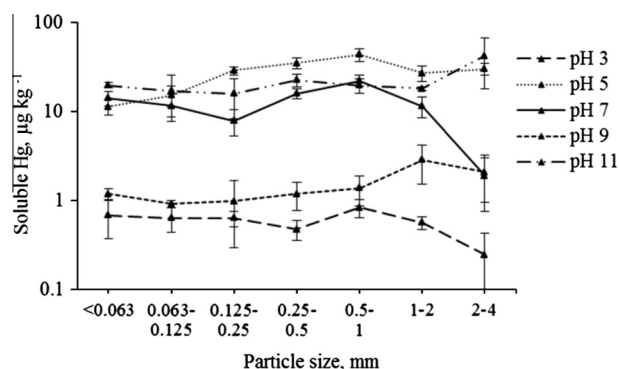
Fig. 2. The pH-dependent dissolution of Hg in soil particle size fractions using HNO$_3$ as the titration acid. Note the logarithmic concentration scale. Error bars represent standard deviation of means, n = 3.
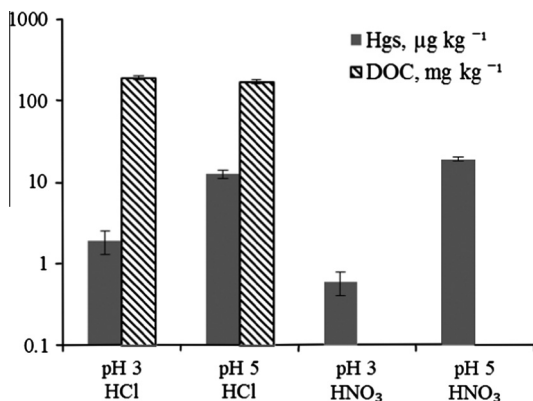
Fig. 3. Soluble Hg (Hg$_S$) and dissolved organic carbon (DOC) after titrations to pH 3 and 5 using HCl and HNO$_3$ as the titrants. Note the logarithmic concentration scale. Error bars represent standard deviation of means, n = 3.

Table 4
Mercury dissolution in NaOH solution (± SD, n = 3).

NaOH concentration (M)	0	0.1	0.4
Soluble Hg (µg kg^{-1})	30 ± 3	500 ± 48	470 ± 56
pH	6.5	12.6–12.8	12.9–13.1
Electrical conductivity (EC, mS cm^{-1})	2.3 ± 0.1	12.3 ± 0.2	54.3 ± 2.9

pH dependent dissolution of Hg (similar to what was shown in Fig. 2) was observed regardless of the presence of chlorides.

4. Discussion

4.1. Soil washing efficiency by particle size separation

Usually, fine soil particles have a larger specific surface area and tend to bind more contaminants than large particles (Table 2).

However, even sand/gravel fractions (below 4 mm) contained substantial concentrations of Hg (Table 2). Different curves obtained for dry and wet sieving (Fig. 1) indicate that coarser particles contained some amount of fines attached to them, demonstrating the lower effectiveness of particle size separation via dry sieving. Mercury concentrations in particle size fractions after wet sieving were not measured, but judging from the leaching test results (Table 2), the removal of Hg from all particle size fractions with water is expected to be very low.

Organic content of soil might also influence Hg distribution (Dermont et al., 2008). TOC usually exhibits a strong correlation with Hg concentration due to the high affinity of Hg for SOM functional groups (Schuster, 1991; Kwaansa-Ansah et al., 2012). Nevertheless, there was no correlation between TOC and total Hg in this case (r = 0.4), which could be due to the competition with soil mineral particles that have larger specific surface area (e.g., Fe, Mn, Al (hydr)oxides) (Liao et al., 2009). On the other hand, the quality of functional groups of SOM might be more important than the quantity of SOM for Hg binding (Jing et al., 2007). Hg(II) is expected to preferentially bind with thiol (–SH) and other reduced sulfur-containing groups (Skyllberg et al., 2006), which are present only in trace quantities in SOM (Ravichandran, 2004). Quantification of functional Hg-binding groups of SOM in different particle sizes might help to better understand the Hg association with SOM (Xia et al., 1999; Manceau and Nagy, 2012).

4.2. Influence of DOM, pH and chlorides on Hg mobilization in the soil

4.2.1. DOM

In general, strong positive correlation between soluble Hg and DOC is expected in cases where Hg is primarily derived from wetlands and soils, where Hg is released and co-transported with the natural DOM (Wallschläger et al., 1996; Åkerblom et al., 2008; Miller et al., 2011). The Hg contamination in the studied soil resulted from anthropogenic activities and it is likely that organic contaminants have altered the distribution of organic carbon in soil particle size fractions, which have interfered with the correlations between Hg and SOM. Other elements, such as Cd and Zn, that were present in elevated concentrations showed significant positive correlations with DOC, hence they are competing with Hg for sorption sites (Lin and Chen, 1998; Turer and Maynard, 2003; Amir et al., 2005).

Several studies have demonstrated that DOM might act as a reducer by transforming Hg(II) in the solution to Hg(0) thereby decreasing Hg dissolution (Ravichandran, 2004; Gu et al., 2011). Although a higher level of DOC was observed, while lower Hg dissolution was shown at pH 3 compared to pH 5 (Fig. 3), no species transformation to Hg(0) was indicated by the thermo-desorption (Fig. 4a and b).

4.2.2. pH

It is well established that pH is an important factor controlling the mobility of Hg in soil by changing Hg speciation (Barrow and Cox, 1992; Yin et al., 1996). The observed low dissolution of Hg
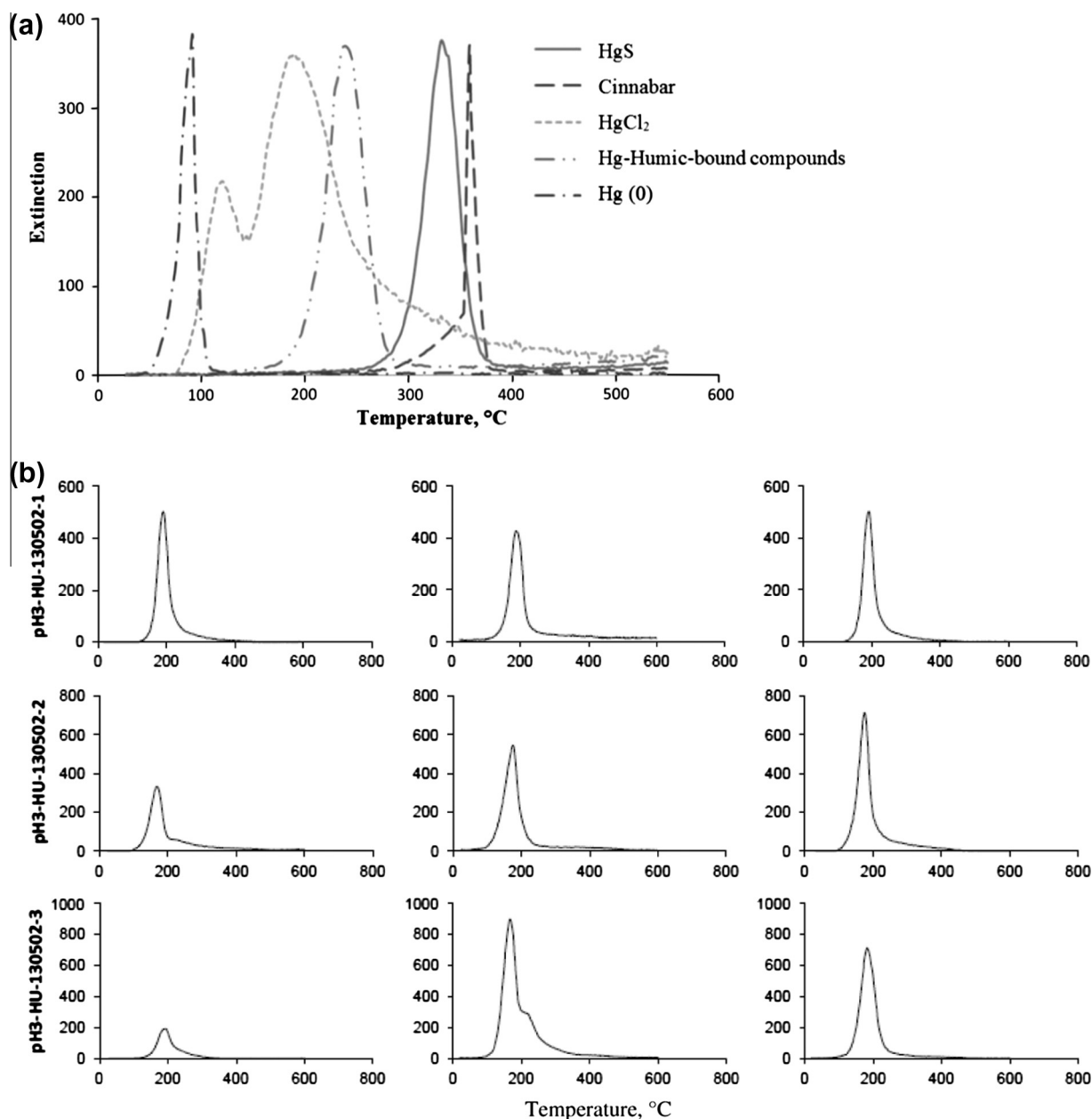
Fig. 4. (a) Reference measurement of Hg thermo-desorption curves of soil; (b) Thermal desorption curves of Hg in bulk soil after HCl titration to pH 3. Three replicate samples after titration examined in triplicate (hence nine graphs in total).

at pH 3 (Figs. 2 and 6) was earlier reported by Yin et al. (1996), who explained it by the precipitation of SOM at this pH. HA is one of the main constituents of SOM and is insoluble under strongly acidic conditions; therefore, Hg bound to HA would be expected to co-precipitate at pH 3 (Hem, 1970; Reimers and Krenkel, 1974; Walls-chläger et al., 1996). Besides, adsorption of Hg(II) to soil mineral particles could also occur at this pH range. Maximal adsorption of Hg(II) just below pH 4 was reported for goethite (Barrow and Cox, 1992). Considerable Hg adsorption on hydrous MnO_2 between pH 2.5 and 3 were found by Lockwood and Chen (1973). They explained the formation of the Mn-hydroxide complex was responsible for Hg adsorption in this pH range. The apparent increase in Hg dissolution at pH 5 and 7 compared to pH 3 (Figs. 2 and 6) was likely to result from the dissolution of HA that retained Hg in the soil at pH 3. The decrease of desorbed Hg from pH 5 to 7 could be caused by the competition between Hg^{2+} and hydronium ions

(H_3O^+) for adsorption sites on the soil particles at acidic pH (Semu et al., 1987). When pH increased from 7 to 9, Hg dissolution decreased (Fig. 2), most likely due to that elevated pH increases the quantity of negative charges of soil particles, which might attract and retain Hg(II) ions (Semu et al., 1987). In addition, hydrolysis of Hg increases with increasing pH, and mercuric hydroxide has been reported to be increasingly adsorbed by soil constituents from pH 5 to 9 (Farrah and Pickering, 1978). This mechanism could also contribute to a higher Hg adsorption at pH 9 in comparison to pH 7. An obvious increase in Hg dissolution from pH 9 to 11 (Fig. 2) was probably due to the continuous dissolution of HA that bound Hg. Results of the dissolution of Hg associated with humic matter (Table 4) further confirmed this hypothesis, with much enhanced Hg dissolution at pH around 13 compared to neutral pH. Moreover, decreasing concentrations of Hg^{2+} and $HgOH^+$ with increasing pH, as hydrolysis of these charged species proceeds to uncharged
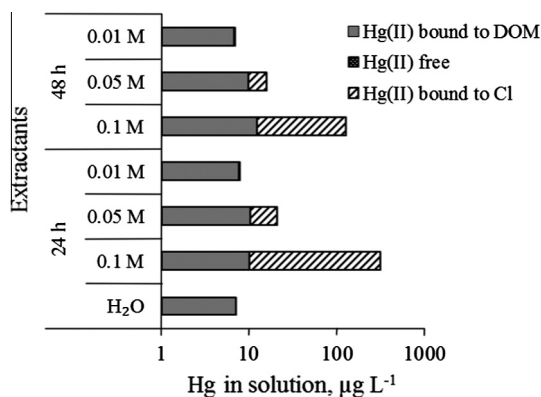
Fig. 5. Model output of Hg species in solution after extraction by HCl of various concentrations for 24 h and 48 h and distilled water for 24 h. Note the logarithmic concentration scale.
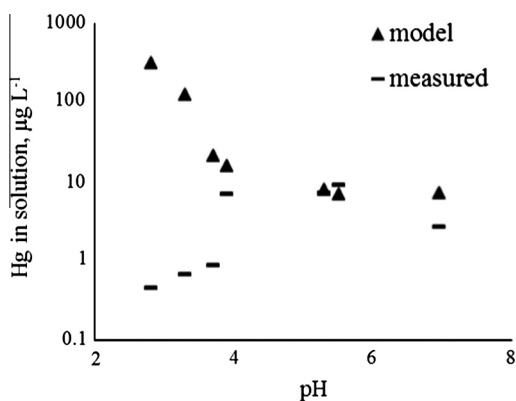

Fig. 6. Experimental data and model simulation of Hg released into solution at different pH using distilled water and HCl of 0.01, 0.05 and 0.1 M as the extractants for 24 and 48 h. Note the logarithmic concentration scale.

$Hg(OH)_2$ might also cause the elevated Hg dissolution at alkaline pH with negative charges of the soil (Farrah and Pickering, 1978).

4.2.3. Chlorides

Chlorides are regarded as one of the most mobile complexing agents for Hg (Kabata Pendias, 2011) and able to compete with OH^- and even organic ligands for Hg bonding (Payne, 1964; Reimers and Krenkel, 1974; Gabriel and Williamson, 2004). According to the model simulation, chlorides were very competitive with SOM for complexing dissolved Hg at acidic pH (Fig. 5). Besides, Hg solubility was predicted to increase at lower pH values in HCl leachates (Figs. 5 and 6). However, the results of the experiment indicate that chlorides had little effect on Hg mobilization in the studied soil (Fig. 6). These discrepancies might be due that data in the Visual Minteq database used to obtain the binding parameters for Hg(II)-FA/HA (Gustafsson, 2012) were insufficient to simulate the conditions in our study. Moreover, much higher chloride concentrations than studied (0.1 M HCl) might be needed to mobilize Hg (USEPA, 2007).

Similar findings as what was shown in Fig. 3 were obtained by Yin et al. (1996), who found that the addition of Cl^- at pH 3 had almost no effect on the desorption of Hg(II) in soils high in SOM. It is possible that the non-soluble SOM forms ternary complexes with Hg-Cl (Yin et al., 1996). Chloride content in the leachate after HCl titration to pH 3 was calculated to decrease (not shown), indicating that chlorides were sorbed to the soil. Additionally, thermal desorption curves indicate that Hg(II) was bound to SOM as well

as to chlorides (Fig. 4a and b), implying possible formation of SOM bound Hg-Cl.

5. Conclusions

The studied soil was coarse-grained and the total Hg concentration decreased with increasing particle sizes. However, even the least contaminated fraction (2–4 mm) substantially exceeded the Swedish generic guideline value for Hg in soils with less sensitive use (2.5 mg kg^{-1}). Particle size separation and the use of water were insufficient to remove Hg to the acceptable levels. As little as 0.03–0.2% of the total Hg was removed from soil fractions with water, showing a strong affinity of Hg for soil constituents. No correlation between the total and dissolved Hg and SOM (both total and dissolved) was identified for the studied soil.

Although different pH values affected Hg dissolution and an enhanced Hg desorption was observed at pH 5 and 11, soil washing by pH adjustment was insufficient for Hg removal, as the highest amount of mobilized Hg at these two pH values was only up to 0.3% of the total soil Hg. The pH 3 should be avoided for Hg washing since Hg was shown to be least soluble at this pH. Increased chloride concentration through addition of 0.1 M HCl acid did not improve Hg mobilization either in the studied soil.

The best result was obtained by the use of NaOH solution at pH ~13, where 1.5% of the total Hg was removed from soil. This, however, was still not sufficient to reach the acceptable Hg levels in soil. Mercury was firmly bound to soil particles and the studied soil washing conditions were not sufficient to extract Hg. Stabilization techniques should be considered for the future soil treatment.

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