

Remediation of heavy metal polluted sediment by suspension and solid-bed leaching: Estimate of metal removal efficiency

Christian Löser^{a,*}, Andreas Zehnsdorf^b, Petra Hoffmann^c, Heinz Seidel^c

^a Dresden University of Technology, Institute of Food Technology and Bioprocess Engineering, 01062 Dresden, Germany

^b UFZ Centre for Environmental Research Leipzig-Halle, Environmental and Biotechnology Centre (UBZ),
Permoserstrasse 15, 04318 Leipzig, Germany

^c UFZ Centre for Environmental Research Leipzig-Halle, Department of Bioremediation, Permoserstrasse 15, 04318 Leipzig, Germany

Received 3 April 2006; received in revised form 4 July 2006; accepted 7 July 2006

Available online 5 September 2006

Abstract

Remediation of heavy metal polluted sediment by extracting the metals with sulfuric acid can be performed as follows: abiotic suspension leaching, microbial suspension leaching, abiotic solid-bed leaching, and microbial solid-bed leaching. Abiotic leaching means that the acid is directly added, while microbial leaching means that the acid is generated from sulfur by microbes (bioleaching). These four principles were compared to each other with special emphasis on the effectiveness of metal solubilization and metal removal by subsequent washing. Abiotic suspension leaching was fastest, but suspending the solids exhibits some disadvantages (low solid content, costly reactors, permanent input of energy, high water consumption, special equipment required for solid separation, large amounts of waste water, sediment properties hinder reuse), which prevent suspension leaching in practice. Abiotic solid-bed leaching implies the supply of acid by percolating water which proceeds slowly due to a limited bed permeability. Microbial solid-bed leaching means the generation of acid within the bed and has been proven to be the only principle applicable to practice. Metal removal from leached sediment requires washing with water. Washing of solid beds was much more effective than washing of suspended sediment. The kinetics of metal removal from solid beds 0.3, 0.6 or 1.2 m in height were similar; when using a percolation flow of $20 \text{ l m}^{-2} \text{ h}^{-1}$, the removal of 98% of the mobile metals lasted 57–61 h and required 8.5, 4.2 or 2.3 l kg^{-1} water. This means, the higher the solid bed, the lower the sediment-mass-specific demand for time and water.

© 2006 Elsevier Ltd. All rights reserved.

Keywords: Dredged sludge; Percolator; Bioleaching; Column; Metal extraction; Washing

1. Introduction

Heavy metal contaminated aquatic sediment is still an unsolved environmental problem. There is an increasing worldwide interest in sustainable and cost-effective remediation methods. A remediation process has therefore been developed in which the heavy metals are removed by solid-bed bioleaching. In a first step, freshly dredged sludge

is divided into a less polluted coarse fraction and a strongly polluted, organic-rich fine fraction to reduce the quantity of sediment which has to be treated by leaching (Löser et al., 2001b). In a second step, the fine fraction is planted with helophytes converting the sludge into a soil-like material (Löser et al., 1999b, 2001a; Löser and Zehnsdorf, 2002; Seidel et al., 2004). Pretreatment in such a manner makes sediment sufficiently permeable for solid-bed leaching: elemental sulfur (S^0) added to the sediment is oxidized by indigenous microbes to sulfuric acid that, in turn, dissolves the toxic heavy metals which, in the end, are extracted by percolating water (Seidel et al., 1998, 2004; Löser et al., 1999a, 2001a, 2006). The leached material is finally

* Corresponding author. Tel.: +49 351 46 33 25 95; fax: +49 351 46 33 77 61.

E-mail address: christian.loeser@tu-dresden.de (C. Löser).

neutralized with lime and revitalized with compost (Seidel et al., 2004). The heavy metal-loaded water from the leaching process is treated by alkalization (Seidel et al., 2004) or electrolysis (Fischer et al., 2004, 2005). Feasibility studies on a pilot scale have demonstrated the practicability of this concept (Seidel et al., 1998, 2004).

Besides our studies on sediment, the principle of solid-bed leaching had also been tested for remediation of heavy metal polluted soil (Ried, 1988; White et al., 1998; Heil et al., 1999; Dessi et al., 2000; Sun et al., 2001; Tampouris et al., 2001; Thayalakumaran et al., 2003; Hauser et al., 2005). Metal solubilization and metal removal occurred simultaneously or successively. The following operation modes have been described:

1. Abiotic leaching by a liquid containing the leaching agent, passing the bed once only (Heil et al., 1999; Sun et al., 2001; Thayalakumaran et al., 2003).
2. Abiotic leaching by a liquid containing the leaching agent and subsequent washing with water, both passing the bed once only (Dessi et al., 2000; Sun et al., 2001; Hauser et al., 2005).
3. Abiotic leaching by a continuously circulating liquid containing the leaching agent and simultaneous regeneration of the liquid by metal removal (Ried, 1988; Tampouris et al., 2001).
4. Abiotic leaching by continuously circulating liquid containing the leaching agent and subsequent washing with water, the latter passing the bed once only (Löser et al., 1999a).
5. Bioleaching at continuous liquid circulation and partial exchange of the liquid by water for metal removal (Seidel et al., 1998).
6. Bioleaching at continuous liquid circulation and subsequent washing with water, the latter passing the bed once only (White et al., 1998; Löser et al., 1999a; Seidel et al., 2004).

Four various leaching procedures have been formerly studied on a laboratory scale using heavy metal polluted Weisse Elster River sediment as an example: (1) abiotic suspension leaching (Löser et al., 2001a, 2005), (2) microbial suspension leaching (Löser et al., 2001a, 2005), (3) abiotic solid-bed leaching (Löser et al., 1999a, 2006), and (4) microbial solid-bed leaching (Löser et al., 1999a, 2001a, 2006). In this contribution, these four leaching principles are compared to each other with special emphasis on the effectiveness of metal solubilization (leaching in a narrower sense) as well as metal removal by subsequent washing. As yet only minimal information on the washing process has been available, although this step offers some opportunities for saving time and material.

2. Materials and methods

Leaching experiments were carried out with ripened Weisse Elster River sediment – an oxidized and soil-like

material – which has been formerly extensively characterized (Löser et al., 2005). The sediment contained the following target heavy metals related to dry mass (in mg kg^{-1}): Zn 3291, Cr 515, Cu 322, Pb 312, Ni 286, and Cd 36. The total amount of target heavy metals was 4792 mg kg^{-1} .

The leaching process comprises two steps: the solubilization of metals and the removal of solubilized metals by washing the sediment with water. In connection with the washing step, the term of ‘mobile metals’ is used. This term refers to the six target heavy metals that can be removed by washing the sediment with water. These metals are dissolved in the liquid phase (process water and pore water in the solids) and bound adsorptively to the sediment (see below).

At suspension leaching, sediment of 20 g dry mass was leached in 200 ml aqueous phase (Löser et al., 2005), the solid and liquid phase were separated by centrifugation, the supernatant was removed, the solids were suspended in 200 ml fresh water on a rotary shaker at 130 rpm for 1 h, and then the liquid phase was removed again as already described. The extraction procedure was repeated three times and the quantity of metals removed in each step was calculated from the volume and metal content of the aqueous solutions, where the latter was analyzed by atomic emission spectroscopy as formerly described (Löser et al., 2005).

At solid-bed leaching, the sediment was leached with 2 l aqueous phase per kg dry mass using a percolator system (Löser et al., 2006). After leaching, the liquid phase was withdrawn from the system and the solid bed was washed with an average water flow of 150 ml h^{-1} (equal to $20 \text{ l m}^{-2} \text{ h}^{-1}$). In detail, 150 ml water was sprinkled within 15 min followed by a break lasting 45 min. Periodic sprinkling ensured a better spreading of water over the bed surface. The leachate, i.e., the liquid which leaked from the solid bed, was collected at intervals and analyzed regarding its volume and metal content. In addition, the leachate flow was sampled repeatedly to follow the washing kinetics.

The quantity of metals solubilized during leaching and removed by washing were calculated as follows: $m_{\text{Me}}(\text{Solubilization}) = C_{\text{Me,Liquid}} (V_{\text{Liquid}}(t=0) - \sum V_{\text{Sample } i} + V_{\text{Acid}}) + \sum (C_{\text{Me,Sample } i} V_{\text{Sample } i})$ and $m_{\text{Me}}(\text{Removal}) = \sum (C_{\text{Me,Sample } i} V_{\text{Sample } i}) + \sum (C_{\text{Me,Leachate } j} V_{\text{Leachate } j})$. These equations take into consideration changes in the liquid volume by sampling and supplying acid during leaching (the latter only at abiotic leaching), and the discharge of metals by sampling (where the withdrawal of the liquid phase at the end of leaching was also regarded as sampling). The masses of the target heavy metals (Zn, Ni, Cu, Cd, Pb, and Cr) were summarized and then divided by the mass of these metals initially present in the sediment to obtain the fraction of metals solubilized or removed. The fraction of heavy metals removed was also calculated from the residual metal content of leached and washed sediment.

3. Results and discussion

3.1. Suspension leaching versus solid-bed leaching

Four various leaching principles were comparably studied by using heavy metal polluted Weisse Elster River sediment (Fig. 1). Sulfuric acid acted as the leaching agent which was directly added (abiotic leaching) or microbially generated from S^0 by means of indigenous leaching-active bacteria (bioleaching).

In comparison with other leaching principles, abiotic suspension leaching was fastest because of the direct addition of the acid and homogeneous mixing, where the latter minimized possibly rate-limiting transport phenomena. Nevertheless, adjustment of the equilibrium concerning metal solubilization lasted some days due to intra-particle diffusion processes (Löser et al., 2005).

Microbial suspension leaching ran slower since the formation of sulfuric acid from the added S^0 required time (Fig. 1). The kinetics of this leaching process was primarily governed by the kinetics of S^0 oxidation. In the case considered, the S^0 was oxidized by a purely hyperbolic kinetics (Löser et al., 2005) and, thus, the rate of metal solubilization gradually diminished with time.

When solely evaluated from the reaction kinetics, abiotic suspension leaching seems to be superior to all other leaching principles. However, suspension leaching exhibits some disadvantages compared to solid-bed leaching, preventing its application on a larger scale, as follows:

1. Suspension reactors contain lower volume-specific quantities of sediment than solid-bed reactors (dry mass of solids related to the reaction volume). Suspension

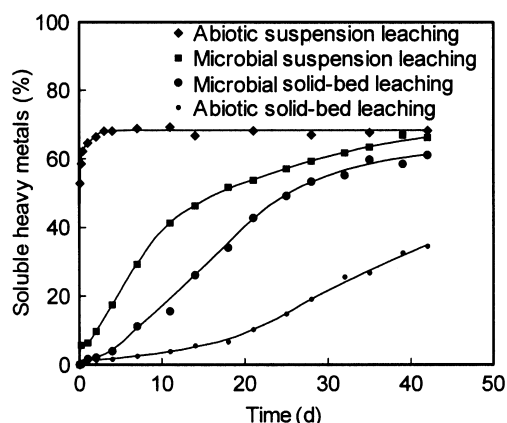


Fig. 1. Fraction of solubilized heavy metals (sum of Zn, Ni, Cu, Cd, Cr and Pb) during abiotic leaching or bioleaching of Weisse Elster River sediment in suspension or in the solid bed (abiotic suspension leaching with $624 \text{ mmol kg}^{-1} \text{ H}_2\text{SO}_4$ at 20°C ; microbial suspension leaching with $624 \text{ mmol kg}^{-1} \text{ S}^0$ – i.e., with a mass fraction of 2% S^0 w/w – at 25°C ; microbial solid-bed leaching with $624 \text{ mmol kg}^{-1} \text{ S}^0$; abiotic solid-bed leaching with process water acidified with H_2SO_4 to pH 2.8; in both solid-bed experiments a bed 1.2 m high was percolated with $801 \text{ m}^{-2} \text{ h}^{-1}$ at 25°C).

leaching of sediment has been studied in a wide range of solid content: 2 to 200 g l^{-1} with 50 g l^{-1} as an average (17 references). In our studies, suspension leaching was always operated with 100 g l^{-1} since higher densities hampered homogenization. At solid-bed leaching, the bulk density determines the quantity of solids per bed volume. In microbial solid-bed leaching of Weisse Elster River sediment on a pilot scale (with 1000 kg dry mass), the bulk density amounted to 513 g l^{-1} (Seidel et al., 2004), which was significantly higher than in suspension leaching.

2. Suspension reactors are cost-intensive. Equipment for suspending the solids is necessary (stirrer tanks or rotary barrels), acidic conditions require corrosion-resistant construction materials, and suspension leaching is operated in closed reactors while solid-bed leaching may be done in open, only downward sealed basins.
3. Suspending the solids requires a permanent input of energy. The more solids a suspension contains, the higher the energy costs for agitation.
4. Suspending the solids during leaching and subsequent washing for metal removal requires large quantities of water (for details see below).
5. The separation of the leached sediment from the suspension requires special equipment. The separation of the solids by simple sedimentation is difficult and time-consuming due to the fineness of sediment particles (agitation stresses the particles and makes them smaller), but separation by filtration or centrifugation is costly. At solid-bed leaching, liquid-solid separation proceeds simply by gravity.
6. Waste water which arises from suspension leaching (leachate and washing water) is characterized by large volume but low concentrations of metals, which complicates waste water treatment.
7. Due to mechanical stress during suspension leaching, the sediment becomes finer, muddy-pasty and swollen which hinders its further use. But sediment leached in the solid bed maintains its soil-like structure.

Although solid-bed leaching shows many advantages over suspension leaching, no information on remediation of polluted sediment by solid-bed leaching is available – apart from our own studies (Löser et al., 1999a, 2001a, 2006; Seidel et al., 1998, 2004). This is surprising at first, but becomes clear upon closer inspection: solid-bed leaching requires material sufficiently permeable to air and water (oxygen is needed for microbial S^0 oxidation in bioleaching, water percolation is necessary for sulfuric acid supply in abiotic leaching, and washing the bed is required for metal removal in both processes), but freshly dredged sediment is nearly impermeable and therefore unsuitable for solid-bed leaching (Seidel et al., 1998). An upstream process, in which freshly dredged sediment is planted with helophytes, converts the sludge into a soil-like and easily permeable material (Löser et al., 1999b; Löser and Zehndorf, 2002; Seidel et al., 2004). Freshly dredged Weisse

Elster River sediment had a water permeability of $0.3 \text{ l m}^{-2} \text{ h}^{-1}$ while sediment treated by helophytes had a permeability of at least $200 \text{ l m}^{-2} \text{ h}^{-1}$ (Löser and Zehndorf, 2002).

For abiotic solid-bed leaching, sulfuric acid as the leaching agent is supplied to the sediment by percolating liquid. The leaching kinetics in this process is primarily determined by the transport of the acid into the bed; the higher the percolation flow, the lower the pH of the circulating water, and the lower the solid-bed height is, the faster the heavy metals are solubilized (Löser et al., 2006). However, the percolation flow and pH are subjected to restrictions. And a low solid bed is uneconomical since capital costs for a solid-bed reactor arise related to its base area rather than its height or, in other words, an increase in bed height lowers the specific costs. Abiotic solid-bed leaching on a large scale – i.e., leaching of a high solid bed with a limited permeability at a restricted pH – requires too much time (Fig. 1).

Bioleaching in the solid bed and bioleaching in suspension proceeds similarly (Fig. 1) since the rate of microbial S^0 oxidation, which determines the kinetics of metal solubilization, was analogous in both processes. The rate of solid-bed bioleaching was nearly independent of the percolation flow as well as the solid-bed height since the acid is generated homogeneously within the bed rather than supplied from outside (Löser et al., 2006).

When Weisse Elster River sediment was leached as described for experiments shown in Fig. 1 (application of $624 \text{ mmol kg}^{-1} \text{ S}^0$ in bioleaching, or $624 \text{ mmol kg}^{-1} \text{ H}_2\text{SO}_4$ in abiotic suspension leaching, or pH-controlled application of H_2SO_4 in abiotic solid-bed leaching), then the equilibrium pH was uniformly 2.8, i.e., independent of the leaching procedure. The same was true for the solubilized heavy metals (Löser et al., 1999a, 2001a, 2005, 2006; Seidel et al., 2004); when the equilibrium concerning pH and dissolved metals was reached, then the six target heavy metals were mobilized to about 65%, and could be removed by washing the sediment with water. However, the six target heavy metals reacted quite different to acidification (Löser et al., 2005): Zn, Ni and Cd were easily dissolved (between 80 and 90%), Cu was moderately leached (about 50%), Cr was little soluble (less than 10%), and Pb was insoluble under the given leaching conditions. In the case considered, the most critical toxic metals Zn and Cd could be removed to a high degree (residual content 400 and 6 mg kg^{-1} , respectively).

3.2. Removal of solubilized heavy metals by washing

3.2.1. Kinetics of metal removal from the solid bed

After treatment of the sediment by abiotic or microbial leaching in the solid bed at a continuous percolation, the package was washed with fresh water passing through the bed once only as described above. The metal content of the leachate leaving the solid bed was repeatedly analyzed (Fig. 2). Target heavy metals and other mobile metals aris-

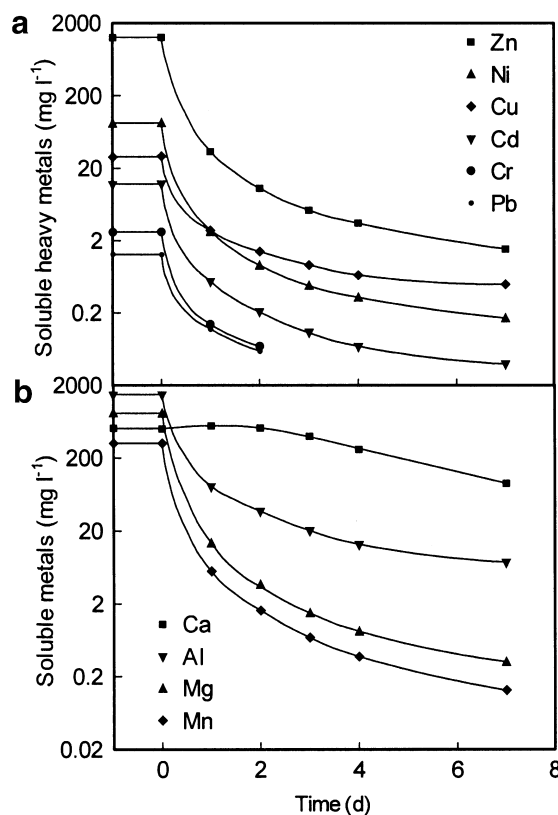


Fig. 2. Content of target heavy metals (a) and other mobile metals (b) in the process water at the solid-bed outlet during washing of sediment after 42 d of solid-bed bioleaching in a percolator system (leaching of 1 kg Weisse Elster River sediment with 2% S^0 w/w in a solid bed 0.3 m in height at a percolation rate of $20 \text{ l m}^{-2} \text{ h}^{-1}$ and $25 \text{ }^\circ\text{C}$ (Löser et al., 2006) and washing with water at an average percolation rate of $20 \text{ l m}^{-2} \text{ h}^{-1}$).

ing from dissolution of mineral compounds were analyzed since both affect the downstream waste water treatment.

The kinetics of metal extraction from the solid bed depended highly on the kind of metal and was influenced by the solubility of its sulfate and its adsorption to the sediment matrix. A limited solubility was found for Ca. The leached sediment contains large quantities of this metal in the form of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). The solubility of gypsum in water at $20 \text{ }^\circ\text{C}$ amounts to 2 g l^{-1} which equals 470 mg l^{-1} of Ca. As long as the sediment package still contained gypsum, the leachate was saturated with this compound resulting in a Ca content of about 500 mg l^{-1} (Fig. 2b).

The adsorption of a metal to the leached sediment becomes visible from its retarded discharge during washing. The weakest adsorption was detected with Mg and Mn: their leachate concentrations were reduced by the factor 2500 during 7 d of washing. Zn, Ni, Cd, Al and Cu were more readily adsorbed, resulting in a weaker decrease in their leachate concentrations. The strongest adsorption was found with Cu: its leachate concentration was reduced only by the factor 60. A retarded Cu discharge was also observed at leaching of heavy metal polluted soil (White et al., 1998; Dessi et al., 2000; Vogeler, 2001; Hauser et al., 2005).

3.2.2. Metal solubilization versus metal removal

Heavy metals which have been solubilized during leaching have to be removed from the sediment by subsequent washing. The quantities of heavy metals solubilized during leaching and removed through washing were compared to each other for various solid-bed bioleaching processes (Table 1).

The amount of heavy metals removed by washing was on the average 4% higher than the fraction of solubilized metals. This discrepancy can be explained by the above-mentioned adsorption of metals to the sediment. Adsorptively bound heavy metals were not detected in the circulating liquid during the leaching period, but were extracted by water passing through the leached sediment during the washing period.

The fraction of metals removed seemed to be dependent on the way in which the metal removal had been quantified (Table 1). In detail, the fraction calculated from the amount of metals still present in the leached and washed sediment was on average 2.5% smaller than the fraction calculated from the amount of metals discharged with the leachate. This apparent discrepancy results from a shrinking of sediment mass during leaching and washing; the acid dissolved toxic and other metals like Ca, Al and Mg originating from the mineral matrix, the sediment mass reduced by about 6%, the content of readily soluble metals like Zn, Ni, Cu and Cd decreased less than expected, and the content of less soluble metals such as Cr and Pb even increased.

3.2.3. Demand on water for leaching and washing

The quantity of water necessary for leaching and washing is an important economic factor in the developed remediation process. An extensive usage of water increases material costs as well as results in large amounts of waste water which requires special treatment.

The demand on water for metal extraction depends on the extent to which mobile heavy metals are to be removed. Their complete removal seems to be illusory; this would require too much water and time. This is not necessary as well since the leached and washed sediment is finally neutralized and stabilized by lime which also immobilizes heavy metals still dissolved in the pore water by precipita-

tion (Seidel et al., 2004). Leaching and subsequent washing is solely aimed at a reduction in the absolute content of heavy metals in the sediment shortly below their threshold values.

For suspension leaching, large amounts of water are necessary to suspend the sediment and to extract solubilized metals. Suspension leaching of Weisse Elster River sediment was always carried out with a solid content of 100 g l^{-1} which corresponds to 10 l kg^{-1} water, and further 20 l kg^{-1} of water was required to remove at least 98% of the mobilized metals – in part dissolved in the adhesive and pore water and in part bound adsorptively to the sediment – by repeated extraction of the leached sediment. Thus, 30 l of water was used to clean-up sediment of one kg dry mass.

For solid-bed leaching, various modes of leaching and washing are possible (see above). In our studies, the sediment was at first leached with continuous liquid circulation (Löser et al., 2006). A quantity of 1.5 l kg^{-1} water was necessary in this process (2 l kg^{-1} liquid phase less 0.5 l kg^{-1} moisture in the used sediment). Thereafter, the sediment was washed with water passing the bed once only to remove the solubilized metals. This is a low-technology process since water simply passes through the bed driven by gravity, and no equipment is required to separate the liquid from the solids as it is necessary for washing suspended sediment.

In order to evaluate the efficiency of metal removal by solid-bed washing, the mass of target heavy metals removed at a given time (calculated as described above) was related to the mass of these metals extractable, at a maximum, from the sediment (estimated from the former by extrapolation). The heavy metal content of the leachate quickly diminished and, after 2 d, more than 95% of the mobile metals had been removed even with the highest solid bed examined (Table 2). In this period, the removal of heavy metals was predominantly based on the displacement of metal-rich pore water by washing water. The removal of adsorbed metals required further washing.

Surprisingly, the height of the solid bed only slightly affected the rate of metal removal. Although a solid bed 1.2 m in height contained four times more sediment than a solid bed 0.3 m high, the kinetics of metal removal were

Table 1

Fraction of soluble heavy metals after 42 d of solid-bed bioleaching (column 2), and fraction of removed heavy metals calculated from the metals extracted with water (column 3) or from the metals remaining in the washed sediment (column 4) (leaching of 1 kg Weisse Elster River sediment with 2% S^0 in a solid bed 0.3 m in height at various percolation rates and 25 °C (Löser et al., 2006) and washing with water for 7 d at a percolation rate of $20 \text{ l m}^{-2} \text{ h}^{-1}$)

Percolation rate ($\text{l m}^{-2} \text{ h}^{-1}$)	Soluble heavy metals ^a (from process water analysis) (%)	Removed heavy metals ^a (from washing water analysis) (%)	Removed heavy metals ^a (from sediment analysis) (%)
1	69	74	69
5	64	68	66
20	57	62	62
80	68	70	67
On average:	64.5	68.5	66.0

^a Heavy metals = sum of Cd, Cr, Cu, Ni, Pb and Zn.

Table 2

Efficiency of heavy metal removal by washing of sediment with fresh water after solid-bed leaching at various solid-bed heights (washing of 1, 2 or 4 kg Weisse Elster River sediment in solid beds 0.3, 0.6 or 1.2 m in height at a percolation rate of $20 \text{ l m}^{-2} \text{ h}^{-1}$ and 25°C)

Solid-bed height (m)	Heavy metal removal relative to extractable heavy metals ^a				
	After 1 d (%)	After 2 d (%)	After 3 d (%)	After 4 d (%)	After 7 d (%)
0.3	92.7	96.8	97.9	98.5	99.4
0.6	89.6	97.1	98.3	98.9	99.7
1.2	80.2	96.4	98.3	99.0	99.8

^a Heavy metals = sum of Cd, Cr, Cu, Ni, Pb and Zn.

rather similar (Table 2). This fact becomes even more pronounced when the fraction of heavy metals removed is depicted against the sediment-mass-specific washing water consumption (Fig. 3).

For reasons of cost-effectiveness, the mobile toxic metals should not be removed completely by washing. But rather, 98% of the mobile metals should be removed while the residual 2% should be immobilized by lime during the sub-process of sediment revitalization (Seidel et al., 2004). The removal of 98% of mobile metals from solid beds 0.3, 0.6 or 1.2 m in height required 8.5, 4.2 or 2.3 l washing water per kg leached sediment (mean values from several experiments, respectively). The higher the solid bed was, the better the washing power of the percolated water was utilized, and the smaller the quantity of water required was. Taking the usage of 1.5 l kg^{-1} water in the leaching period into account, 3.8, 5.7 or 10 l kg^{-1} of water were necessary for the whole process, which was substantially lower than the 30 l kg^{-1} required in suspension leaching. As a result of a minor quantity of water applied, all metals removed were more concentrated in a smaller volume of waste water which makes waste water treatment more effective.

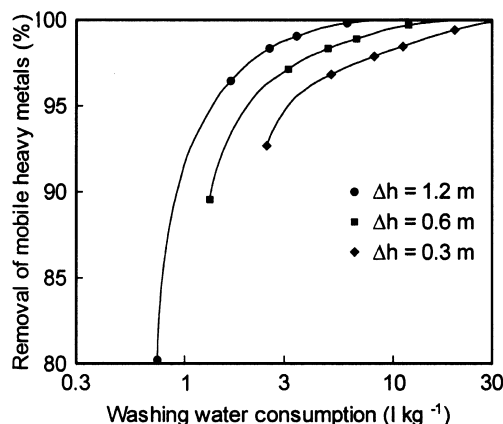


Fig. 3. Removal of mobile heavy metals by washing sediment after solid-bed leaching at various solid-bed heights depending on the specific washing water consumption (washing of 1, 2 or 4 kg Weisse Elster River sediment in solid beds 0.3, 0.6 or 1.2 m in height at an average percolation rate of $20 \text{ l m}^{-2} \text{ h}^{-1}$ and 25°C); Heavy metals = sum of Zn, Ni, Cu, Cd, Cr and Pb.

Moreover, the time necessary to remove 98% of the mobile metals was nearly independent of the solid-bed dimension. In using a percolation flow of $20 \text{ l m}^{-2} \text{ h}^{-1}$, the washing process lasted only 57–61 h.

4. Conclusions

Four various leaching procedures were compared to each other: (1) abiotic suspension leaching, (2) microbial suspension leaching, (3) abiotic solid-bed leaching, and (4) microbial solid-bed leaching. Several arguments were given that explain why suspension leaching is not feasible on a larger scale. For example, the quantity of water necessary for leaching and extracting the heavy metals is up to eight times higher when sediment is leached in suspension rather than the solid bed. Abiotic and microbial solid-bed leaching differ in the way by which the sediment and the leaching agent come together. In abiotic solid-bed leaching, the H_2SO_4 is supplied from outside by percolating water with a rate highly dependent on the solid-bed height and percolation flow. But in microbial solid-bed leaching, the acid is produced within the bed with a rate independent of these two process parameters. When solid-bed leaching is performed on a larger scale (i.e., leaching a bed of extended height and limited permeability), then microbial leaching is much faster than abiotic leaching (Löser et al., 2006) and therefore more cost-effective. All these facts condensed shows that remediation of heavy metal polluted sediment by leaching is most efficient when solid-bed bioleaching is performed.

Successful large-scale solid-bed bioleaching has been demonstrated with Weisse Elster River sediment of 20000 kg dry mass (Löser et al., 2002). Estimates of treatment costs for the solid-bed bioleaching resulted in an amount of 37–110 € per 1000 kg dry sediment depending on the scenario (see appendix in Löser et al. (2002) and Seidel et al. (2004)).

Acknowledgements

The authors would like to express their gratitude for the kind financial support provided by the Deutsche Bundesstiftung Umwelt foundation (AZ 12099). We are grateful to Dr. R. Wennrich and Dr. P. Morgenstern from the UFZ Department of Analytical Chemistry for providing the metal analysis data.

References

- Dessi, R., Fadda, S., Peretti, R., Serci, A., Zucca, A., 2000. Soil decontamination at the Montevecchio–Levante mine site with experimental washing and leaching techniques. *Ann. Chim.* 90, 687–694.
- Fischer, R., Seidel, H., Rahner, D., Morgenstern, P., Löser, C., 2004. Elimination of heavy metals from leachates by membrane electrolysis. *Eng. Life Sci.* 4, 438–444.
- Fischer, R., Seidel, H., Morgenstern, P., Förster, H.-J., Thiele, W., Krebs, P., 2005. Treatment of process water containing heavy metals with a two-stage electrolysis procedure in a membrane electrolysis cell. *Eng. Life Sci.* 5, 163–168.

- Hauser, L., Tandy, S., Schulin, R., Nowack, B., 2005. Column extraction of heavy metals from soils using the biodegradable chelating agent EDDS. *Environ. Sci. Technol.* 39, 6819–6824.
- Heil, D.M., Samani, Z., Hanson, A.T., Rudd, B., 1999. Remediation of lead contaminated soil by EDTA. I. Batch and column studies. *Water Air Soil Pollut.* 113, 77–95.
- Löser, C., Zehndorf, A., 2002. Conditioning of freshly dredged heavy metal-polluted aquatic sediment with reed canary grass (*Phalaris arundinacea* L.). *Acta Biotechnol.* 22, 81–89.
- Löser, C., Seidel, H., Hoffmann, P., Zehndorf, A., 1999a. Remediation of heavy-metal-polluted river sediments by bioleaching using the percolation principle. In: De Schutter, G. (Ed.), *CATS 4 Characterisation and Treatment of Sediments*. Technologisch Instituut vzw, Antwerpen, pp. 213–222.
- Löser, C., Zehndorf, A., Hoffmann, P., Seidel, H., 1999b. Conditioning of heavy metal-polluted river sediments by helophytes. *Int. J. Phytoremed.* 1, 339–359.
- Löser, C., Seidel, H., Hoffmann, P., Zehndorf, A., 2001a. Remediation of heavy metal-contaminated sediments by solid-bed bioleaching. *Environ. Geol.* 40, 643–650.
- Löser, C., Zehndorf, A., Fussy, M., Morgenstern, P., 2001b. Möglichkeiten zur Kostenreduzierung bei der Entsorgung Schwermetall-kontaminierter Flußsedimente – ein Fallbeispiel. *Altlasten Spektrum* 10 (1), 18–27.
- Löser, C., Zehndorf, A., Hoffmann, P., Seidel, H., 2002. Reinigung schwermetallbelasteter Sedimente durch Bioleaching. In: *UFZ-Bericht 13/2002*, UFZ Centre for Environmental Research, Leipzig, pp. 1–192.
- Löser, C., Zehndorf, A., Görsch, K., Seidel, H., 2005. Bioleaching of heavy metal polluted sediment: kinetics of leaching and microbial sulfur oxidation. *Eng. Life Sci.* 5, 535–549.
- Löser, C., Zehndorf, A., Görsch, K., Seidel, H., 2006. Remediation of heavy metal polluted sediment in the solid bed: comparison of abiotic and microbial leaching. *Chemosphere* 65, 9–16.
- Ried, M., 1988. Cadmium-Elimination aus Böden. *Wasser Luft Betrieb* 32 (4), 57–59.
- Seidel, H., Ondruschka, J., Morgenstern, P., Stottmeister, U., 1998. Bioleaching of heavy metals from contaminated aquatic sediments using indigenous sulfur-oxidizing bacteria: a feasibility study. *Water Sci. Technol.* 37 (6–7), 387–394.
- Seidel, H., Löser, C., Zehndorf, A., Hoffmann, P., Schmerold, R., 2004. A bioremediation process for heavy metal contaminated sediments: feasibility study on a pilot scale. *Environ. Sci. Technol.* 38, 1582–1588.
- Sun, B., Zhao, F.J., Lombi, E., McGrath, S.P., 2001. Leaching of heavy metals from contaminated soils using EDTA. *Environ. Pollut.* 113, 111–120.
- Tampouris, S., Papassiopi, N., Paspaliaris, I., 2001. Removal of contaminant metals from fine grained soils, using agglomeration, chloride solutions and pile leaching techniques. *J. Hazard. Mater.* B84, 297–319.
- Thayalakumaran, T., Vogeler, I., Scotter, D.R., Percival, H.J., Robinson, B.H., Clothier, B.E., 2003. Leaching of copper from contaminated soil following the application of EDTA. I. Repacked soil experiments and a model. *Aust. J. Soil Res.* 41, 323–333.
- Vogeler, I., 2001. Copper and calcium transport through an unsaturated soil column. *J. Environ. Qual.* 30, 927–933.
- White, C., Sharman, A.K., Gadd, G.M., 1998. An integrated microbial process for the bioremediation of soil contaminated with toxic metals. *Nature Biotechnol.* 16, 572–575.