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1947

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Effect of sediment size on bioleaching of heavy metals from contaminated sediments of Izmir Inner Bay

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Received 04 October 2012; revised 11 January 2013; accepted 17 January 2013

Abstract
The effect of sediment size on metals bioleaching from bay sediments was investigated by using fine (\(< 45 \mu m\)), medium (45–300 \(\mu m\)), and coarse (300–2000 \(\mu m\)) size fractions of a sediment sample contaminated with Cr, Cu, Pb, and Zn. Chemical speciation of the metals in bulk and size fractions of sediment were studied before and after bioleaching. Microbial activity was provided with mixed cultures of Acidithiobacillus thiooxidans and Acidithiobacillus ferrooxidans. The bioleaching process was carried out in flask experiments for 48 days, by using 5% (\(W/V\)) of solid concentration in suspension. Bioleaching was found to be efficient for the removal of selected heavy metals from every size fraction of sediments, where the experiments with the smaller particles resulted in the highest solubilization ratios. At the end of the experimental period, Cr, Cu, Pb and Zn were solubilized to the ratios of 68%, 88%, 72%, and 91% from the fine sediment, respectively. Higher removal efficiencies can be explained by the larger surface area provided by the smaller particles. The changes in the chemical forms of metals were determined and most of the metal releases were observed from the reducible and organic fractions independent from grain size. Higher concentrations were monitored in the residual fraction after bioleaching period, suggesting they are trapped in this fraction, and cannot be solubilized under natural conditions.

Key words: sediment; sediment size; heavy metals; bioleaching; Acidithiobacillus; microbial activity
DOI: 10.1016/S1001-0742(12)60198-3

Introduction
Among the inorganic contaminants, heavy metals are known to be serious components of pollution in aquatic sediments. The treatment of metal contaminated sediments can be achieved by physical or chemical methods. Recently, biological treatment technologies have become available for remediation of metal contaminated sites. Biological technologies are commonly used for the removal of organic contaminants and are being applied for metal remediation (Evanko and Dzombak, 1997). Bioleaching is a biological method adapted from the mining industry, and recently it has been used for the metals remediation of soils, sediments, and sludge. The principle is to solubilize metal contaminants, following treatment with the activity of the chemolithotrophic bacteria, mainly Acidithiobacillus ferrooxidans and Acidithiobacillus thiooxidans. Under aerobic conditions, the bacterial activity of thiobacilli leads to the production of sulfuric acid, and metals are extracted from the sediment (Seidel et al., 1995). Various physicochemical and biological parameters have effects on the bioleaching process. Among them, microorganism type and concentration, the initial pH, oxidation-reduction potential, nutrients, and substrate concentration are of particular importance and their influences on bioleaching efficiency have been widely studied (Chen and Lin, 2000, 2001; Tsai et al., 2003a; Seidel et al., 2005, 2006; Gomez and Bosecker, 1999; Akinci and Guven, 2011). In addition, the chemical fractionation of heavy metals after bioleaching has been reported for sewage sludge (Lombardi et al., 2006), soil (Kumar and Nagra, 2009), mine tailings (Liu et al., 2008), and sediments (Tsai et al., 2003b). Although there are many different findings and discussions on heavy metals bioleaching, such studies continue to be of great interest, therefore this subject is still valid to be investigated in detail.

Size is a fundamental property of sediment particles. It provides important clues about the nature and provenance of the sediments (Pye and Blott, 2004). The concentrations of absorbed metals depend on particle size and it also affects the solubility of metals depending on the changing surface area. Therefore, distinctive sediment bodies with different size structures will show separate leaching prop-
erties depending on the size distributions. The influence of particle size on metals leaching has been a subject of various investigations in the past. With regards to the mining activities, a large portion of these studies were carried out using sulphide ores and pyrite in order to observe the leaching ability of the metals from the mineral material (Ilyas et al., 2010; Nemati and Harrison, 1999; Deveti, 2004). However, there is no other study investigating the sediment size impact on the efficiency of bioleaching as a pollution treatment method.

The aim of this study was to determine the effect of sediment size on bioleaching efficiencies of Cr, Cu, Pb, and Zn by considering the environmental concerns. Izmir Inner Bay sediments were used in this study, since previous pollution monitoring and bioleaching studies with different bacteria type were conducted for this region (Guven and Akinci, 2008; Akinci and Guven, 2011). A general approach was made for size distribution, and the sediments smaller than 45 μm were regarded as fine fraction. A midpoint between medium and coarse sand fraction was chosen as 300 μm and the range of 45–300 μm was named as medium fraction. Sediments between 300 and 2000 μm were regarded as the coarse fraction which may also include smaller particles such as silt and clay that may be aggregated and captured between larger particles. Any pre-treatment method, such as disaggregation, was avoided not to influence the chemical speciation of metals in sediment. After the bioleaching experiments, the metals remaining in the sediments were measured in order to establish the mass balance of Cr, Cu, Pb and Zn in the system. The changes in the chemical speciation of heavy metals after bioleaching experiments were also reported to observe the qualitative removal efficiencies of the mentioned metals. Therefore, the effect of sediment size fractions on bioleaching efficiency will be extensively analyzed.

1 Experimental

1.1 Sediment and particle size separation

The bulk sediment sample were taken from a point where the water is 15 m deep in Izmir Inner Bay where is located on the eastern zone in the Aegean Sea. It is known that Izmir Inner Bay sediments are highly contaminated with heavy metals and the sampling station in this study represents a typical polluted point in the inner bay. Particles larger than 2000 μm were removed from the solid mass. The bulk sediment was classified into three different size fractions by wet sieving. Fine (< 45 μm), medium (45–300 μm), and coarse (> 300 μm) sediments were obtained as subsamples of the bulk sample. Wet subsamples were dried overnight at 65°C in an oven, homogenized, and kept in plastic bags for the chemical analysis and bioleaching tests. For comparison, the bulk sample was also prepared in the same way to be used in bioleaching experiments.

1.2 General properties of sediment fractions

The chemical composition of the sediment grain fractions were detected by X-ray fluorescence and the primary minerals in the samples were identified by XRD analysis. The specific surface areas of the grain fractions were detected by multi point BET method. The organic matter contents were detected by ignition of the dried samples at 550°C for four hours.

Major element oxides concentrations in grain fractions were analyzed by X-ray fluorescence spectrometry by using an AMATEK-Spectro IQ II. The samples were powdered prior to the analyses. Mineralogical analysis on sediment size fractions were performed with a diffractometer (X’Pert ProX-ray, Phillips, The Netherlands) operated at 45 kV/40 mA by using copper anode material. The diffractograms were processed using Crystallographica Search Match software for the identification of the minerals.

For surface area detection, the samples were degassed overnight at 300°C in a vacuum oven prior to the analysis. Surface area analysis were carried out by the multi-point (7) BET method by using N₂ adsorption with a Quantochrome NovaWin 2.

1.3 Heavy metal analysis

Total heavy metal contents of the sediment subsamples were determined by using microwave acid digestion method with a closed vessel extraction system. A microwave digestion system (MicroPrep Q20, Questron Technologies, Canada) with four digestion vessels was used for the extraction. A digestion procedure previously developed by the authors (Guven and Akinci, 2011) was used for the extraction of metals from the sediments. According to the procedure, 0.1 g of samples were digested with a mixture of concentrated HNO₃ (3 mL), HF (1 mL), and HCl (0.8 mL). The heating protocol used was as follows: heat under 200 W for 8 min, then under 400 W for 6 min, finally 600 W for 2 min, and keep in the digester for another 10 min without applying any power. The accuracy of the method was tested by using the Estuarine sediment-1646A (NIST, 2003) as the standard reference material. The chemical partitioning of Cr, Cu, Pb and Zn were determined by following the Community Bureau of Reference Sequential Extraction Procedure which is a scheme developed by the European Commission for Standards, Measurement and Testing (Rauret et al., 1999). In this technique, metals are divided into 4 fractions including: exchangeable and acid soluble, reducible, bound to organics, and residual forms which indicate the binding properties of metals on sediment particles. The technique includes the applications of acetate acid (CH₃COOH), hydroxylamine hydrochloride (NH₂OH·HCl), hydrogen peroxide (H₂O₂), and ammonium acetate (CH₃COONH₄) solutions as the extracting agents, respectively. In this method, shaking, centrifugation and washing were applied.
to the sample for each step of the extraction procedure (Rauret et al., 1999; Dean, 2003).

1.4 Microorganisms

In the literature, generally mixed cultures are used for the bioleaching studies, mainly the mixtures of *T. thiooxidans*, *T. ferrooxidans* or *T. Thiooxidans*, *T. thioparus* (Chen and Lin, 2000, 2001; Gomez and Bosecker, 1999). This study was planned with the mixed culture to make it comparable with the previous studies conducted with bulk sediment samples.

The mixed cultures of *Acidithiobacillus ferrooxidans* (11477) and *Acidithiobacillus thiooxidans* (11478) obtained from DSMZ (German Collection of Microorganisms and Cell Cultures) were used. The recommended medium DSM 271 was used to cultivate both species. The media includes 2 g of (NH₄)₂SO₄, 0.5 g of K₂HPO₄, 0.5 g of MgSO₄·7H₂O, 0.1 g of KCl, 0.01 g of Ca(NO₃)₂, 8 g of FeSO₄·7H₂O, 10 g of elemental sulfur in 1000 mL of distilled water (DSMZ, 2004). The cultures were maintained in a shaking incubator at 170 rpm at 30°C. Plate count method on sterile petri plates was applied in order to determine the initial cell concentrations of the bacteria (Franson et al., 1992).

1.5 Bioleaching experiments

Before the bioleaching experiments, the cultures of thiobacilli were acclimated to the contaminated sediment subsamples. In the acclimation process, the mixed inoculum of 1% (V/V) of *A. ferrooxidans* and *A. thiooxidans* were transferred to 150 mL of water containing 2% (W/V) total solids of autoclaved sediment. Technically available elemental sulfur in yellow-cristal solid form with a density of 2.06 kg/L was used in the bioleaching medium. In consideration of the previously conducted experiments, optimum elemental sulfur dose detected as 0.5% (W/V), was added to each of the suspensions (Guven and Akinci, 2010). The acclimation flasks were prepared separately for bulk, fine, medium, and coarse sediment samples. Acclimation was terminated when the pH in the flasks was < 2 (Chen and Lin, 2001).

The bioleaching experiments were carried out in 1000 mL flasks including 250 mL reaction which contain the dilution medium necessary for the bacterial growth. The solid liquid ratio was kept constant with 5% (W/V) and the sulfur concentration was arranged to 0.5% (W/V). A 5% (V/V) actively mixed cultures of bacteria, obtained from the acclimation process, was added to the flasks, and the initial pH values were set to 4.0 ± 0.4 by using dilute H₂SO₄. The bioleaching experiments were conducted in shaking incubator at 170 rpm/min under 30°C. Aeration of the flasks was maintained by orbital shaking of the incubator. The process was monitored by periodic sampling and analysis of the sediment suspension for pH, oxidation-reduction potential (ORP), sulfate, and solubilized Cr, Cu, Pb, and Zn concentrations. The pH and ORP values were monitored every second day. For the redox potential, the Eh (V) values were calculated by adding the reference voltage (E_ref) value to the measured ORP values. Soluble metals and sulfate concentrations were measured once a week. In order to check for the mass balance during the bioleaching period, the concentrations of metals retained in the leached sediments were determined by removing solid samples from the flasks on day 18, 32, and 48 of the experiments. Approximately 0.2 g of solid samples were obtained in each sampling, and the above mentioned digestion procedure was applied to these solid sediment drawn from the suspensions. The chemical speciations of Cr, Cu, Pb and Zn in the leached sediments were monitored at the end of bioleaching period. The control sets without bacteria were conducted in parallel with the bioleaching experiments for fine, medium, coarse, and bulk samples.

For precision, all analyses were duplicated in the two parallel bioleaching experiments and the mean values of the parameters are reported.

1.6 Instruments

The pH values were measured by using a WTW pH 720 pH/ORP meter. A WTW Sentix Electrode (Germany) was used for the detection of ORP values.

Perkin Elmer Optima 4300 DV inductively coupled plasma-optical emission spectrometer (ICP-OES) was used for the determination of metals. The calibration of the instrument was conducted by using standard solutions (MERCK Spectroquant-1.19776.0100, 1.19806.0100, 1.19786.0100, 1.19780.0500) and the calibration curves were checked after the initial calibration for every 10 samples. For all the elements studied, the detection limits were fixed between 0.005 mg/L and 5 mg/L and seven points calibration curves were drawn. In the case of more than ± 10% deviation, the ICP OES was recalibrated. Cr and Cu were detected at the wavelength of 267.716 nm and 327.393 nm, where Pb and Zn were determined at 220.353 nm and 206.200 nm, respectively.

Sulfate ions in suspensions were analyzed by using Dionex IC-3000 ion chromatography system.

2 Results and discussion

2.1 Properties of sediments

The features of the size fraction sediments are shown in Table 1. The chemical composition of different sediment grain fractions were similar and important concentration distinctions could not be seen. The obtained values were compared with the levels measured in global subducted sediments (Plank and Langmuir, 1998) and Bremen Harbor sediment (Hamer and Karius, 2002). Besides some minor dissimilarities, Al₂O₃ levels were higher and SiO₂ levels were lower than the values of the reported sediments. The elemental levels of the cations in their oxides were also cal-
The specific surface areas (SSAs) of the sediment grain fractions were determined by BET method. The SSAs of the medium and coarse fractions may not seem adequately low when compared to fine sediment. This situation depends on the disaggregation of the material prior to the sieving procedure.

Identified minerals in sediment grain fractions were well matched with their chemical compositions. Quartz, calcite, feldspar, mica and clay minerals (chlorite or kaolinite, and smectite) were detected in all fractions, where dolomite was additionally found in medium fraction.

The organic matter content of the fine, medium, and coarse sediments were detected as 15.03%, 18.36%, and 14.91%. Kontas (2006) reported the maximum organic matter content as 10.1% in Izmir middle-inner Bay which means the higher values in this study indicate a serious organic pollution in the sampling area.

2.2 Heavy metals and their chemical speciations in the sediment

The bulk sample and fine, medium, and coarse sediments were analyzed for their total Cr, Cu, Pb, and Zn concentrations and the results are shown in Table 2. The weighted average concentrations of the metals (cumulative) were calculated by using the mass distribution of the size fractions. The cumulative and the bulk concentration values were compatible with each other. The deviations changed between 3% and 12% for the metals studied. Aksu et al. (1998) presented the elemental background concentrations of the selected metals. The background level for each element was calculated as the mean value of that element in 50 subsurface samples collected from the Bay. In this study, Cr (581 ± 48 mg/kg) and Zn (574 ± 26 mg/kg) exhibit the higher concentrations in the bulk sample. These values are much higher than the natural background values (Cr 175 mg/kg and Zn 65 mg/kg) detected for Izmir Bay. The Cu (108 ± 12 mg/kg) and Pb (138 ± 12 mg/kg) in bulk samples were also detected above the reported concentrations (17 mg/kg for Cu; 85 mg/kg for Pb) for...
Izmir Bay (Aksu et al., 1998). The high concentrations arise from the past discharges of metals to the inner Bay from the metal and manufacturing industries (Guven and Akinci, 2008).

The chemical speciation of the metals in the fine, medium, coarse and bulk sample is illustrated in Fig. 1. For all the samples, Cr was found mostly in the residual fraction with a portion over 50%. Cu was mostly accumulated in the organic form except the fine particles, where it was detected over 44% in the residual fraction. On the other hand, Cu in coarse sediment was highly found in the exchangeable and acid soluble fraction with 36.6%, which means under acidic conditions, significant Cu can pass to water from the coarse particles. Pb accumulation showed distinctions for different particle sizes of the sediments, and the accumulation in reducible fraction was the highest of all the metals studied (19.1%–41.9%). In contrast to the other metals, Zn was found evenly in all particle sizes of the sediments, mostly in the exchangeable and acid soluble form (25.7%–34.3%) when compared with the other metals accumulated in this fraction.

### 2.3 Initial bacteria concentration and acidification during bioleaching

The cell concentrations of the mixed culture were counted in all the acclimation flasks, and the observed concentrations were around $7 \times 10^4$ and $3 \times 10^5$ CFU/mL, for *A. ferrooxidans* and *A. thiooxidans*, respectively. These values were found to be comparable with the previous bioleaching studies, and they were sufficient for the bioleaching startup (Chen and Lin, 2001; Akinci and Guven, 2011).

Rapid pH decreases in flasks were observed within the first ten days of the experiments but the decreases slowed afterwards. This depends on the activity of the mixed culture. In the first days of the experiments, the pH decrease in the coarse sediment were slightly slower when compared with the other sediments. However, pH values in all the bioleaching flasks were measured between 1.5 and 1.7 at the end of 48 days (Fig. 2). The results obtained from the control flasks were detected similar to each other. Since the most aggressive pH level and the highest metal solubilizations were observed with the fine fraction, the results of control-fine sediments are presented in the graphs. The pH in the control test increased in the first ten days and fluctuated between 5 and 6 until the end of the experiments.

The Eh values in the flasks increased rapidly with the decreasing pH and the final Eh values reached 0.60–0.62 V. After the first four days of bioleaching tests, the Eh values were above 0.52 V (coarse fraction, 4th day) and the highest pH level was 3.2 (coarse fraction, 4th day) in the flasks. The Eh in the control flask varied around 0.39

### Table 2  Total heavy metals in the bulk, fine, medium size, and coarse sediment samples

<table>
<thead>
<tr>
<th>Sediment sample</th>
<th>Cr (mg/kg)</th>
<th>Cu (mg/kg)</th>
<th>Pb (mg/kg)</th>
<th>Zn (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fine (mg/kg)</td>
<td>612 ± 36</td>
<td>326 ± 18</td>
<td>128 ± 10</td>
<td>729 ± 33</td>
</tr>
<tr>
<td>Medium (mg/kg)</td>
<td>569 ± 41</td>
<td>108 ± 12</td>
<td>138 ± 12</td>
<td>574 ± 26</td>
</tr>
<tr>
<td>Coarse (mg/kg)</td>
<td>581 ± 48</td>
<td>36 ± 19</td>
<td>8 ± 12</td>
<td>10 ± 11</td>
</tr>
<tr>
<td>Buln (mg/kg)</td>
<td>608 ± 23</td>
<td>28 ± 10</td>
<td>1 ± 1</td>
<td>2 ± 1</td>
</tr>
</tbody>
</table>

Data are expressed as mean ± standard deviation ($n = 4$).
V (Fig. 2). Since the solid content (5%, W/V) and sulfur content (0.5%, W/V) in the flasks were kept constant, no significant differences depending on the buffering capacity or sulfur oxidation were observed in pH or Eh variations.

2.4 Sulfate production

Figure 3 presents the changes in sulfate production which is a significant indicator of bioleaching process. The normalized sulfate concentrations in suspension were monitored. The sulfate productions were found to be in correlation with the pH decreases, and finally reached to 13959, 14125, 15004, and 11728 mg/L for bulk, fine, medium, and coarse sediments. According to these findings it was calculated that 98%, 92%, and 91% of S$^0$ were oxidized to SO$_4^{2-}$ for medium, fine and bulk sediment samples in the bioleaching flasks. On the other hand, 77% of the S$^0$ was converted to SO$_4^{2-}$ for the coarse sediment. The relatively low conversion of sulfur in the coarse sediment system may attribute to either the Eh–pH changes in this system, or the structure of the dominant minerals in this fraction. For the first case, the utilization of elemental S in the process and the leaching of metals from the sediment occur according to the following reactions:

$$S^0 + H_2O + \frac{3}{2}O_2 \xrightarrow{Thiobacilli} H_2SO_4$$

(1)

$$H_2SO_4 + \text{(Sediment-Metal)} \rightarrow \text{(Sediment-2H$^+$)+Metal-SO_4}$$

(2)

The initial sulfur and sediment amounts in the systems were 1250 and 12500 mg, respectively, and the heavy metal concentrations in the sediments are given in Table 3. According to the Reactions (1) and (2), the added sulfur dose is much higher than the necessary sulfur amount for the bioleaching of whole studied heavy metal mass in the sediments. In the systems operated with fine and medium size sediments, some other metals might be bioleached due to the lower pH values at the beginning. However the same process might not happen in the coarse sediment system. In the first 15 days of the experiments, it is noted that, the pH in the coarse sediment system was between 4 and 2.8. This range may lead the formation of some certain insoluble metal sulfides such as CuS, HgS, PbS, and CdS due to the Eh-pH predominance diagrams (Hermann and Neumann-Mahlkau, 1985; Pecora, 1970). The dissolutions of these metal sulfides are only available either in the presence of non-oxidizing acids, such as HCl, or in high temperature concentrated H$_2$SO$_4$ (Brescia et al., 1980). Therefore some part of the available S$^0$ could be used in the formation of MeS compounds in the coarse sediment system, and could not be converted to H$_2$SO$_4$. On the other hand, these insoluble metal sulfides may precipitate on the residual fraction of the coarse particles.

The bacterial sulfur oxidation systems do not only depend on the presence of available substrate, but also the attachment process itself (Vandevivere and Kirchman, 1993). During the process, the bacteria do not attach the whole mineral surface but prefer specific sites of crystal imperfections (Bosecker, 1997; Rohwerder and Sand, 2007). According to the wet sieving procedure applied in this study, the sand sized particles are only dominated in the coarse sediment which serves larger quartz particles in this system. It is known that quartz has trigonal or hexagonal smooth crystal surfaces which do not supply

<table>
<thead>
<tr>
<th>Table 3</th>
<th>Comparison of metals remained in the sediments after bioleaching with the limit values given in Ecotox Thresholds</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Bulk</td>
</tr>
<tr>
<td>Cr (mg/kg)</td>
<td>45</td>
</tr>
<tr>
<td>Cu (mg/kg)</td>
<td>35</td>
</tr>
<tr>
<td>Pb (mg/kg)</td>
<td>46</td>
</tr>
<tr>
<td>Zn (mg/kg)</td>
<td>22</td>
</tr>
</tbody>
</table>
sufficient interaction area for the bacteria, resulting as the lower sulfur oxidation in the system.

2.5 Metal solubilization during bioleaching

Figure 4 presents the metal solubilization ratios and the amounts of heavy metals remaining in the sediments on the day 18, 32, and 48 of bioleaching period. The results show variations depending on sediment size and the metals studied. Cr solubilization started significantly on the day 11 for fine particles with 26% and slowed down on day 39. Remarkable releases from medium and coarse particles were observed on day 18 and 32, with 21% and 31%, respectively. At the end of the experiments, Cr solubilization efficiencies were noted as 58%, 68%, 53%, and 43% for bulk sample, fine, medium and coarse sediments, respectively. According to the Eh-pH predominance diagram of Cr (Reeder and Schoonen, 2006), this heavy metal can only be in ionic (Cr$^{3+}$) form in the aqueous solutions under the above mentioned bioleaching circumstances. The pH values were between 1.5 and 1.7 in the bioleaching flasks, which mean extremely acidic conditions are required for Cr for higher solubilization.

According to the Eh-pH predominance diagram, total copper in the solution can only be in the form of Cu$^{2+}$ (Hermann and Neumann-Mahlkau, 1985). Rapid solubilization of Cu started on the 4th day of the bioleaching period for fine, medium, and coarse particles with 46%, 33%, and 31%, and the reaction rate slowed down on day 11. Finally, the solubilization efficiencies of Cu depending on increasing particle size were determined as 88%, 68%, and 62%, respectively. In the bulk sample, Cu was solubilized to the ratio of 73%, finally.

Noticeable bioleaching of Pb from fine particles was observed on day 4 with 11%, and increased gradually till day 25 (66%), and then the solubilization rate slowed down. Significant Pb releases from medium particles started on day 11 (10%), and from coarse particles on day 25 (12%). Final lead removals were detected as 58%, 72%, 55%, and 25% for bulk, fine, medium and coarse particles. The Eh-pH predominance diagram of Pb defines that, it will be in the form of Pb$^{2+}$ when the pH is below 1.6 while the formation of PbSO$_4$ is observed between pH 1.6 and 3.2 (Hermann and Neumann-Mahlkau, 1985). As illustrated in Fig. 2, the lowest pH level in the flasks of medium and coarse fractions are 1.67 (day 39) and 1.71 (day 43), respectively, which allows the occurrence of PbSO$_4$ in the solution. Since the solubility of PbSO$_4$ is low (Sillen and Marten, 1964), it may settle down and result in the lower Pb removals in medium and coarse fractions.

In contrast to Pb, Zn salts and compounds are highly soluble in water and will be in ionic form (Zn$^{2+}$) under acidic conditions (Hermann and Neumann-Mahlkau,
Zn removal from sediments started on day 4 with 22%, 15%, and 15% for fine, medium, and coarse particles. Thereafter, the process continued regularly from different size sediments. At the end of the experimental period, comparatively higher bioleaching efficiencies of Zn were determined as 81%, 91%, 76%, and 61% for bulk, fine, medium, and coarse sediments, respectively.

For all the metals studied, solubilization efficiencies increased with the decreasing particle size of the sediment samples. Metals were solubilized from fine particles in higher ratios, while the lowest efficiencies were determined with coarse particles. This situation depends on the relatively larger surface area of fine sediments (17.596 m²/g) provided by the unit mass of the smaller particles while the specific surface area of the coarse sediment was detected as 16.119 m²/g (Table 1). In other words, larger interaction area of particles resulted in better solubilization achieved by the microbial activity. It was also observed that the solubilization of the metals in fine particles started earlier than the others. The bigger particles make reactions slower because of the decreasing surface area of contact. Therefore, smaller particles also increase the speed of bioleaching as well as the process performance. Another point is fine fraction exhibits relatively lower trace metal concentrations which means initial metal content in the sediment may also influence the bioleaching efficiency.

The efficiency of metal solubilization from contaminated sediment particles in the decreasing order is: Zn > Cu > Cr > Pb. Higher solubilization of Zn depends on its mobility and formation of highly soluble compounds. The lower efficiency of Pb in the process was attributed to the low solubility of PbSO₄. The minor distinctions between the chemical and mineralogical compositions of the grain fractions show that the effect of these features on bioleaching efficiencies can be neglected in this study. The mass balance was checked between the metals solubilized in suspension and remained in the sediment and the results were compatible with each other (Fig. 4). Deviations between 2% and 13% were observed due to the heterogeneity of metal distribution in the sediment sample.

The weighted average of the solubilization efficiencies (WASE) in fractions are determined by considering the contribution of each sediment size fraction to individual metal solubilization. WASE is calculated by considering the Cr, Cu, Pb and Zn solubilization efficiency of each size fraction, and its % distribution in the bulk mass. The WASE of Cr, Cu, Pb and Zn were determined as 59%, 70%, 60%, and 79%, respectively. For Cr and Cu, the maximum contributions to metal solubilization were achieved with the fine sediments (31.5% for Cr and 34.5% for Pb) depending on the high mass distribution and higher solubilization efficiencies of the fine particles. On the other hand, medium sediment consist of the maximum contribution of Cu (29.8%) and Zn (41.1%) to the total solubilization values due to the higher initial concentrations of Cu (113 ± 9 mg/kg) and Zn (739 ± 33 mg/kg). When the WASE are compared with the solubilization efficiencies in the bulk sample, the values are found to be satisfactorily close to each other. The general sediment size characteristics of the sediment samples show that fine and medium particles contribute to the bulk sediment with an average ratio of 90%. Even though the coarse particles have considerable concentrations of heavy metals, the load of metals they carry is lower than fine and medium size particles. Therefore, overall treatment efficiency allows lower removal of metals from the coarse sediment sample.

The data obtained during this study largely meet the expectations. Especially, for the fine sediment samples, the solubilization efficiencies were found to be remarkably higher than previously completed bioleaching studies conducted with contaminated bulk sediments (Chen and Lin, 2000, 2001, 2004; Lombardi and Garcia, 2002; Löser et al., 2007). Satisfactory solubilization ratios obtained here, shows considerable performance of the bioleaching process for the selected metals in this study.

In Turkey, there are no legal obligations related to the sediment quality criteria presenting the limit levels of organic and inorganic contaminants in aquatic sediments. Therefore, the metals remained in the sediments after bioleaching were compared with the Ecotox Thresholds published by EPA’s Office of Solid Waste and Emergency Response (US EPA, 1996). As can be seen from Table 3, Cr (29–56 mg/kg) and Zn (7–36 mg/kg) concentrations remained in the sediment were much lower than the Ecotox Thresholds values (81 mg/kg for Cr and 150 mg/kg for Zn) for all the samples. Cu remained in fine fraction (17 mg/kg) were also lower than the Ecotox Thresholds value (34 mg/kg). Cu in the medium sediment slightly exceeded the limit value with 35 mg/kg. For the case of Pb, only coarse fraction was found to contain higher Pb concentration (73 mg/kg) than the reported Ecotox Thresholds concentration (47 mg/kg) while the other size fractions included Pb lower than the limits. In consequence, bioleaching with the size fractions will be a useful remediation strategy for some selected metals. Especially for the fine sediments, satisfactory removal efficiencies were obtained which correspond to the international standard defined in this study.

2.6 Changes in chemical speciations

Figure 5 presents the fractionation of Cr, Cu, Pb and Zn in bulk, fine, medium, and coarse sediment before and after bioleaching. The changes in the chemical forms of metals are presented individually since each metal was found in different concentrations in the bulk, fine, medium, and coarse particles. Cr was mainly present in the residual fraction before bioleaching, and showed complex fractionation behavior after bioleaching. It was solubilized mostly from the reducible and organic fraction for all grain sizes. In addition, increasing Cr concentrations in the exchangeable and acid soluble fraction were observed after
bioleaching. The losses of Cr in residual fraction decreased due to the increasing sediment size.

After the bioleaching period, Cu was considerably removed from all fractions of sediment except the residual fraction. There was practically no Cu found in the reducible fraction after bioleaching and the organic fraction released most of the Cu load from the sediment. However, the concentrations of Cu in the residual fraction increased for medium (from 15 to 29 mg/kg) and coarse (from 25 to 68 mg/kg) sediment at the end of the experimental period.

Pb releases were mostly observed in the reducible and organic fractions of the sediments. The exchangeable and acid soluble fraction remained almost unchanged for Pb, only slightly increasing concentrations were observed for fine (from 6 to 9 mg/kg) and medium (from 5 to 6 mg/kg) sediments after bioleaching. Similar to Cu, Pb in residual form increased for medium (from 29 to 43 mg/kg) and coarse (from 57 to 86 mg/kg) sediment after bioleaching.

For every size of sediments, Zn was distributed evenly in each bounding fraction, and different than other metals, it was present at higher concentrations in the exchangeable and acid soluble form. After bioleaching, Zn was significantly removed from all fractions of bulk, fine, and medium sediments. Only higher Zn concentrations were detected in the residual fraction for coarse sediments (Fig. 5).

Significant amounts of concentration decreases were observed from all fractions in bulk and fine sediment. Besides, for all the metals studied, the content ratio of exchangeable and acid soluble fraction in the sediment increased after bioleaching. This is in agreement with the study conducted by Chen et al. (2005). On the other side, a remarkable occasion was observed for medium and coarse sediments as the content ratio of the metals in residual fraction increased significantly after bioleaching process (Fig. 5). This is compatible with the literature (Kumar and Nagendran, 2009). This situation may be due to the precipitation of insoluble metal sulfides on the residual matrix of the coarse sediment depending on the relatively higher pH values of the coarse sediment system. This means, although relatively high metal concentrations are left in medium and coarse samples, the remnant metals are bound to the stable fraction from which they are not expected to be released under normal conditions.

3 Conclusions

Bioleaching was found to be efficient for removal of Cr, Cu, Pb, and Zn from every size fraction of sediments, where the experiments with the finest sediments resulted in the highest metal removal efficiencies. Finer particles also increased the speed of bioleaching, as well as process performance. This is related with the larger surface area of unit amount of particles with lower diameters. At the end of the experimental period, Cr, Cu, Pb and Zn were solubilized to the ratios of 68%, 88%, 72%, and 91% from the fine particles. The metal contents remained in the sediment were remarkably lower than the reported limit values of Ecotox Thresholds, except for the Cu and Pb in coarse sediment. The effect of chemical and mineralogical distinctions of the samples, on bioleaching efficiency could be ignored, since the differences were minimal. After the bioleaching process, metal releases were mostly observed from reducible and organic fraction independent from grain size, where coarse particles could not release Cr, Cu, Pb and Zn from the residual fraction. Moreover, higher accumulations in the crystal matrix were observed for Cu, Pb, and Zn after bioleaching, due to the formation of insoluble metal sulfides which might precipitate in the residual fraction of coarse sediment. This shows that, even the metals cannot be solubilized from the coarse particles sufficiently; they are trapped in the residual
fraction from which they are not released under natural conditions. Bioleaching with the size fractions will be a useful remediation strategy, especially for the fine particles of the selected metals, since it significantly reduces the metal concentrations that may be harmful to ecological receptors. For the metals studied here, bioleaching can be applied as a proper treatment method, especially for the sediment with a grain size structure dominated by clay, silt, and fine sand particles.

Acknowledgments

This work was supported by the Research Fund of Dokuz Eylul University (No. 03.KB.FEN.018) and by the Turkish State Planning Organization (No. DPT.2003K120360).

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