

Trace element exposure in the environment from MSW landfill leachate sediments measured by a sequential extraction technique

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Abstract

Dredged leachate sediments from eight MSW landfills were dried and homogenized, and metals sequentially extracted. The concentrations of cadmium (Cd), copper (Cu), chromium (Cr), iron (Fe), lead (Pb), manganese (Mn), nickel (Ni) and zinc (Zn) were found to be similar to those reported for sewage sludge, and generally below the EU limits for use of sludge on agricultural land. Sequential extraction of the samples showed that the largest fractions of the heavy metals were associated with Fe- or Mn-oxides. Cr and Cu were the only metals investigated that were associated with the organic matter in the sediments to any significant extent (2–10% of Cr and 10–28% of Cu). Since the largest fractions of the metals studied were bound to Fe-oxides, and thus had a low mobility, these metals will generally have a low bioavailability under aerobic conditions when present in leachate sediments. This is most likely also valid for particulate matter suspended in MSW landfill leachate and released to the environment, since this is the source of the sediments.

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1. Introduction

The levels of hazardous metals detected in municipal solid waste (MSW) (e.g. [1,2]), suggest that continued attention to these metals after deposition of such wastes is necessary. The retention of metals in MSW landfills is high [2,3]. Long-term discharge of low concentrations of metals from landfills may, however, cause environmental problems in the future. The heavy metals present in landfill leachate have been shown to be to a large degree bound in or onto particulate matter [4,5]. This binding strongly reduces the mobility and bioavailability of the metals, compared to free metal species. The prevalence of metals bound in or onto particles simplifies leachate treatment, since mere separation of the solids from the leachate (e.g. in settling tanks) will remove a large portion of the elements of concern [6].

To determine the fate of hazardous metals contained within the solid matter of leachate, it is important to identify the metal forms present and assess their mobility. This can enable assessment of the likelihood of dissolution of metals from sediments collected in leachate treatment plants, or from solids discharged into an ecosystem. Any subsequent dissolution would adversely affect the apparent high efficiency of treatment plants, and also increase the mobility and bioavailability of the metals once discharged into the environment.

The sediments intentionally collected in sedimentation tanks or unintentionally in the leachate discharge system need to be removed from time to time. It is then a question of how these sediment masses should be handled. At some landfills these sediment masses are handled as hazardous waste, while at other locations the masses are deposited onto the landfills. Except for studies reporting on sediments in stream systems being influenced by particulate matter discharged from a landfill [7], there are few published studies into metal concentrations in isolated leachate sediments, and it is thus difficult to give any clear advice to the landfill community on how to handle the sediment masses. Determination of total metal concentration to ascertain whether the masses are hazardous waste or not, can

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be done by a simple acid digestion of sediments in a microwave oven.

Both the chemical forms of metals and their mobility can be studied by the use of sequential extraction methods. The sequential extraction technique was initially proposed by Tessier et al. [8], and has been adopted by many research groups [9–11]. By performing sequential extraction (over six steps) on leachate sediments collected from various landfills, knowledge of the binding and mobility of heavy metals can be obtained. In a well-constructed landfill site there should be few other sources of particulate matter generating leachate sediments than suspended matter washed out from the landfill by the leachate. Assuming that particulate matter suspended in leachate has the same composition as the sediments, the study will also show the binding and mobility of heavy metals in this suspended matter which is released to the environment. Knowledge of this is important for the assessment of the mobility and potential eco-toxicological impact of heavy metals discharged from landfills.

2. Materials and methods

2.1. Sampling and treatment

Leachate sediments were sampled from eight sanitary landfills situated on the west coast of Norway (Fig. 1). All the landfills had been in use for 15 years or more and only MSW had been deposited. All landfills were still active at the time of the sampling. The characteristics of the leachate from the landfills prior

or during the sediment sampling are shown in Table 1. All the metal results are found in acid-digested leachate samples that included suspended particulate matter.

Where sedimentation tanks were present in the system, the sediments were sampled from the bottom of these tanks, immediately prior to the discharge pipe. At the other sites, samples were collected from the leachate volume measurement points, where sediments had collected, or, if necessary, sediment-rich leachate collected from the bottom of the sedimentation tanks was centrifuged (Beckman J2-21m centrifuge, 15,000 rpm for 10 min). After direct collection, or separation by centrifugation, the samples were kept frozen until further processing. Where sediments could be sampled directly, approximately 1.5–2 kg of sediments (wet weight) was sampled, and further processed. Where the sediment was collected from the leachate, approximately 100 g of sediment was separated from around 40 L of sediment-rich leachate.

Samples were dried at 40 °C, and subsequently sieved to 2 mm. More than 90% of the dry sample weight had a grain size <2 mm for all the samples. The sieved samples were carefully homogenized by stirring by a glass rod, and kept in a desiccator until sequential extraction or digestion for total analysis.

2.2. Sequential extraction

Sequential extraction involves the treatment of a solid material with a series of chemical reagents with increasing dissolution and displacement power, in order to extract met-

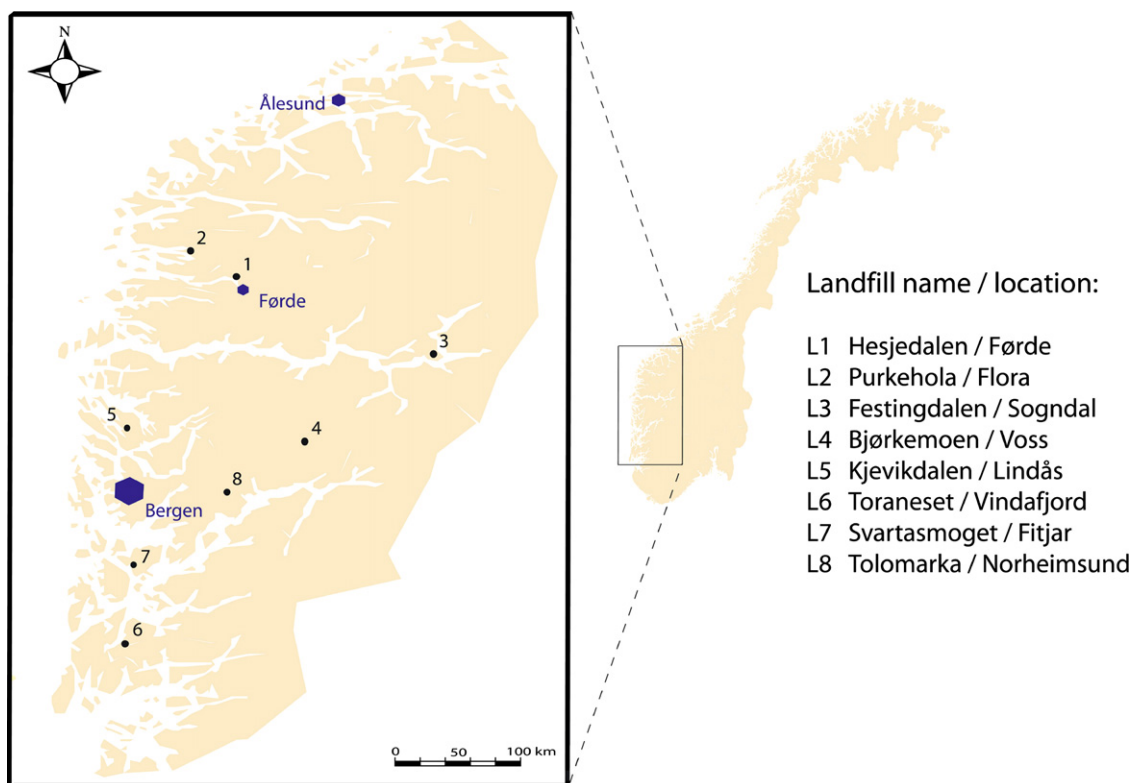


Fig. 1. Geographical locations of the landfills where the leachate sediments were investigated.

Table 1

Concentration of various parameters in leachate from the eight investigated landfills prior (2003–2004) or during (2004–2005) the sampling of the leachate sediments

Parameter	Landfill							
	L1 (n=8)	L2 (n=8)	L3 (n=8)	L4 (n=24)	L5 (n=24)	L6 (n=12)	L7 (n=24)	L8 (n=24)
pH	7.27 ± 0.16	7.15 ± 0.45	6.55 ± 0.62	7.29 ± 0.14	6.83 ± 0.13	7.41 ± 0.70	6.76 ± 0.12	6.59 ± 0.16
Conductivity (mS m ⁻¹)	660 ± 150	110 ± 21	390 ± 340	540 ± 77	300 ± 79	290 ± 94	220 ± 73	130 ± 20
Total N (mg L ⁻¹)	300 ± 66	42 ± 12	220 ± 180	350 ± 63	120 ± 46	160 ± 72	130 ± 52	45 ± 8.0
Total P (mg L ⁻¹)	3.6 ± 1.7	0.4 ± 0.2	6.9 ± 7.7	1.5 ± 0.96	0.82 ± 0.26	–	0.76 ± 0.26	400 ± 76
COD (mg L ⁻¹)	730 ± 190	170 ± 200	1600 ± 840	1900 ± 860	290 ± 110	–	290 ± 100	180 ± 180
Cl ⁻ (mg L ⁻¹)	–	370 ± 370	–	510 ± 130	450 ± 120	–	140 ± 52	53 ± 17
Fe (mg L ⁻¹)	22 ± 35	15 ± 9.6	35 ± 34	110 ± 40	24 ± 13	10 ± 3.9	20 ± 8	59 ± 30
Zn (µg L ⁻¹)	150 ± 110	220 ± 190	550 ± 630	240 ± 130	52 ± 22	320 ± 160	110 ± 51	50 ± 24
Cr (µg L ⁻¹)	63 ± 42	7.9 ± 4.7	44 ± 28	58 ± 19	30 ± 15	36 ± 13	31 ± 11	12 ± 2.1
Cu (µg L ⁻¹)	31 ± 18	8.4 ± 6.3	190 ± 310	16 ± 6.9	11 ± 11	24 ± 11	21 ± 20	5.3 ± 4.6
Pb (µg L ⁻¹)	11 ± 10	3.4 ± 1.8	–	4.0 ± 3.2	3.1 ± 3.1	7.1 ± 5.3	4.3 ± 2.9	1.5 ± 0.7
Cd (µg L ⁻¹)	0.44 ± 0.42	0.16 ± 0.12	0.32 ± 0.32	0.15 ± 0.11	0.09 ± 0.06	1.4 ± 1.2	0.65 ± 2.5	0.09 ± 0.05
Ni (µg L ⁻¹)	–	–	–	31 ± 10	13 ± 5.1	20 ± 5.4	12 ± 4.4	8.5 ± 1.0

als according to the chemical activity and property of their binding to the solid. Depending upon the reagents chosen, the method can assess the level of water-soluble and exchangeable metals, as well as metals contained in easily reducible or oxidizable compounds. Thus, a sequential extraction scheme will operationally distinguish between reversibly and irreversibly (or slowly reversibly) associated metal fractions in the sediments.

In this experiment, sequential extraction was performed broadly as described by Oughton et al. [10], but modified according to the work done by Gómez Ariza et al. [12]. The leachate sediments had a relatively high iron content (4.1–24%), and Gómez Ariza et al. reported that it was necessary to perform several repetitions (up to seven) of extraction step 4 (0.4 M

NH₂OH·HCl in 25% (v/v) acetic acid) in order to achieve complete extraction of reducible compounds from iron-rich sediments. The extraction procedure was thus modified by increasing the concentration of NH₂OH·HCl from 0.04 to 0.4 M in step 4, and by performing this step seven times instead of once. It is known that concentrations of NH₂OH·HCl even as high as 1 M will not dissolve metal bound to organic matter or silicates [13]. The entire procedure is shown schematically in Fig. 2.

The extractions were performed using Nalgene (40 ml PPCO Oak Ridge) centrifuge tubes, with 20 ml extractant added in each step, except in step 5, where 15 ml of H₂O₂ and later 5 ml NH₄Ac solution was added. Due to the high organic matter content in the sediments it was necessary to perform extraction step 5

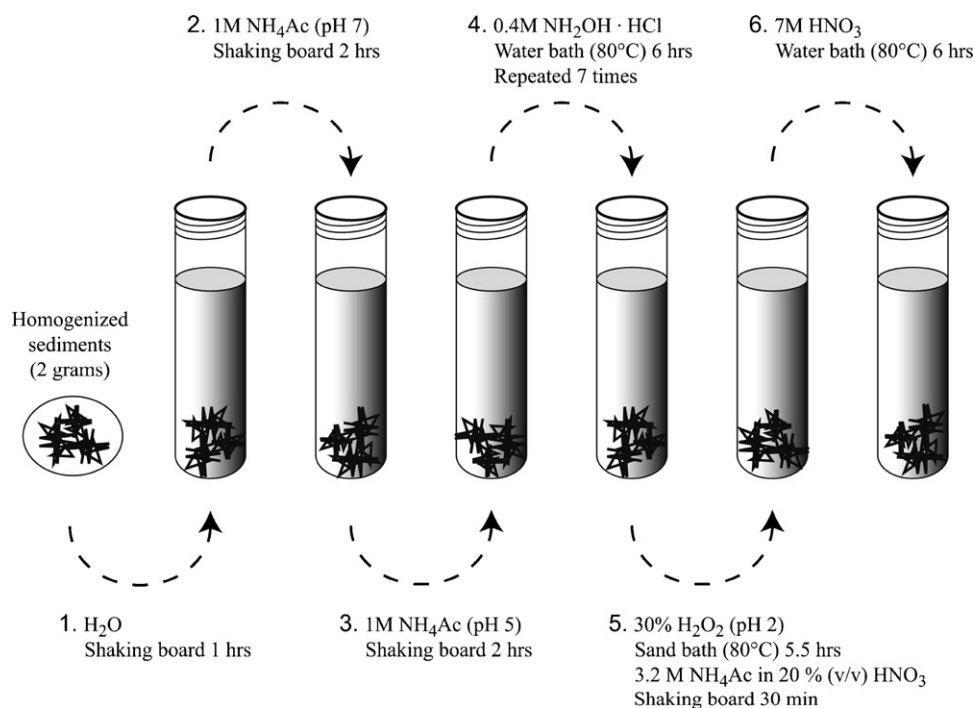


Fig. 2. Schematic view of the sequential extraction scheme used on the dried and homogenized landfill leachate sediments.

very carefully, with gradual addition of H₂O₂. The sediments were washed with de-ionized water between each extraction step. Three parallel samples consisting of 2 g of the homogenized sediments were extracted in order to get representative results.

All equipment used for the experiment was acid washed prior to use. Reagents of analytical grade or better were used throughout, and all water was de-ionized (>18 MΩ cm⁻¹).

2.3. Analysis

The determination of S, Fe, Mn, Zn, Cu and the higher concentrations of Ni ($\geq 50 \mu\text{g/L}$) was performed on a Perkin-Elmer Optima DV 5300 ICP-OES, after calibration with standards which were matrix matched to each extractant solution. The determination of Cd, Pb, Cr and the lower levels of Ni (<50 $\mu\text{g/L}$) was done with a Perkin-Elmer SIMAA 6000 ET-AAS with Zeeman background correction. Accuracy of the analysis of the extractant solutions were done by determining the recovery of spiked metals in the various extractant solutions. The recovery was within $\pm 5\%$ of the true value.

The total (acid extractable) metal contents in leachate sediments were determined by microwave acid digestion of sediment samples. Accurately weighed samples of about 0.25 g were digested with 4 ml concentrated HNO₃ (65%, w/w) and 1 ml H₂O₂ (30%, v/v), in a Perkin-Elmer Anton Paar Multiwave microwave oven. A digestion program with an upper temperature limit at 230 °C was chosen to achieve complete digestion of the sediment. The digested samples were diluted to 25 ml with de-ionized water prior to analysis, as described previously. Accuracy of the total analysis was checked by concurrent analysis of the certified reference material River sediment (LGC6187, Laboratory of the Government Chemist, UK). All analysed metals were determined with results within 5% of the certified value of the reference material, except Cr and Zn, which were within 10% of the certified value.

Loss on ignition was determined by dry combustion of 2 g of homogenized sample at 550 °C for 12 h. X-ray examinations were done on a Philips PW 1370 X-ray diffractometer.

3. Results and discussion

3.1. Composition of sediments

Total metal, sulphur and organic matter contents in the samples are given in Table 2. The sediments contained a considerable proportion of organic matter. The most distinct feature of the sediments, however, was the relatively high iron content. All the sediments were red in colour in their oxidized form. X-ray diffraction investigations of the sediments gave the diffractograms shown in Fig. 3. From the X-ray diffraction investigations the iron compounds in the sediments could be identified as Goethite ($\alpha\text{-FeOOH}$). This identification was confirmed by treating the sediments by dithionite [14] and comparing diffractograms before and after treatment, and also corresponds well with the findings of goethite in leachate-polluted stream sediment by Ettler et al. [7]. Assuming that all the iron in the sediments was in the Fe(OOH) form, iron oxide accounts for 6.5–38% of the total mass of the dry sediments.

Apart from the levels of Zn, which were relatively high in landfills 2 and 4, the heavy metal contents of the sediments were moderate; lower than, or at the same levels, as those reported for sewage sludge [15–17]. According to the EU directive on the use of sewage sludge on agricultural lands [18], the concentrations of heavy metals in the sediments (except for Zn in landfills 2 and 4) were lower than the upper legal limits for use. In most cases there seems to be no reason to handle the sediments as hazardous waste.

When comparing concentrations of heavy metals in the leachates (Table 1) with the sediments, it could be seen that the sediment/leachate concentration ratios of Zn, Cu, Pb and Cd were higher than those of Ni and Cr. From the fractionation study of leachate done by Øygaard et al. [5], with leachate sampled from many of the same landfills as in this study, the results can be explained by Ni and Cr being present to a large degree in matter <0.45 μm in the leachate, whereas Zn, Cu and Pb are present to a large degree in particulate and colloidal matter >0.45 μm , thus accelerating their deposition and pre-concentration in the sediments. It would therefore be expected to find a lower degree

Table 2
Total (acid extractable) metal, sulphur, organic matter and pH in dried and homogenized samples of landfill leachate sediments ($n=3$)

Landfill	L1	L2	L3	L4	L5	L6	L7	L8	EU limit ^a
pH	6.8	6.8	7.1	6.2	7.4	6.8	7.2	7.2	–
Loss on ignition (%)	48	43	19	29	22	6.7	11	21	–
S (%)	5.5	0.42	0.51	11	0.24	0.84	1.1	1.1	–
Fe (%)	10	24	5.1	18	19	4.1	5.7	15	–
Mn (mg kg ⁻¹)	1500	1100	250	570	330	530	370	1900	–
Cd (mg kg ⁻¹)	1.1	1.3	0.55	1.1	0.23	4.8	0.35	1.5	20–40
Pb (mg kg ⁻¹)	49	96	18	13	47	22	37	30	750–1200
Cr (mg kg ⁻¹)	91	57	31	56	70	15	50	77	1000–1750
Ni (mg kg ⁻¹)	26	48	47	64	15	9.2	15	37	300–400
Cu (mg kg ⁻¹)	220	130	67	34	300	19	130	83	1000–1750
Zn (mg kg ⁻¹)	1300	2800	410	4400	490	370	500	1200	2500–4000
As (mg kg ⁻¹)	98	15	7.1	46	15	8.1	12	14	–

^a EU concentration limit for the use of sewage sludge in agriculture (EU directive 86/278/EEC (European Union, 1986)); concentration range depending on soil pH.

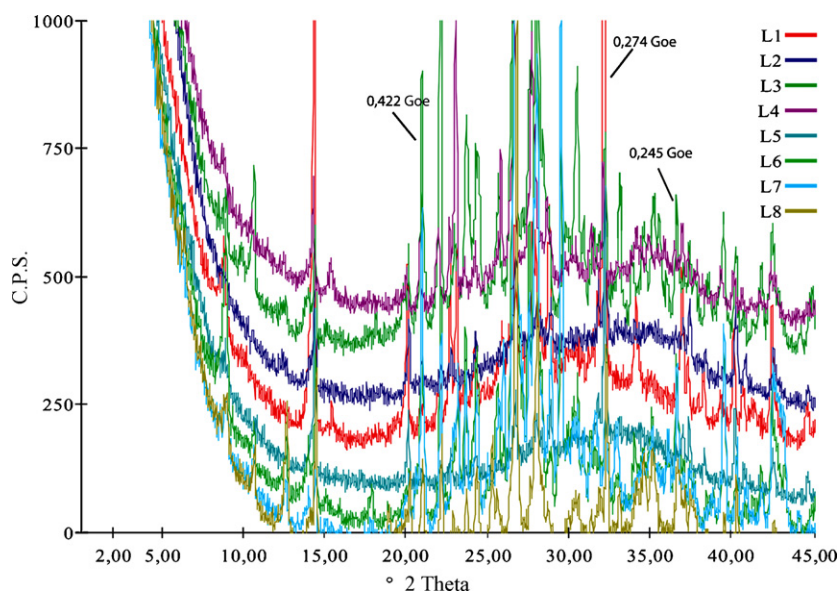


Fig. 3. XRD-diffractograms of the leachate sediment from the eight landfills. The characteristic peaks for Goethite are identified in the diffractograms.

of accumulation of Ni and Cr in leachate sediments than of Zn, Cu and Pb.

3.2. Sequential extraction of sediments

The proportions of each of the metals extracted in each of the six steps, relative to the total concentration of the metals from all extraction steps, are shown in Fig. 4. The results presented for step 4 are the sum of the seven successive extractions with 0.4 M $\text{NH}_2\text{OH}\cdot\text{HCl}$. For most of the metals, large proportions (70–95%) of the total extracted in step 4 were dissolved in the first of the seven repeat extractions. However, this was not the case for Pb, of which as little as 4% was extracted in the first extraction, and relatively more (up to 50%) in the second. This may be due to considerable re-adsorption of Pb to the sediments in the first of the seven successive extractions.

Except for Ni, only very small amounts of the metals were soluble in the first extraction medium (water), indicating that the metals were not present as water-soluble salts. Metals extracted in step 2 are thought to represent exchangeable compounds, i.e. electrostatically adsorbed metals. Small amounts of the Cd, Zn and Cu present in the sediments were extracted in this step, but Ni was the only metal where a significant fraction was extracted (up to 9% of the total Ni content for landfill 1).

Metals extracted in step 3 are thought to be bound to carbonates. Negligible amounts of Cu and Pb were extracted in this fraction. The carbonate-bound fractions of Ni and Zn were variable, ranging from 3 to 33% of the total in the sediments. Up to 50% of the Mn was also in this fraction. Except for the low levels of Pb, these findings are quite similar to those found in a sediment sequential extraction study by Panda et al. [19].

Kersten and Förstner [9] as well as Stephens et al. [20] suggested that aerobic treatment of initially anoxic sediments could convert some of the initially sulphide-bound Zn, Cd and Ni into the carbonate-bound form. The proportions of Zn, Cd and Ni present as carbonate-bound species might be overesti-

mated compared to the initial situation in the anoxic sediments. However, for the oxic form of the sediments, the results are valid.

Step 4 represents easily reducible materials, such as Fe- or Mn-oxides. In all the sediment samples, the largest concentrations of all the metals were in this fraction. The heavy metals might be present as metal oxides, but it is more likely that the metals are adsorbed in or onto the large amounts of iron oxide present in the sediments. In this case, this confirms the scavenging effect that precipitating iron has on heavy metals in landfill leachate and in aquatic systems in general [20–22]. If the heavy metals in the sediments were initially sorbed onto precipitating $\text{Fe}(\text{OOH})$, it is likely that they would also be dissolved if that iron is re-dissolved.

Step 5 extracts metals bound to organic matter or as sulphides. The hydrogen peroxide used in this step decomposes organic matter completely, dissolving any associated metals. As mentioned earlier, it is likely that some of the initially sulphide-bound Ni, Zn and Cd had been converted to carbonates during aerobic treatment of the sediments [9,20], which could account for their relatively low proportion in this fraction. On the other hand, as seen from Fig. 4 between 10 and 30% of the total Cu and 3–15% of the Cr in the sediments was in this fraction. Pb is known to bind strongly to organic material, but in this investigation, only <0.7% of Pb was found to be present in this fraction. Plots (not shown) of the percentage of total metals extracted in this fraction against the total organic contents and total sulphur contents in the sediments, give poor correlations. Thus, a high level of either sulphur or organic matter in the sediments does not necessarily indicate that a high proportion of the metals will be associated with these parts of the sediments.

The last extraction step represents the residual metals in the sediments. Significant amounts of the Cr, Cu, Ni and Pb were found in this fraction. This might be due to incomplete extraction of these metals in step 4 [12], but most likely due to the metals being present associated with poorly soluble minerals (e.g. silicates) [13]. In contrast, little of the Fe, Zn and Cd were present

in the residual fraction. Others have reported that in open sea marine sediments and unpolluted lake sediments, most of the Cr, Ni, Cu, Zn and Fe are in the residual fraction [19,23]. In sewage sludge, the dominant fractions have been found to be the oxidizable and residual fractions [17]. In the current study, the metals were found mainly in the reducible fraction. The leachate sediments thus show a significantly different metal distribution compared to naturally occurring sediments and sewage sludge.

3.3. Bioavailability and mobility of metals in landfill leachate

In the work by Øygard et al. [5], it was shown that almost all of the Cu and Pb (>90%), and a large part of the Cd and Zn (65–80%) in freshly discharged leachate was present as particulate matter >0.45 μm . Only 20–30% of the Cr and Ni in the leachate was present as particulate matter >0.45 μm , with almost all of the remainder as non-labile complexes/colloids <0.45 μm . The investigated landfills all had constructed leachate collection systems draining the leachate directly from the waste masses, and the sampled sediments could only be derived from the leachate, which after the sample collection point was discharged directly into the environment. As large amounts of the heavy metals discharged from landfills are in the form of particulate matter, the results found for the leachate sediments are relevant for assessing the mobility and bioavailability of at least Cu, Pb, Cd and Zn in leachate in general, since the particulate matter suspended in the leachate is the source of the sediments investigated in this work. It is likely, however, that only the larger size fraction of the suspended colloids will settle out of the leachate. As shown by Gounaris et al. [24] and Jensen et al. [25] organic matter will be present in the colloidal matter with the smallest size fraction, and thus this part of the leachate might not be present in the sediments. This might have implications for the level of metal in the sediments, since it has been suggested that metals to a large degree will be associated with the organic matter [25,26].

The metals extracted in the various steps in the sequential extraction procedure represent fractions of varying metal mobility and bioavailability. Metals extracted in the first two steps can be considered mobile and bioavailable. Since only very small portions of the heavy metals were extracted in these steps, this indicates that the heavy metals contained in sediments are generally not readily mobile or bioavailable. Metals extracted in step three are associated with carbonates [8], and thus their dispersion in the environment is pH dependent [27]. Immediate release could occur in acidic freshwater systems, but in a buffered marine environment this would be less likely. If we consider steps one to three as relatively mobile and bio-available, this fraction includes 20–30% of the total Cd, Zn and Ni in the sediments, but only small amounts (on average <3%) of the total Pb, Cu and Cr. As a result of the drying and oxidation of the sediments, it is likely that some of the initially sulphide-bound Ni, Zn and Cd have been transformed to more easily-extractable species, such as carbonates [9,20], and thus included in these figures. Thus, the bioavailable fractions could

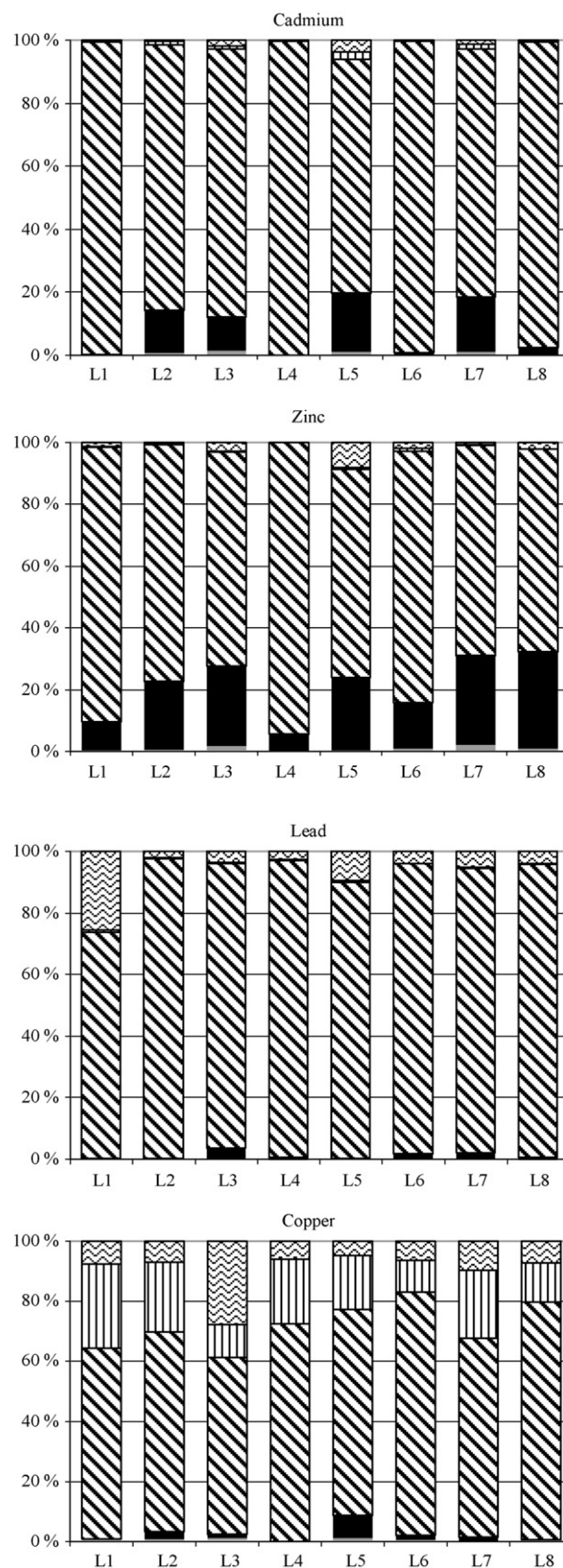


Fig. 4. Distribution of metals (% of total content in the sediment) extracted by the various steps in the sequential extraction of landfill leachate sediments from landfills L1 to L8. The presented results are the average of three extractions for each sample.

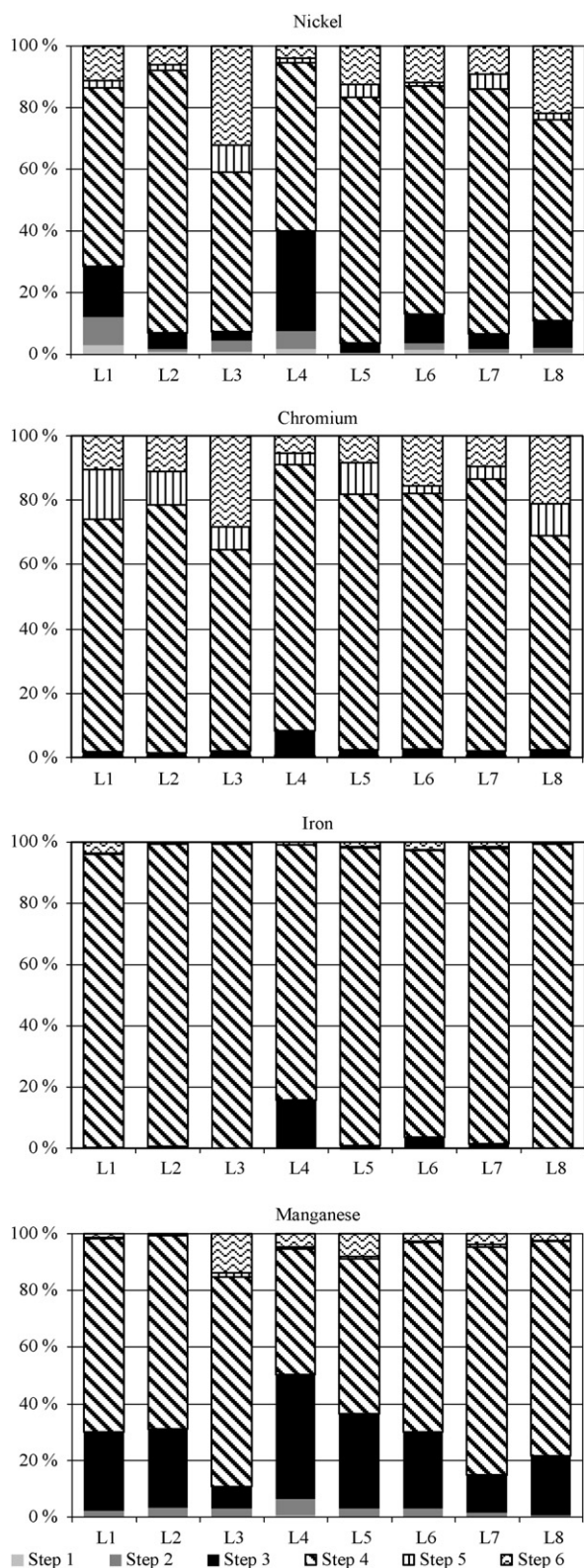


Fig. 4. (Continued).

be overestimated here compared to the initial situation in the sediments. The results in this investigation can only be considered valid for dried dredged sediments. Metals in untreated sediments and particulate matter might have a lower mobility and bioavailability than that suggested here, unless exposed to oxidizing conditions. However, as leachates are often exposed to aerobic conditions once discharged into the natural environment, if not before, the results presented here could in fact give a better indication of the likely behaviour and bioavailability of the metals in the environment.

The fraction extracted in step 4 accounts for the bulk of the metal contents and it is therefore important to consider the mobility of metals obtained in this step. Generally, this portion of the sediments can be thought to be inert, as long as the pH is neutral. In an aerobic environment, metals contained within this fraction have a low mobility, and are not considered bioavailable. The assumed low mobility corresponds with the findings by Ettler et al. [7]. Their status could change, however, in the event of changes in the red-ox conditions [23,28]. Reducing conditions would dissolve Fe(III)-oxyhydroxides, due to the reduction of Fe(III) to Fe(II), with the subsequent solution of the metals bound in or onto those Fe(III)-oxyhydroxides.

In contrast to this, the fractions of metals found to be associated with organic matter or as sulphides (extracted in step 5) could become bioavailable in an aerobic environment, due to the potential oxidation of sulphides and degradation of the organic matter [28]. As seen in Fig. 4 this includes some Ni and Cr (2–10% of the total content in the sediments) and a significant fraction of the total Cu (10–28% of the total content).

Metals extracted in the residual extraction step (step 6) are generally considered neither mobile nor bioavailable. It is unlikely that any naturally occurring processes could dissolve these metals.

4. Conclusion

Leachate sediments from landfills in western Norway have levels of heavy metals that are equal to or lower than those found in sewage sludge, but a significant content of iron (4.1–24%).

The sequential extraction of dried, homogenized landfill leachate sediments shows that the bulk of the heavy metals are associated with Fe- or Mn-oxides, or present as metal oxides. Cr and Cu were the only metals that were associated with organic matter to a significant degree (2–10% for Cr and 10–28% for Cu). Up to 30% of the metals were extracted only in the final (residual) step in the sequential extraction, indicating that they are associated with minerals with a low solubility and low bioavailability.

In the first three steps of the sequential extraction, generally considered to contain the bio-available metal fractions, 20–30% of the total Cd, Zn and Ni were extracted, but only small fractions of the other metals. Since the bulk of the metals are bound to Fe-oxides, which also have low solubility, metals in both the leachate sediments and in particulate matter suspended in leachate can be said to have a low bioavailability and thus probably also a low eco-toxicological effect under aerobic conditions.

The sediments were dried in an aerobic atmosphere prior to the investigation, so the results are only totally applicable to dry dredged sediments, although they also have relevance for the release of sediments into aerobic environments, which is often the case with landfill leachate. However, more research needs to be done to assess the metal mobility in anoxic sediments.

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