

# Possibility Of Using Cardboard Mill Sludge In Remediation Of Contaminated Sediment

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## Abstract:

The sludge from cardboard mill is commonly landfilled, but it could be recycled into production on-site or reused in some other way. In this study the use of sludge from cardboard mill as stabilizing agent in the solidification/stabilization (S/S) treatment of lead polluted sediment was examined. The effectiveness of S/S treatment was evaluated by determining cumulative percentage of lead leached and by applying different leaching tests. Applied S/S treatment was effective in immobilizing lead irrespective of high concentration in the untreated sample.

**Keywords:** cardboard mill sludge, contaminated sediment, lead, remediation, solidification/stabilization, leaching mechanism, leaching tests

## Introduction

Paper and cardboard mill sludge is generated by various processes in the production of paper and cardboard, and increasing quantities produced make the disposal of this sludge a problem (Battaglia et al., 2007; Monte et al., 2008).

Waste is mainly generated from pulping, deinking unit operations and wastewater treatment. The amount and the composition of the waste depends on the paper and cardboard grade produced, the raw materials used, the process techniques applied and the properties to be achieved. Cardboard mill wastewater treatments typically include a primary treatment consisting of neutralization, screening, and sedimentation to remove suspended solids, which are then dewatered into a sludge that requires disposal. This primary sludge contains wood fibers as the principal organic component, as well

as paper and cardboard making fillers (inorganic materials such as kaolin, CaCO<sub>3</sub>, TiO<sub>2</sub>, etc.), pitch (wood resin), lignin by-products, inert solids rejected during the chemical recovery process, and ash. The primary method of disposal for this type of sludge has been landfilling (Jackson et al., 2000; O'Brien et al., 2003). Landfilling costs in the EU are rising because of increasingly stringent regulations, taxes, and declining capacity. With landfill space becoming scarce and expensive, some sludges are being burned or incinerated to reduce their volume and to recover part of the energy they contain (Navaee-Ardeh et al., 2006).

The use of paper and cardboard mill sludges could be a good practice for sediment remediation, because of their organic matter, silicate and carbonate content. The organic matter is able to form stable complexes with several metals; the silicates are materials of high cation exchange capacity (CEC) and the bicarbonate/carbonate system is able to increase the pH value of soil. These chemical features were able to reduce the harm-

ful mobile metals in polluted soils when the paper mill sludge was added to them (Calace et al., 2005).

In this study stabilization/solidification (S/S) was used in order to remediate lead contaminated sediment, since this is a widely accepted treatment process for the immobilization of heavy metals contained in wastes (Coz et al., 2009; de Groot and van der Sloot, 1992; Voglar and Leštan, 2010). S/S of sludge originating from cardboard industry is a potential tool to stimulate correct industrial sludge disposal in a safe and profitable manner. In this regard, we propose incorporation of this sludge into contaminated sediment in order to combine two types of wastes. At present, dredged sediment, contaminated or not and cardboard mill sludge in Serbia are deposited in a landfill.

Lead (Pb) is ubiquitous heavy metal pollutant in the environment due to wide-spread use that have received a great deal of attention because of its toxic effects. Sediments are the final sink for lead input to the water environment, originating from both natural and/or anthropogenic sources, but they may also act as endogenous contamination sources for aquatic systems (Zhang et al., 2010). Toxicity and the mobility of elements depend strongly on their distribution in sediment and its identification helps in understanding geochemical processes in order to evaluate the remobilisation potential and the risks induced (Wise et al., 2000). Most commonly used methods for that are the single and sequential extraction procedures. Several sequential fractionation procedures have been suggested to identify element distribution in "operationally" defined pools; from water soluble to forms immobilized in mineral lattices. The five step protocol developed by Tessier et al. (1979) is the most widely used while many modified procedures with different reagents or operational conditions have been proposed (Pertsemli and Voutsas, 2007). In some studies (Ghrefat and Yusuf, 2006; Jain, 2004; Martley et al., 2004) the risk assessment code (RAC) has been used to assess environmental risks and estimate possible damage to benthic organisms caused by contaminated sediments based on the results of sequential extraction procedures. The RAC considers the percentage fraction of metals that are exchangeable and associated with carbonates. In this fraction, the metals are weakly bound to the sediment, and present a greater environmental risk since they are more available to the aquatic system. The RAC classification defines risk levels as zero, low, medium, high and very high, depending on the percentage value (Ghrefat and Yusuf, 2006; Jain, 2004; Martley et al., 2004; Passos et al., 2010).

The mechanisms governing heavy metals leachability of contaminants from solidified waste forms and evaluation of the long-term behaviors of S/S wastes can be effectively examined using the American Nuclear Society's (ANS) semi-dynamic leaching test (ANS, 1986).

The ANS 16.1 provides substantially more information regarding the "real time" rate at which heavy metals are released from the solidified product as compared to other leaching tests. The leaching results extend over a 90-day period instead of a single result at the end of the test. The most often used leaching test, recommended by the USEPA, but which provides only one result for defining the waste toxicity, is the toxicity characteristic leaching procedure - TCLP test (USEPA, 2002). The TCLP was specifically designed to mimic acidic conditions in a sanitary landfill and identify wastes that have potential to contaminate ground water. The Waste extraction test - WET is used in California (CCR, 1998), US, in a similar manner as the TCLP (determination of whether a solid waste is a hazardous waste), with the exception of the liquid-to-solid ratio (10:1) and the leaching time, the remain of the test was the same as the TCLP. Also, synthetic precipitation leaching procedure - SPLP tests was developed by USEPA in order to assess metal mobility in wastes (USEPA, 2002a). The SPLP test reproduces acid rain conditions and estimates metal mobility when wastes are disposed in an open area. The SPLP is conducted in a similar fashion as the TCLP with the exception of the leaching fluid.

In view of the above, the objectives of this study were to define lead distribution in dredged sediment and evaluate environmental risk based on the results of sequential extraction procedure and to assess the effectiveness of lead immobilization, S/S treatments with cardboard mill sludge based on the results of different leaching tests.

## Materials and methods

Fresh sediment was collected, using Eijkelkamp core sampler, from a middle of sediment reach Great Backa Canal (Serbia) and was placed in a sealed plastic, acid-rinsed box (15x15 cm and 20 cm deep) immediately after sampling. Organic matter content was determined as ignition loss, and it was  $4.48 \pm 0.1$  (NEN, 1994). The sediment pH was measured according to ASTM D 4972-01 (2007) and it was  $7.3 \pm 0.4$ . The given values represent the means of three measurements.

The cardboard mill sludge used was obtained from cardboard factory situated in Belgrade (Serbia) after physico-chemical wastewater treatment and its chemical characterisation was: pH  $8.6 \pm 0.3$ , CEC 14.8 (cmol  $\text{kg}^{-1}$ ), water content 39%, organic matter 31%, carbonates 25%, silicates 47%, Pb  $0.8 \text{ mg kg}^{-1}$ . The sludge was dried, crushed and passed through a 2.0 mm sieve, then characterised and used in the experiment.

Pseudo-total lead content was assessed on sample triplicate after aqua regia digestion (ISO, 1995) and mean

values were used. The standard deviations (% R.S.D.) obtained ( $n=3$ ) were below 10%. The results of sediment lead pseudo-total concentrations are discussed in reference to Dutch regulation standards and Canadian guidelines, since Serbia has neither an established system of continual monitoring of sediment quality nor regulations concerning the quality standards (CCME, 1995; Ministry of Housing, Spatial Planning and Environment Directorate-General for Environmental Protection, 2000). Microwave assisted sequential extraction procedure (MWSE) was performed as described by Jamali et al. (2009), using identical operating conditions applied in each individual BCR fraction. Mean values were used and the RSDs ( $n=3$ ) were below 5%. The sum of the four steps sequential procedure and pseudo-total lead content differed less than 10%. Pseudo-total sediment metal content and lead content in sequential extraction procedure steps were analyzed by AAS (Perkin Elmer AAnalyst™ 700) or ICP-MS (Perkin Elmer Sciex Elan 5000) according to the standard procedures (NEN, 1990).

Sediment and cardboard mill sludge possessing an average initial moisture content of 75% and 39% respectively were dried at 105°C to a constant mass. The sediment and paper mill sludge were mixed in proportion of 95:5 (M1), 90:10 (M2), 80:20 (M3), 70:30 (M4) and 50:50 (M5) by wt. Samples were prepared in the form of monolithic cubes ( $(3 \pm 0.1) \times (3 \pm 0.1) \times (3 \pm 0.1)$  cm) by compaction. The compaction was performed according to ASTM D1557-00 (2000), providing a compactive effort of 2700 kN m m<sup>-3</sup>. Samples were cured at 20°C in sealed sample bags for 28 days and then subjected to the leaching tests: standard TCLP leaching method and ANS 16.1.

TCLP test was done according to the USEPA protocol (USEPA, 2002) 0.1 M acetic acid solution with a pH of 2.88 was used to extract control sample and S/S-treated samples since the pH was above 5. The sediment samples were extracted at a liquid to solid (L/S) ratio of 20 in capped polypropylene bottles on a rotary tumbler at 30 rpm for 18 h. After the extraction, the final pH of the leachate was measured and the liquid was separated from the solids by filtration through a 0.45 µm pore size membrane filter. This test was applied on every sample in triplicate and the RSDs were below 10%.

The WET test (CCR, 1998) is similar to the TCLP in that it uses a buffered organic acid solution as the extraction fluid. This test uses a pH buffered citrate acid solution with sodium hydroxide, a 10:1 L/S ratio, and a 48-hour testing period. The WET extraction solution is prepared with a combination of 0.2 M citric acid solution and 4.0 N NaOH to pH  $5.0 \pm 0.1$ . One liter of this solution is added to a 100-g sample and rotated for 48 hours. After rotation, the final pH is measured, and

the samples were filtered through a 0.45 µm pore size membrane filter. This test was applied on every sample in triplicate and the RSDs were below 10%.

The SPLP test is performed in the same manner as the TCLP. The extraction fluid is made of two inorganic acids (nitric and sulfuric acid) to simulate acidic rain-water. In a similar fashion as the TCLP, a 100-g sample of waste material is placed in a 2-liter extraction vessel and mixed with the extraction fluid. The leachate is then filtered through a 0.45 µm pore size membrane filter (USEPA, 2002a). This test was applied on every sample in triplicate and the RSDs were below 10%.

The ANS 16.1 test was applied in duplicate only on the M2 sample, with deionized water as leachant, and the RSDs were below 10%. The ANS 16.1 method was modified by including 0.014 M acetic acid (AA) pH 3.25 as leachant instead of deionized water (DI). This was applied with the objective to mimic the worst possible conditions of the S/S waste disposed in the environment (influence of acidic rains). All materials in contact with the leachant were pre-cleaned with HNO<sub>3</sub> and subsequently rinsed with deionised water. All results are expressed with respect to sediment dry matter.

## Results and discussion

### Pseudo-total metal concentrations and sequential extraction of untreated sample

Pseudo-total lead concentration in sediment was  $2085.6 \pm 59.6$  mg kg<sup>-1</sup>. According to Dutch regulation standards, sediment is polluted with lead (class 4). Class 4 sediments are of unacceptable quality and need highest urgency situation, dredging, disposal in special storage reservoirs, and, if possible, sediment clean-up measures (Ministry of Housing, Spatial Planning and Environment Directorate-General for Environmental Protection, 2000). Compared with Canadian Sediment Quality Guidelines (CCME, 1995) for aquatic life protection, Pb content is above the PEL. Sediment concentrations above PEL values are expected to be frequently associated with adverse biological effects. Although PEL is considered to be applicable to a variety of sediment types, it cannot define uniform values of sediment pollution as the bioavailability (and hence toxicity) of contaminants may be different.

First fraction in sequential extraction procedure is the most dangerous for the environment and a small proportion of lead (1.70%) is present in the exchangeable forms and bound to carbonates. Metals in exchangeable fraction can be exchanged and are in equilibrium with the ionic content in water. Carbonates fraction mainly refers to the metals that are precipitated or co-precipitated with carbonate. This fraction is sensitive to pH variations. Larger portion of lead is bound to reducible

Table 1. Total cumulative fraction of lead leached (CFL, %) after ANS 16.1 test completion using deionized water (DI) and acetic acid (AA) as leachants for untreated (SO) and treated sediment samples (M1, M2, M3, M4, M5)

DI	SO	M1	M2	M3	M4	M5
CFL (%)	18.5	0.08	0.07	0.18	0.25	0.57
AA						
CFL (%)	19.4	0.27	0.15	0.43	1.20	1.88

phases, Fe and Mn oxides (26.9%). The Fe–Mn oxides fraction includes the soluble metal oxides/hydroxides under slightly acidic pH as well as the metal associated with reducible amorphous Fe–Mn oxyhydroxides. This fraction can be dissolved with oxidation–reduction potential change. The distribution profile of lead suggests that the major portion of lead is associated with residual fraction (70.6%). Residual fraction represents the stable metal forms associated with anthropogenic or geogenic components, the influence of which on the ecological system is much smaller than the others under the majority of conditions (Quevauviller, 2002). Other studies also reported similar lead fractionation (Guevara-Riba et al., 2004; Jamali et al., 2009; Pertsemli and Voutsas, 2007; Pueyo et al., 2008).

The risk connected with the type of association, or the type of bounding of metals in sediment is described by risk assessment code (RAC). This criterion indicates that a sediment which can release in exchangeable and carbonate fractions less than 1% of the total metal will be considered safe for the environment. On the contrary, a sediment releasing in the same fraction more than 50% of the total metal has to be considered highly dangerous and can easily enter the food chain (Jain, 2004). According to RAC, 1.70% of cadmium in carbonate phases comes under the low risk category.

The nonresidual fractions (exchangeable+reducible+oxidizable) of lead was 30.7%. This is an indication that these fractions of lead in sediment are potentially available for exchange and/or release into the environment.

#### Leaching Tests: ANS 16.1, TCLP, WET, SPLP

Samples of sediment-cardboard mill sludge mixtures were subjected to the semi-dynamic leaching test ANS 16.1 (ANS, 1986). This test has not been previously ap-

plied on the mixtures of metal-contaminated sediment with cardboard-mill sludge. Total cumulative fraction of lead leached (%) after ANS 16.1 test completion using deionized water (DI) and acetic acid (AA) solution as leachants are presented in Table 1.

The amount of lead released during the ANS 16.1 tests for the untreated samples did not exceeded 20% of the total mass of the contaminant in the waste, which is the upper limit for the diffusion model to be still applicable (ANS, 1986). The cardboard mill sludge exhibit good sorption properties reducing significantly its leachability compared to untreated sample. This is in agreement with literature data for soil remediation (Battaglia et al., 2007; Calace et al., 2005).

Amount of lead leached sometimes increases with higher sorbent loading (Table 1). Similar results have been reported by other authors (Bhattacharyya and Gupta, 2008; De la Rosa et al., 2011, Pérez-Novo et al., 2008). This may be attributed to two reasons: (i) higher adsorbent amount creates particle aggregation, resulting in a decrease in the total surface area and an increase in diffusional path length both of which contribute to decrease in amount adsorbed per unit mass and (ii) a large adsorbent amount effectively reduces the unsaturation of the adsorption sites and correspondingly, the number of such sites per unit mass comes down resulting in comparatively less adsorption at higher adsorbent amount.

At present, TCLP is used by the USEPA to evaluate whether a particular S/S process is effective in treating a given waste in terms of reduction of contaminant mobility and toxicity (USEPA, 2002). TCLP leaching test results are presented in Table 2.

A comparison of the concentrations obtained in the TCLP test showed that lead concentration in treated

Table 2. Results of the TCLP, WET and SPLP leaching test ( $\text{mg L}^{-1}$ ) for untreated (SO) and treated sediment samples (M1, M2, M3, M4, M5)

	SO	M1	M2	M3	M4	M5
TCLP	2.5	0.03	0.04	0.05	0.06	0.08
SPLP	2.3	0.01	0.02	0.03	0.05	0.07
WET	3.9	0.59	0.21	0.66	0.69	0.82

samples is below concentration obtained for untreated sediment which means that the lead is successfully incorporated in treated matrix.

The TCLP Pb concentration was  $2.5 \text{ mg L}^{-1}$  of leachate for untreated sediment, which was lower than the regulatory limit of  $5 \text{ mg L}^{-1}$  for hazardous waste (USEPA, 2002). This is agreement with the results of sequential extraction procedure. Despite of high pseudo-total lead concentrations lead poses low risk based on the results of SEP and TCLP. The five S/S treatments reduced the TCLP concentration to less than  $0.08 \text{ mg L}^{-1}$ , which was well below the TCLP limit.

Lower concentrations of Pb were leached in SPLP than the TCLP test in all samples. This is consistent with literature (Townsend et al., 2004; Janin et al., 2009). Differences in metal leachability between TCLP and SPLP might result from changes in the solution pH that occur during the 18 h of leaching that differ between SPLP and TCLP, and thus result in different amounts of metal leaching (Townsend et al., 2004).

The WET extracted higher concentrations of Pb than the TCLP and SPLP but below WET limit values (CCR, 1998). This is consistent with literature (Townsend et al., 2004; Townsend et al., 2005). TCLP is conducted at a 20:1 liquid-to-solid ratio and WET is carried out at 10:1 liquid to solid ratio; the TCLP is twice diluted compared to WET and in general higher leachate concentrations are observed at lower liquid-to-solid ratio. The greater element concentrations observed in the WET leachates relative to the leachates most likely result, however, from citrate's propensity to chelate Pb. Citric acid has multidentate ligands while acetic acid has monodentate ligands, and in general, complexes with monodentate ligands are less stable than those with multidentate ligands (Townsend et al., 2004; Townsend et al., 2005).

While the single leaching tests TCLP, SPLP, WET are rapid they do not simulate complex environmental settings. Also, sequential extractions provide semi-quantitative information on element distribution between operationally defined geochemical fractions. Therefore, the fractions obtained from sequential extractions do not necessarily reflect true chemical distribution. Not to forget that one of the main limitations of sequential extraction procedures are that they are extremely time-consuming, so that they are less used for routine analysis. Although leaching techniques such as column leaching tests are probably more realistic to field conditions, sequential extractions and single step extractions can give an indication of the 'pools' or 'sinks' of heavy metals that are potentially available under changing environmental conditions.

Overall, the treatment was efficient having in mind that sediment sample had high initial lead concentration even under the conditions that mimic acidic rains. This may be difficult to readily explain due to the degree of complexity of the natural sediment samples. In nature there are many constituents that could participate in and influence lead leachability. Further research is required to obtain some of this information in order to evaluate the lead speciation and subsequent mechanisms of incorporation and release in these samples.

## Conclusion

The assessment of the sediment quality based on the pseudo-total lead content according to the corresponding Dutch standards and Canadian guidelines showed its severe contamination. Sequential extraction can be used as a valuable tool to provide information on the mobility, bioavailability and potential toxicity of trace metals in the environment. Lead seems to pose low risk based on the modified BCR sequential extraction and risk assessment code, which is not in accordance with the assessment of the sediment quality based on the Dutch standards and Canadian guidelines.

The immobilization treatment with cardboard mill sludge applied appeared to be efficient in the remediation of sediment contaminated with lead. Based on the cumulative fraction of lead leached and results of TCLP, WET and SPLP tests it appeared the treatment was efficient in reducing lead mobility. This could be explained by the presence in the sludge of organic matter that is able to bind the lead very strongly.

Due to the large volumes of waste generated in the cardboard, paper and pulp industry, it is necessary to continue environmental awareness on different applications of wastes, while taking into account the environmental and economic factors of these waste treatments. Conventional waste management methods, which might have been acceptable in the past, might not be optimal to meet present and future requirements. Cardboard mill sludge utilisation to "remediate" a contaminated sediment could be an interesting approach.

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