
Chemical Speciation & Bioavailability

ISSN: 0954-2299 (Print) 2047-6523 (Online) Journal homepage: https://www.tandfonline.com/loi/tcsb20

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**To cite this article:** Nan-Nan Zhao, Xiao-Jia He, Zhen-Shan Li & Hua-Zhang Zhao (2015) Effects of paper sludge addition on the bioavailability and distribution of Pb in contaminated soil, Chemical Speciation & Bioavailability, 27:4, 169-174, DOI: 10.1080/09542299.2015.1121519

**To link to this article:** https://doi.org/10.1080/09542299.2015.1121519

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Published online: 07 Jan 2016.

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CHEMICAL SPECIATION & BIOAVAILABILITY, 2016
VOL. 27, NO. 4, 169–174
http://dx.doi.org/10.1080/09542299.2015.1121519

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# Effects of paper sludge addition on the bioavailability and distribution of Pb in contaminated soil

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**ABSTRACT**

Heavy metal contamination of soil poses risks and hazards to environment and human being. Many amendments were used to remediate the contaminated soil. In this report, paper sludge was used to reduce the bioavailability of Pb in soil, and the remediation mechanism was studied by investigating the redistribution of Pb speciation after paper sludge addition. In pot experiments, significantly increased weights of shoots (from 1.6 to 3.3 mg per plant) and roots (from 0.7 to 0.8 mg per plant), as well as significantly decreased Pb content in shoots (from 153.8 to 24.4 mg $kg^{-1}$) and roots (from 467.1 to 38.0 mg $kg^{-1}$) of rape were observed after paper sludge was added. The addition of paper sludge resulted in redistribution of Pb from the liable fractions (carbonate-bound Pb from 58.3 to 3.7%) to the Fe–Mn oxide-bound fraction (from 29.2 to 74.9%). Paper sludge addition increased the content of organic matters and pH of soil, and induced Pb redistribution, which ultimately inhibited Pb uptake and improved plant growth.

**ARTICLE HISTORY**
Received 5 July 2015
Accepted 13 November 2015

**KEYWORDS**
Paper sludge; contaminated soil; metal fraction; bioavailability; heavy metal

## 1. Introduction

Heavy metal contamination of soil poses risks and hazards to humans, which has become a serious environmental problem due to the rapid industrial development worldwide.[1,2] Metal bioavailability does not necessarily correspond with total metal concentrations, but is determined largely by the chemical fractions.[3] Based on solubility, heavy metals are often classified into five operationally defined chemical forms, including exchangeable (EXC), carbonate-bound (CAR), Fe–Mn oxide-bound (OX), organic matter-bound (OM), and residual (RES) fractions.[4] Solubility is an important factor controlling bioavailability,[5] and the metal bioavailability decreases gradually in the order from the EXC to the RES fraction.[6] The sequential extraction procedure has been widely applied in the mobility and bioavailability analysis of heavy metals in soil.[6] Several soil characteristics, such as pH, cation exchange capacity, and organic matter content, may together contribute to the metal distribution.[7,8]

Soil amendments, mainly by adding materials to soil to improve soil quality, are often used to reduce the metal concentration and bioavailability.[9–12] Commonly used soil amendments include organic matter,[13] calcium hydroxide,[14] phosphates,[15] and nanometallic materials.[16] Municipal sludge has been demonstrated to have several advantages for immobilization of heavy metals in soil, including low cost and high effectiveness.[17–20]

In recent years, paper sludge has been suggested to be a potential soil remediation for metal-contaminated soils.[21–23] Paper sludge, a municipal solid waste produced by the paper industry during papermaking and wastewater treatment, contains abundant organic matters (i.e. cellulose fibers) and inorganic matters (i.e. $CaCO_3$).[14,24] The increase in biological activity, porosity, and stability of aggregates in soil added with paper sludge has positive effect on the plant growth, suggesting that paper sludge could be an effective soil amendment.[22,23,25] To date, most studies have been focused on the effectiveness, adsorption–desorption kinetics, and vegetation growth in soil with paper sludge.[26–29] Unfortunately, the mechanism triggered by paper sludge, especially how the distribution of metal chemical fractions is affected by paper sludge, has not been investigated so far.

The objective of this study was to investigate the effect of paper sludge on the bioavailability and distribution of heavy metal in soil. Heavy metal Pb was used as a model contaminant, as it showed acute toxicity and carcinogenicity, and could find its way into human populations through the food chain.[30] In pot experiments of rape grown in Pb-contaminated soil with and without sludge,

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Supplemental data for this article can be accessed at http://dx.doi.org/10.1080/09542299.2015.1121519.

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we collected the shoots and roots to examine the weight and metal content for Pb bioavailability assessment. We also examined distribution of Pb when soil was added with paper sludge at different ratios or incubated with different times to probe the remediation mechanism.

## 2. Materials and methods

### 2.1. Preparation of Pb-contaminated soil

Red soil collected from Hunan Province, China, was classified as wet sticky iron-rich soil according to the *Chinese Soil Taxonomy*.[31] The soil collected 25–50 cm below the ground surface was air-dried and passed through a 2-mm sieve. Soil characteristics are summarized in Table S1 (Supporting materials). The Pb concentration in the soil was 32.2 mg $kg^{-1}$. Uncontaminated soil was sprayed with appropriate lead chloride solution to produce Pb-contaminated soil samples with Pb concentrations of 500, 1000, and 1500 mg $kg^{-1}$, respectively. The mixture of soil with metal was mixed thoroughly and incubated at room temperature (25 ± 2 °C) for about three weeks. Then, the contaminated soil was oven-dried at 60 °C, crushed, and passed through a 2-mm sieve for analysis. The final Pb concentrations in the soil were determined to be 500.4, 1000.1, and 1500.3 mg $kg^{-1}$.

### 2.2. Addition of paper sludge

Paper sludge provided by the Gold East Paper Co. Ltd., Jiangsu, China, was utilized as soil amendment. Some important physicochemical characteristics of the paper sludge are listed in Table 1. The Pb in the paper sludge was undetectable.

Paper sludge was dried, crushed, and added to the soil samples with initial Pb concentrations of 500, 1000, and 1500 mg $kg^{-1}$, respectively, at different ratios of soil to paper sludge by weight. The mixture was kept in a plastic bottle at room temperature and maintained at constant wet condition for about 15 days. The soils with paper sludge were denoted by weight ratio of soil/paper sludge as SP1 (9:1), SP2 (9:4), and SP3 (9:8), and soil without paper sludge was denoted as CK for control. A 90-day incubation experiment was performed.

During the 90-day incubation, soil samples were collected at day 7, day 35, day 55, and day 90, respectively, and placed in an oven and dried at 30 °C for 48 h. A smaller-sized sample is obtained by grinding sample with pestle and mortar, and preserved in a plastic bottle for the metal fraction analysis. Results were obtained in triplicates, and data presented represent the average of three independent experiments.

### 2.3. Pot experiments

Rape was selected as the target plant. Seven rapeseeds were sown in each pot in order to ensure at least five sprouts per pot. Five plants were left in each pot after their germination. Plants were grown in plastic containers filled with 600 g of Pb-contaminated soil (500 mg $kg^{-1}$). All experiments were carried out in a greenhouse with natural light and constant temperature. Light (14 h)/no light-time temperatures were 23/18 °C with 60% relative humidity. After 55 days growth, plants were harvested and cut to ground level for metal concentration test. The roots and shoots were rinsed with the tap water and deionized water, and dried in an oven at 75 °C for 48 h before analysis. The dried shoots and roots were weighed.

The above-mentioned roots and shoots (0.2–0.5 g) were digested by dry ashing procedure at 450 °C for 4 h.[32] One gram of air-dried soil sample was digested with 11 mL of $HNO_3$ + HF + $HClO_4$ (5:3:3) at 180 °C. Pb concentrations were measured by atomic absorption spectrophotometry after digestion.

### 2.4. Pb fractionation

The Pb fractionation in soil was assessed following the modified five-step sequential extraction procedure to separate Pb into five fractions including EXC, CAR, OX, OM, and RES Pb.[6,26] For example, for EXC fraction, 4 mL $MgCl_2$ (1 mol $L^{-1}$, pH 7) was added to 0.5 g

**Table 1.** Physicochemical characteristics of the paper sludge.

| Characteristics | Result |
|---|---|
| pH | 8.21 |
| Water (%) | 40.60 |
| Volatile (%) | 56.31 |
| Ash (%) | 45.68 |
| IEC (cmol $kg^{-1}$) | 17.48 |
| C (%) | 25.36 |
| H (%) | 2.63 |
| N (%) | 0.13 |
| TOC (%) | 25.36 |
| Exchangeable-Ca (cmol $kg^{-1}$) | 14.28 |
| Exchangeable-Mg (cmol $kg^{-1}$) | 1.82 |
| Exchangeable-K (cmol $kg^{-1}$) | 0.22 |
| Exchangeable-Na (cmol $kg^{-1}$) | 1.16 |
| Ca (%) | 16.2 |
| Si (%) | 0.49 |
| Al (%) | 0.40 |

air-dried soil as mentioned before and shaken for 1 h (25 °C, 200 r $min^{-1}$). The mixture was filtrated to separate the extract (EXC fraction) from the residue. The residue was used for the next extraction. The CAR, OX, and OM fractions were separated following the same procedure with different extracting agents as shown in Table 2. The RES fraction was obtained after the complete digestion of the residue of the extraction for OM fraction. Pb concentrations in all fractions were measured, respectively, and the percentage of each fraction Pb in the total Pb (the sum of the five fractions) in the soil was calculated.

Pb concentrations were determined by atomic absorption spectrophotometry. pH was measured by a 201 acidimeter (Hanna instruments Ltd., Italy). All reagents used were of analytical or higher grade. All plastic materials were soaked in 5% $HNO_3$ overnight and rinsed with distilled water before use.

### 2.5. Statistical analysis

Analysis of variance of the measured parameters was performed using MSTAT-C, and the means were compared using Duncan's multiple range test at 5% probability level.

## 3. Results and discussion

### 3.1. Effect of paper sludge addition on plant growth

Figure 1(A) shows the dry weights of the shoot and root of rape grown for 55 days in Pb-contaminated soils with or without paper sludge addition. We observed significantly higher weight of shoot than root in all samples, as well as significantly higher weight of shoots in soils with sludge than without sludge ($p < 0.05$). The weight of shoots and roots increased (1.6–3.3 mg per plant for shoots and 0.7–0.8 mg per plant for roots, $p < 0.05$) with the addition of paper sludge in Pb-contaminated soils. In Figure 1(B), the total content of Pb decreased in dried shoots (from 153.8 to 24.4 mg $kg^{-1}$) and roots (from 467.1 to 38.0 mg $kg^{-1}$) significantly when the soil was added with paper sludge ($p < 0.05$). In SP3 soil, the total content of Pb in shoot reached a minimum value of 13.4 mg $kg^{-1}$. The results demonstrated that paper sludge significantly reduced the bioavailability and uptake of Pb, which relieved the stress generated from Pb on rape and then promoted the rape growth. The change of the bioavailability of Pb must be related to the change of Pb speciation.

### 3.2. Effect of paper sludge addition on Pb distribution

Figure 2 shows Pb distribution before and after the addition of paper sludge in the Pb-contaminated soils for 35 days. Regardless the initial Pb concentration (500, 1000, and 1500 mg $kg^{-1}$), the change of Pb distribution showed similar tendency. Before the addition of paper sludge, more than 50% Pb was distributed in two liable fractions (EXC and CAR fractions) (Figure 2(A)), which were of high solubility and potential bioavailability for organisms.[33] The addition of paper sludge significantly decreased the proportion of Pb in the EXC and CAR fractions ($p < 0.05$). The loss in the liable fractions was balanced by a significant increase in the OX and OM fractions ($p < 0.05$), though the RES fraction was not changed obviously. After the addition of paper sludge, most Pb was distributed in the OX fraction (Figure 2(B)), which could be related to the improved capacity of Fe–Mn oxide binding to Pb. The results showed that, regardless the initial Pb concentration, the addition of paper sludge transformed EXC and CAR fractions to OX and OM fractions in the soil, resulting in the reduced Pb bioavailability.

Figure 3 shows the percentage of fraction Pb in the total Pb in the soil prepared with different ratios of soil to paper sludge. The percentage of OM or RES Pb showed no significant change with the mixture ratio. In the soil without paper sludge, 11.3% Pb was distributed in the EXC fraction, while after the addition of paper sludge, the percentage of EXC Pb decreased to 2.5%. The decrease in the liable fractions was balanced by a significant increase in the OX Pb ($p < 0.05$). The percentage of CAR Pb decreased from 45.5% in SP1 to 25.3% in SP3, and the percentage of OX Pb increased from 39.2% in SP1 to 60.8% in SP3. With the increased ratio of paper sludge, Pb showed increased transformation from liable forms (EXC and CAR fraction) into stable forms (OX fraction),

Table 2. Chemical extraction of Pb fractions.

| | | Extraction conditions | | |
|---|---|---|---|---|
| Fraction | Extracting agents | Shaking time (h) | Temperature (°C) | Separate method |
| Exchangeable (EXC) | 1 mol $L^{-1}$ $MgCl_2$ | 1 | 25 | Filtration |
| Carbonate -bound (CAR) | 1 mol $L^{-1}$ NaAc | 5 | 25 | Filtration |
| Fe–Mn oxide -bound (OX) | 0.04 mol $L^{-1}$ $NH_2OH{\cdot}HCl$ | 5 | 96 | Filtration |
| Organic-bound (OM) | 68%$HNO_3$ + 30%$H_2O_2$ | 5 | 85 | Centrifugation |
| | 3.2 mol $L^{-1}$ $NH_4Ac$ | 0.5 | 25 | |
| Residual (RES) | 68%$HNO_3$ + 40%HF + 38%$HClO_4$ | 2 | 160 | Not needed |

Notes: The soil was extracted and the residue was separated and used for the next extraction. Pb fractions were obtained after the extractions with different extracting agents and extraction conditions. The RES fraction was the residue of the extraction for OM fraction and was completely digested before Pb analysis. The Pb concentrations in the extracts or the digested solutions were measured by atomic absorption spectrophotometry.

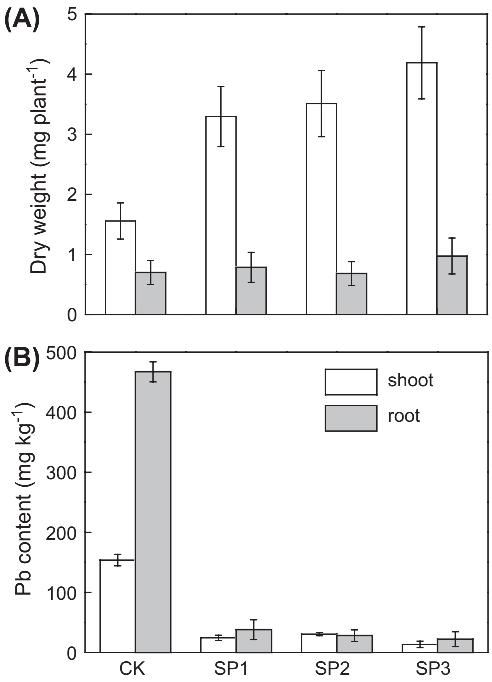


**Figure 1.** Dry weights (A) and Pb contents (B) in shoots and roots of rapes grown for 55 days in the Pb-contaminated soil (500 mg $kg^{-1}$) with different mixture ratios of soil to paper sludge by weight: CK (9:0), SP1 (9:1), SP2 (9:4), and SP3 (9:8). Rapes were grown in plastic containers filled with 600 g of soil. All experiments were carried out in a greenhouse with natural light and constant temperature. Light (14 h)/no light-time temperatures were 23/18 °C with 60% relative humidity.

suggesting that paper sludge reduced the bioavailability in a dose-dependent manner.

### *3.3. Mechanism of Pb redistribution by paper sludge*

Paper sludge consists of inorganic components ($CaCO_3$, etc.) and organic components (residual fiber, etc.), which could alter the Pb speciation redistribution. The soil pH was increased with the increased ratio of paper sludge in Pb-contaminated soil. The soil pH was 6.34, 6.97, 7.45, and 7.64 at the ratio of soil/paper sludge of 9:0, 9:1, 9:4, and 9:8, respectively. Such increase might be related to the high content of $CaCO_3$ in the paper sludge, an inorganic material that is able to increase pH of the soil system.[34] It has been demonstrated that the content of OX fraction increases when pH increases in soil.[7,32,35] Therefore, after paper sludge addition, $CaCO_3$ in the paper sludge increased the soil pH and transformed Pb EXC and CAR fractions into OX fraction.

It was shown in the previous research that organic matters has been associated with redistribution of heavy metals, such as Cu, Zn from mobile fractions to stable fractions.[32] In the present work, the percentage of organic matter was 25.36% in the paper sludge (Table 1) and 0.63% in the soil (Table S1), strongly supporting that paper sludge could increase the organic matter content in soil. The role of organic matters in redistributing Pb speciation was dependent on the structure and property of organic matters, which will change during the incubation of paper sludge added in soil. As a result, Pb fraction in the soil was changed with the incubation time. For the soil without paper sludge, the fractions of Pb remained almost the same with the incubation time (Figure 4(A)). After the addition of paper sludge, the percentages of EXC, RES, and OM fractions did not show significant change with incubation time. However, the percentage of OX Pb increased from 29.2 to 74.9%, and the percentage of CAR Pb decreased from 58.3 to 3.7% during the incubation time from 0 to 35 days and remained constant with longer incubation time (Figure 4(B)). With the increase in incubation period, more organic matters in paper sludge would be transformed into humus by microbes, an adsorbent that can be adsorbed on the surface of soil colloid easily, which may, in turn, result in more adsorption sites of the soil medium. This mineralization and humification processes could ultimately lead to the change of metal fraction in soil.[36,37] As a result, the structure and property of the organic matters in paper sludge changed with

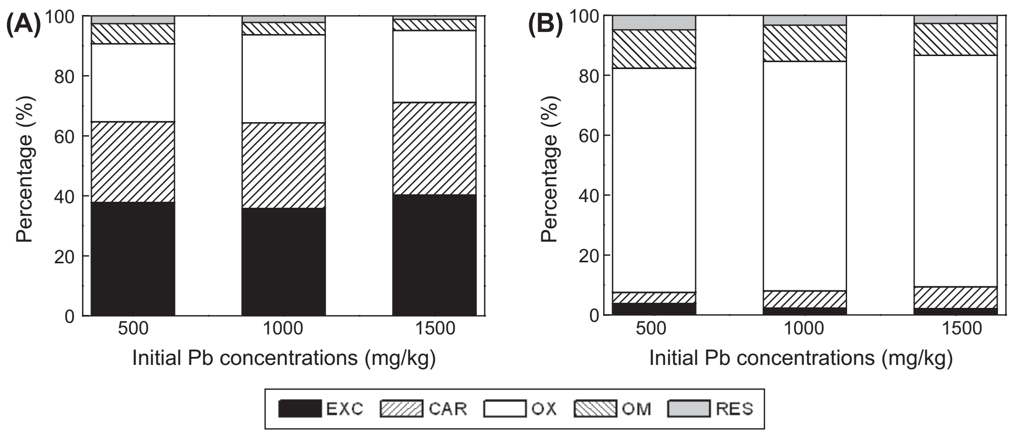


**Figure 2.** The percentage of EXC, CAR, Fe–Mn OX, OM, and RES Pb in the total Pb of the soil with different initial Pb concentrations and with the ratios of soil to paper sludge of 9:0 (A) and 9:8 (B) (by weight). The soils were incubated for 35 days.

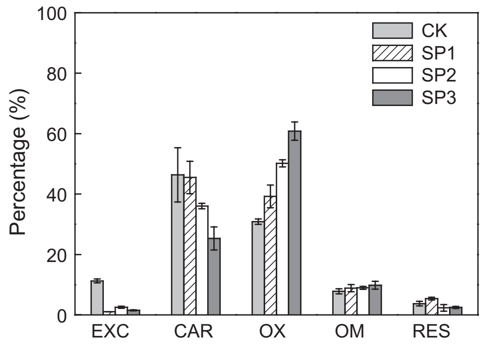


**Figure 3.** The percentage of EXC, CAR, Fe–Mn OX, OM, and RES Pb in the total Pb of the soil (initial Pb concentration of 1500 mg $kg^{-1}$) with 55-day incubation time and different mixture ratios of soil to paper sludge by weight: CK (9:0), SP1 (9:1), SP2 (9:4), and SP3 (9:8).

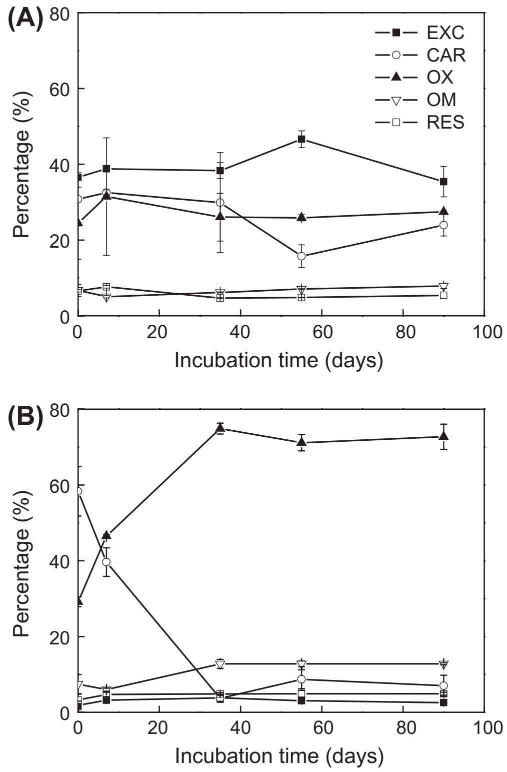


**Figure 4.** The percentage of EXC, CAR, Fe–Mn OX, OM, and RES Pb in the total Pb of the soil (initial Pb concentration of 500 mg $kg^{-1}$) with different incubation time and with the ratios of soil to paper sludge of 9:0 (A) and 9:8 (B) (by weight).

incubation time, which led to the redistribution of Pb speciation in soil and ultimately inhibited Pb uptake to increase the plant yield.

## 4. Conclusions

The effect of paper sludge on the bioavailability and distribution of Pb in soil was investigated. In pot experiment with rape, the addition of paper sludge into the Pb-contaminated soil increased the weights of shoots and roots and reduced the contents of Pb in shoots and roots. Pb in soil was distributed from the liable fractions to OX fraction after the paper sludge addition. The addition of paper sludge increased the content of organic matters and pH of soil and induced Pb redistribution to decrease Pb bioavailability. Our results demonstrated that paper sludge was a potential amendment for Pb-contaminated soils, and it could be used as an amendment for soils contaminated with other heavy metals.

## Disclosure statement

No potential conflict of interest was reported by the authors.

## Funding

The authors are grateful for the financial support from the Major Program of the National Natural Science Foundation of China [grant number 91434132]; National Natural Science Foundation of China [grant number 51378020 and 21077001]; the Program for New Century Excellent Talents in University of Ministry of Education of China [NCET-10-0208]; and the Collaborative Innovation Center for Regional Environmental Quality.

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