

## Single-Step Extraction to Determine Soluble Lead Levels in Soil

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**ABSTRACT:** This study investigated single-step extraction using EDTA to extract the amount of potentially soluble Pb in soil, which would be almost as much as that in the first four Pb fractions of Tessier's sequential extraction procedure (SEP), with or without the immobilization amendment hydroxyapatite under different extraction conditions such as concentration and extractant pH. The results clearly showed that 0.1M EDTA (pH 7.5) with a soil/extractant ratio of 1:100 can extract almost 90% of the first four Pb fractions of SEP, and EDTA-extracted Pb was positively correlated with the first four Pb fractions even in Pb-immobilized soil. In conclusion, single-step extraction using EDTA was found to be a useful method to evaluate potentially soluble Pb even in Pb-immobilized soils.

**Keywords:** EDTA, Immobilization, Pb-Contaminated Soil, Potentially Soluble Pb, Single-Step Extraction

### 1. INTRODUCTION

Heavy metal contamination, which is common in soil throughout the world, poses a potential risk to human and animal health owing to migration through soil profiles into groundwater. Therefore, various remediation techniques have been applied to reduce the mobility and bioavailability of heavy metals in soil.

To effectively remediate heavy-metal-contaminated soil, it is necessary to know the amount of toxic elements in the soil. However, only determining the total concentration of heavy metals in soil is not sufficient because mobility and bioavailability are strongly dependent on the chemical phases of heavy metals in soil [1], [2]. Therefore, determining the chemical forms of such heavy metals is especially important for chemical remediation such as in-situ immobilization and ex-situ soil-washing techniques. Heavy metals in soil exist in various chemical forms including exchangeable, inorganic such as carbonates, bound to Fe/Mn oxide and organic matter, and retained in a clay mineral. Heavy metals bound to Fe/Mn oxide and organic matter, which are less mobilizable than exchangeable heavy metals, have the potential to solubilize with changes in soil conditions such as pH and oxidation-reduction. Therefore, to remediate contaminated soil, especially by using chemical remediation techniques, it is necessary to evaluate the amount of potentially soluble heavy metals including heavy metals bound to Fe/Mn oxide and organic matter as well.

A sequential extraction procedure (SEP) has been widely used to analyze the chemical phases of heavy metals in soil. Tessier's SEP divides the chemical phases into five fractions: exchangeable, carbonates, Fe/Mn oxide, organic matter, and residual [3]. The residual fraction should only be insoluble under natural environmental conditions, which suggests that the first four SEP fractions are possibly being solubilized by ion-exchange and sorption/dissolution onto soil particles. However, SEP is not suitable for analyzing a large number of different contaminated soils because it is time consuming and the analytical technique requires

special facilities.

Ethylenediaminetetraacetic acid (EDTA) is the most widely used chelating agent owing to its ability to form strong chelate complexes with many heavy metals. The extraction efficiency of EDTA, which is closely dependent on many factors of extractant such as its concentration and pH, and the types of contaminated soil [4]-[6], has been extensively studied to predict the mobility and bioavailability of heavy metals in contaminated soil [7], sediments [8]-[10], and wastes [7], [8] as well as to improve the efficiency of soil-washing techniques [6], [11]-[13]. However, only a few studies have compared the amount of heavy metal extracted with EDTA and the amount of each fraction of SEP [8]. It has not been determined whether EDTA can extract the first four fractions of Tessier's SEP. Moreover, EDTA extraction has never been applied to contaminated soil after the addition of an immobilization amendment that would alter the chemical phases of heavy metals in soil. To assess the effectiveness of the chemical remediation technique applied, it is important to be able to determine the amount of potentially soluble heavy metals in soil after chemical remediation.

The present study focused on Pb, which is one of the most toxic and prevalent heavy metal contaminants in Japan and throughout the world, and investigated single-step procedure using EDTA to extract potentially soluble Pb from soil. We speculated that the amount of potentially soluble Pb would be almost the same as the amount of Pb extracted in the first four fractions of Tessier's SEP. In conjunction with potentially soluble Pb, we also investigated whether the same result could be obtained after an immobilization amendment was added.

### 2. MATERIALS AND METHODS

#### 2.1 Study area

This study was conducted at two shooting ranges: Tajimi and Nakatsugawa located in Gifu, Japan. The mean annual precipitation and temperature of both shooting ranges are 2200mm, 14°C, respectively and this area lies between 35° 20' 8" and 35° 28' 6" N and 137° 06' 5" and 137° 29' 2" E.

## 2.2 Soil Preparation and Characterization

The Pb-contaminated soils were collected from depths of 5–15 cm at both shooting ranges. At Tajimi, seven soil samples were collected at 3-m intervals from the shooting point where shooter fires a bullet toward the shooting direction. Table I shows the chemical properties of the Pb-contaminated soils collected at Tajimi (t) and Nakatsugawa (n). Sample t-1 was collected closest to the shooting point; sample t-7 was collected at a point farthest from the shooting point. The Tajimi samples used for chemical analysis were air-dried and passed through a 2-mm sieve without adding any immobilization amendments. One soil sample (n-1) was collected at Nakatsugawa. Hydroxyapatite (hereafter, HAp) was added to sample n-1 at a ratio of 1, 2.5, and 10% (w/w), hereafter, n-2, n-3, and n-4, respectively. After adding HAp, water was added to maintain a water holding capacity of 60%, and then the samples were incubated for 1 week. After incubation, the Nakatsugawa samples were also air-dried and passed through a 2-mm sieve prior to chemical analysis.

The water-extracted Pb and total Pb, Fe, Mn, and C contents of the Tajimi and Nakatsugawa soil samples were determined. The water-extracted Pb was determined by extraction with water at soil/water ratio 1:10. To obtain the total Pb, Fe, and Mn contents in soil, the soil samples were digested with HNO<sub>3</sub> and HCl using a microwave oven. All solutions were filtered with a 0.45-µm filter and analyzed for element concentrations by ICP-OES (ULTIMA 2, Horiba). The total C content in the soil was determined by CHO analyzer (JM10, J-SCIENCE LAB). These analyses were not performed on samples n-2, n-3, and n-4.

## 2.3 Determination of Lead Phases in Soil by Tessier's SEP

SEP was performed on the soil sample following the procedure described in [3]. 1.0 g of soil that has been passed through a 0.425-mm sieve was extracted with 25 mL of a 1 M MgCl<sub>2</sub> solution (exchangeable Pb). The soil remaining after the first extraction procedure was extracted with 25 mL of a 1 M sodium acetate solution with pH 5 (Pb carbonate). The soil remaining after the second extraction was further extracted with 20 mL of 0.04 M NH<sub>2</sub>OH-HCl in

25% (v/v) HOAc in a 95 °C water bath with occasional agitation (Pb bound to Fe/Mn oxide). The soil remaining after the third extraction was extracted with 3 mL 0.02 M HNO<sub>3</sub> and 5 mL 30% H<sub>2</sub>O<sub>2</sub> solution in an 85 °C water bath with occasional agitation. After 3 h of extraction, 5 mL 3.2 M NH<sub>4</sub>OAc in 20% (v/v) HNO<sub>3</sub> was added and the soil was shaken for 20 min (Pb bound to organic matter). The remaining soil was digested with 5 mL HNO<sub>3</sub> and 2 mL HCl using a microwave oven (residual Pb) and diluted in a 50 mL measuring cylinder. After digestion was complete, the solution was passed through a 0.45-µm filter and analyzed for Pb concentration by ICP-OES.

## 2.4 Single-Step Extraction using EDTA

The effects of EDTA concentration, extractant pH, and soil/extractant ratio on the amounts of Pb extracted from soil were investigated using t-1, t-2, t-3, t-5, and t-7 to evaluate the amount of potentially soluble Pb by single-step extraction (SSE) with EDTA.

### 2.4.1 Concentration

10 mL of 0.5, 0.1, and 0.05 M EDTA (pH 7.5) was added to a 15 mL polypropylene tube containing 1.0 g soil samples t-1, t-2, t-3, t-5, and t-7 (1:10 soil/extractant ratio), and then, the tube was shaken for 1 h.

### 2.4.2 pH

10 mL of 0.1 and 0.05M EDTA adjusted to pH 3.5 using 1M HCl was added to a tube containing 1.0 g soil (1:10 soil/extractant ratio), and then, the tube was shaken for 1 h.

### 2.4.3 Soil/Extractant Ratio

100 mL of 0.1 and 0.05M EDTA (pH 7.5) was added to a 100mL tube containing 1.0 g soil (1:100 soil/extractant ratio), and then, the tube was shaken for 1 h.

After shaking, all solutions were passed through a 0.45-µm filter and analyzed for Pb concentration by ICP-OES. All experiments were performed in triplicate, and the average values were calculated.

The extraction ratio in this study, defined as the ratio of EDTA-extracted Pb to the first four fractions of Tessier's SEP, was calculated according to the following equation:

**Table I** Chemical properties of Pb-contaminated soils

No.	Sampling location	Amendment (Type/ratio(w/w))	Water-extracted Pb (mg/kg)	Total			
				Pb (mg/kg)	C (g/kg)	Fe (g/kg)	Mn (g/kg)
t-1	Tajimi	-	0.4	91	8.8	35.8	1.7
t-2	Tajimi	-	0.5	229	19	32.6	1.6
t-3	Tajimi	-	0.7	1551	79	25.8	1.2
t-4	Tajimi	-	0.7	986	37	28.3	1.0
t-5	Tajimi	-	1.0	1341	58	21.0	1.0
t-6	Tajimi	-	1.3	2060	174	13.8	1.0
t-7	Tajimi	-	3.8	9289	135	24.8	6.4
n-1	Nakatsugawa	-	20.8	3966	6.0	30.4	6.0
n-2	Nakatsugawa	HAP/1%	N.A. <sup>*1</sup>	N.A.	N.A.	N.A.	N.A.
n-3	Nakatsugawa	HAP/2.5%	N.A.	N.A.	N.A.	N.A.	N.A.
n-4	Nakatsugawa	HAP/10%	N.A.	N.A.	N.A.	N.A.	N.A.

\*1 Not Analyzed

$$R\% = \frac{Pb_{SSE}}{Pb_{SEP}} \times 100 \quad (1)$$

where  $Pb_{SSE}$  (mg/kg) is the amount of EDTA-extracted Pb and  $Pb_{SEP}$  (mg/kg) is the amount of the first four fractions of Tessier's SEP.

EDTA-extracted Pb of the t-4, t-6 samples was determined by extraction with 0.1M EDTA (pH 7.5) at a soil/extractant ratio of 1:100 following the procedure described above.

### 2.5 Single-Step Extraction using EDTA on Immobilized Soil

Based on the results obtained in the procedures described in Section 2.4, the extraction with 0.1M EDTA (pH 7.5) at a soil/extractant ratio of 1:100 would be suitable to predict potentially soluble Pb in soil. Moreover, the suitability of SSE for Pb-immobilized soils amended with HAp, which would gradually alter the amount of the residual Pb fraction in SEP, was investigated following the procedure described above.

## 3 RESULT AND DISCUSSION

### 3.1 Soil Chemical Properties

The water-extracted Pb and total Pb, C, Fe, and Mn contents in the soil used in this study are shown in Table I. The amount of the water-extracted Pb in Tajimi soil tended to increase with the distance from the shooting point. The amount of the water-extracted Pb in both Tajimi and Nakatsugawa soils exceeded environmental standards for soil contamination in Japan. The total Pb content in Tajimi soil was the lowest for t-1 and then increased gradually with the distance from the shooting point. This increase in the total Pb content might be attributed to the different number of Pb bullets that landed on the soil. From the evidence that the total Pb content in t-7 exceeded 9000 mg/kg, it was considered that heavy Pb contamination has occurred in Tajimi soil. The total C contents in t-6 and t-7 (135–174 mg/kg) were higher than those in t-1 to t-5 (8.8–79 mg/kg). The total Fe content ranged from 13.8 to 35.8 mg/kg. The total Mn content in t-7 was 6.4 mg/kg and was higher than that in the other samples. The water-extracted Pb and total Pb content in Nakatsugawa soil were 20.8 and 3966 mg/kg, respectively. The total Pb content was lower than that in t-7, whereas the water-extracted Pb was higher. This probably resulted from the difference between the Pb phases in Tajimi and Nakatsugawa soils. This result will be discussed in greater detail in the following section.

From these results, it was confirmed that the soils used in this study had different chemical properties such as water-extracted Pb and total Pb, C, Fe, and Mn contents.

### 3.2 Lead Phases in Soil by Tessier's SEP

Figs. 1 and 2 show the results of Pb fractions in Tessier's SEP for Tajimi and Nakatsugawa soils, respectively. In both figures, the upper and lower graphs show the amount and ratio of the Pb fraction, respectively. The average recovery ratio, defined in this study as the ratio of the total amount from SEP to the total amount of Pb amounts in the soil shown in Table I, was  $110 \pm 22\%$ .

The ratio of the Pb fraction in Tajimi soil was the highest in

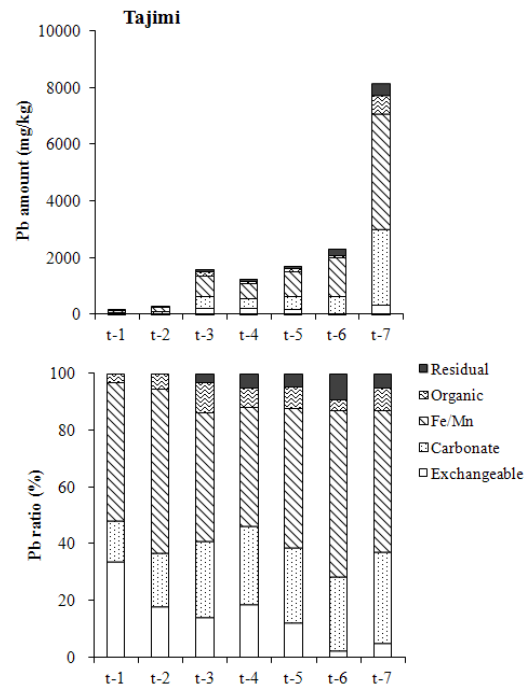


Fig. 1. Sequential extraction of Pb from Tajimi soil

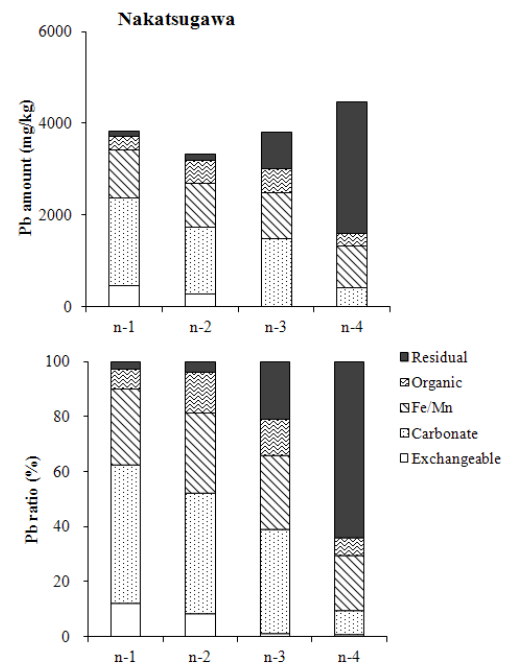


Fig. 2. Sequential extraction of Pb from Nakatsugawa soil.

the fraction bound to Fe/Mn oxide and ranged from 42 to 59%; the second highest was in carbonate ranged from 15 to 32%. In contrast, the highest ratio of Pb fraction in Nakatsugawa soil was carbonates; it was around 50% in n-1 with no added immobilization amendments. The amount and ratio of exchangeable Pb in n-1 were 460 mg/kg and 12%, respectively, although those in t-1 were 355mg/kg and 4%, respectively. In spite of a lower total Pb content in n-1 than in t-7, the water-extracted Pb in n-1 was higher than that in t-7, which probably resulted from the greater

**Table II** EDTA-extracted Pb and extraction ratio under different conditions.

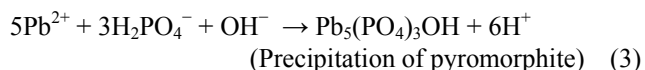
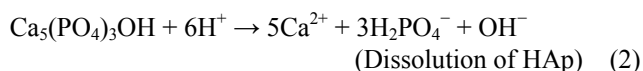
	Concentration	pH	S/E ratio	EDTA-extracted Pb					Ave.	S.D.	C.V.
				t-1	t-2	t-3	t-5	t-7			
				----- (mg/kg) -----							
EDTA-extracted Pb (a)	0.5 M	7.5	1:10	43	175	1029	1096	6552			
	0.1 M	7.5	1:10	59	203	1214	1554	8747			
	0.05 M	7.5	1:10	68	198	1271	1375	7713			
	0.1 M	3.5	1:10	68	210	1342	1650	8253			
	0.05 M	3.5	1:10	83	245	1466	1797	8493			
	0.1 M	7.5	1:100	138	365	1130	1100	6796			
	0.05 M	7.5	1:100	141	180	755	1040	7501			
First four fractions of SEP. <sup>*1</sup> (b)				156	268	1529	1631	7705			
Extraction ratio <sup>*2</sup>	0.5 M	7.5	1:10	27	65	67	67	85	62.4	21.3	34.1
	0.1 M	7.5	1:10	38	76	79	95	114	80.4	28.0	34.8
	0.05 M	7.5	1:10	43	74	83	84	100	77.0	21.0	27.3
	0.1 M	3.5	1:10	44	79	88	101	107	83.6	25.0	29.9
	0.05 M	3.5	1:10	53	92	96	110	110	92.2	23.3	25.3
	0.1 M	7.5	1:100	88	137	74	67	88	90.9	27.1	29.8
	0.05 M	7.5	1:100	90	67	49	64	97	73.6	19.8	26.9

\*1: Pb amount of first four fractions of SEP

\*2: (b)/(a)\*100

amount of exchangeable Pb in n-1 than in t-7.

The residual fractions in n-3 and n-4, where HAp was applied at 2.5% and 25% (w/w), respectively, were significantly increased, although the composition of the Pb fraction in n-2, where HAp was applied at 1% (w/w), was not significantly different from that in n-1. It is widely known that adding HAp to Pb-contaminated soil probably results in the precipitation of Pb phosphate minerals such as pyromorphite (Pb<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>X: X = OH, Cl, F) by immobilizing Pb in the contaminated soil according to the following reactions [14], [15]:



Pyromorphite has a lower solubility product constant at log  $K_{sp} = -25.05$  [16] and is probably included in the residual fraction of SEP. From this consideration, pyromorphite might be precipitated by HAp to increase the residual fraction in n-3 and n-4. On the other hand, the low percentage of HAp added in n-2 probably caused no difference in the Pb composition between n-1 and n-2.

From these results, it was confirmed that the soils used in this study had significant differences in the amount and composition of Pb phases.

### 3.3 Single-Step Extraction by EDTA

#### 3.3.1 Concentration

The EDTA-extracted Pb and extraction ratio are shown in Table II. The extraction ratios for all concentrations tended to be lower at distances closer to the shooting point. It is widely accepted that EDTA forms stable complexes with many heavy metals. Therefore, EDTA can extract Pb

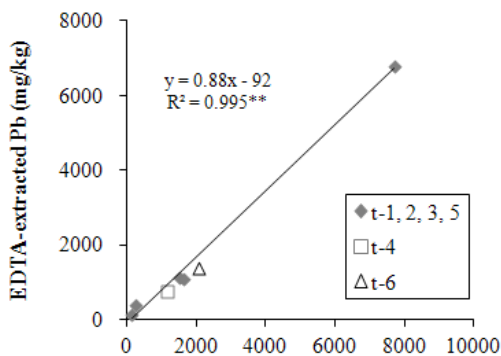
sorbed to Fe/Mn oxide and organic matter in soil as well as inorganic Pb compounds such as Pb carbonate. However, Palma and Mecozzi [10] reported that it was more difficult to extract the exchangeable form compared with other phases. Hence, the extraction ratio in Tajimi soil might be decreased at distances closer to the shooting point owing to a higher ratio of exchangeable Pb. In addition, the extraction ratio for 0.5M EDTA was not higher than that for 0.1 and 0.05M EDTA. According to Elliott and Brown [5], the extraction ratio would not become greater above a certain concentration. Moreover, Finzgar and Lestan [6] revealed that EDTA extraction efficiencies decreased as the molar ratio of EDTA to total Pb in soil increased. From these results, it was considered that 0.1 and 0.05M EDTA should be suitable for the extraction of potentially soluble Pb in soil. Therefore, 0.5M EDTA was not used in the following investigation.

#### 3.3.2 pH

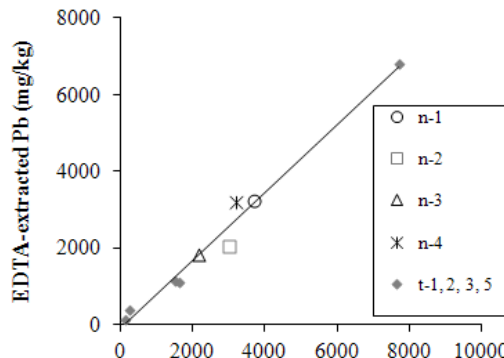
SSE using EDTA was performed again with pH changed to 3.5 (Table II). Although compared with that at pH 7.5, the extraction ratio at pH 3.5 was improved for all samples, the ratio especially in t-1 was still low at 44% and 53% for 0.1 and 0.05M EDTA (pH 3.5), respectively. To perform a simplified analysis rapidly in the field, it is certainly recognized that a neutral pH range is suitable for extractant pH. From these results, it was proposed that an extractant pH of 7.5 would be suitable.

#### 3.3.3 Soil/Extractant Ratio

SSE using EDTA was performed again with the soil/extractant ratio changed to 1:100 (Table II). Compared with the results obtained above, the extraction ratio for 0.1 and 0.05M EDTA (pH 7.5) was improved even in t-1 at 88% and 90%, respectively. The average extraction ratios of five samples were 90.9±27.1 and 73.6±19.8% for 0.1 and 0.05M EDTA (pH 7.5), respectively, and it appears that



**Fig. 3.** Relationship between first four Pb fractions of SEP and EDTA-extracted Pb in Tajimi soil.



**Fig. 4.** Relationship between first four Pb fractions of SEP and EDTA-extracted Pb in Nakatsugawa soil.

**Table III** EDTA-extracted Pb and extraction ratio in Nakatsugawa soil

No.	EDTA-extracted Pb (a)	First four fractions of SEP <sup>*1</sup> (b)	Extraction ratio <sup>*2</sup>
	----- (mg/kg) -----	----- (mg/kg) -----	(%)
n-1	3246	3701	88
n-2	2039	2996	68
n-3	1835	2151	85
n-4	3190	3183	100
Ave.			85.3
S.D.			13.2
C.V.			15.5

\*1: Pb amount of first four fractions of SEP

\*2: (b)/(a)\*100

0.1M EDTA (pH 7.5) with soil/extractant ratio 1:100 may extract potentially soluble Pb in soil. This extraction condition is consistent with that for the soil-washing technique proposed by Garrabrants and Kosson [7].

Fig. 3 shows the relationship between the amounts of 0.1M EDTA-extracted Pb with pH 7.5 and soil/extractant ratio 1:100 and the amount of the first four Pb fractions of SEP. The relationship calculated using the software JMP 8 could be expressed as follow:  $y = 0.88x - 92$   $R^2 = 0.995^{***}$ , where y (mg/kg) is the total Pb in the first four fractions of SEP and x (mg/kg) is the EDTA-extracted Pb. The slope of the line was 0.88, suggesting that 0.1M EDTA can extract almost 90% of the first four Pb fractions of SEP.

The EDTA extraction was performed for the t-4 and t-6 samples under the same conditions, and the results could be represented by the equation presented above (Fig. 3). These results indicated that 0.1M EDTA (pH7.5) with soil/extractant ratio 1:100 can extract almost 90% of the first four Pb fractions of SEP.

### 3.4 Single-Step Extraction of Pb from Immobilized Soil with EDTA

Immobilized Nakatsugawa soil samples were extracted with 0.1M EDTA (pH 7.5) with soil/extractant ratio 1:100 (Table III). The average extraction ratio was  $85.3 \pm 13.2\%$  regardless of the amount of HAP amended and was approximately the same as that for Tajimi soil. If the results of the Nakatsugawa soil samples are plotted in the same manner as shown in Fig. 3, the amounts of EDTA-extracted Pb can also be represented by the same equation for the Tajimi soil samples (Fig. 4). These results suggested that it would be possible to extract potentially soluble Pb in spite

of different kinds of soils and the addition or non addition of immobilization amendment. In addition, pyromorphite, which would be precipitated with the addition of HAp, would remain in soil without dissolution and/or complexation with EDTA owing to its very low solubility. In this study, we have used only two kinds of soils and one type of amendment. Further investigation should consider more types of soil and amendment. Moreover, it is desirable to improve the extraction ratio up to approximately 100%. However, this study did suggest that it would be possible to extract potentially soluble Pb even in immobilized soil with EDTA.

### 4 CONCLUSION

This study investigated single-step extraction using EDTA of potentially soluble Pb that could be almost as much as the amount of Pb in the first four fractions of Tessier's SEP. In conjunction with potentially soluble Pb, we also investigated whether the same result could be obtained after the addition of an immobilization amendment. The results obtained in this study were mainly as follows:

- 1) EDTA extraction under different extraction conditions from soils having different total Pb amounts and different Pb phase compositions was investigated. The results clearly showed that 0.1M EDTA (pH 7.5) with soil/extractant ratio 1:100 can extract almost 90% of the first four Pb fractions of SEP, and EDTA-extracted Pb was positively correlated with the first four Pb fractions of SEP.
- 2) The soils with immobilized Pb using HAp were extracted using 0.1M EDTA (pH 7.5) with soil/extractant ratio 1:100. The results showed that the amount of EDTA-extracted Pb

was also almost 90% of the amount of the first four Pb fractions and could be represented by the same equation as that of the non amended soils.

3) In conclusion, single-step extraction using EDTA was found to be a useful method to evaluate potentially soluble Pb even in Pb-immobilized soils.

## 5 ACKNOWLEDGMENT

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