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Pollution potential leaching index as a tool to assess water leaching risk of arsenic in excavated urban soils



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ABSTRACT

Leaching of hazardous trace elements from excavated urban soils during construction of cities has received considerable attention in recent years in Japan. A new concept, the pollution potential leaching index (PPLI), was applied to assess the risk of arsenic (As) leaching from excavated soils. Sequential leaching tests (SLT) with two liquid-to-solid (L/S) ratios (10 and 20 L kg⁻¹) were conducted to determine the PPLI values, which represent the critical cumulative L/S ratios at which the average As concentrations in the cumulative leachates are reduced to critical values (10 or 5 μ g L⁻¹). Two models (a logarithmic function model and an empirical two-site first-order leaching model) were compared to estimate the PPLI values. The fractionations of As before and after SLT were extracted according to a five-step sequential extraction procedure. Ten alkaline excavated soils were obtained from different construction projects in Japan. Although their total As contents were low (from 6.75 to 79.4 mg kg $^{-1}$), the As leaching was not negligible. Different L/S ratios at each step of the SLT had little influence on the cumulative As release or PPLI values. Experimentally determined PPLI values were in agreement with those from model estimations. A five-step SLT with an L/S of 10 L kg^{-1} at each step, combined with a logarithmic function fitting was suggested for the easy estimation of PPLI. Results of the sequential extraction procedure showed that large portions of more labile As fractions (non-specifically and specifically sorbed fractions) were removed during long-term leaching and so were small, but non-negligible, portions of strongly bound As fractions.

1. Introduction

Arsenic (As) contamination of soil is a serious problem around the world. There have been extensive studies of As mobility in anthropogenically induced contaminated soils (e.g., Ash et al., 2016; Li et al., 2014; Liao et al., 2016), but very few deal with As mobility in deep soils with relative low levels of As. Urban construction projects require excavation of large amounts of soils from great depths (Katsumi, 2015). In most instances, excavated deep soils have not been affected by human activities and the As contents are mostly close to its background values. As a result, its negative environmental impact is easily neglected because of the perception that As in excavated soils occurs at safe levels (Tabelin et al., 2014a); however, such soils may pose a potential threat to the environment because excavation-induced changes in environmental conditions may lead to enhanced As release (Molinari et al., 2013; Tabelin et al., 2012a, 2012b). Excavated soils with relatively low levels of As have now become of great concern in Japan, as reflected by

on the Soil Contamination Countermeasures Law (SCCL) (Ministry of the Environment Government of Japan) (Tabelin et al., 2014a). One environmental standard, known as the Soil Leachate Standard (SLS), is stipulated in SCCL based on the risk associated with ingestion of groundwater. The SLS is determined by deionized water extraction; and for As, the SLS has a critical value of $10 \,\mu g \, L^{-1}$. In Japan, although excavated soils contain low As contents, they are still considered contaminated if the As leaching concentrations are higher than the SLS (Tabelin et al., 2012a, 2012b). After excavation, land owners should therefore take necessary measures to mitigate the potential release of As from these soils; however, whether this method is suitable for assessing the As contamination in excavated soils is still under discussion (Katsumi, 2015). Accordingly, studies of leaching potential and release mechanisms of As from excavated soils are urgently needed.

Leaching tests are typically employed to assess the risk of contamination from contaminated soils to the surrounding water environment (e.g., Ash et al., 2016; Cappuyns and Swennen, 2008; Puga et al.,

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2016). Several typical leaching tests (column, tank and shaking experiments) have been developed, in which different volumes of water are brought into contact with the solid materials (Grathwohl and Susset, 2009). The long-term leaching behaviors over extended periods of time are particularly important for assessment of the risks of hazardous trace elements release from soils. Column tests allow for the measurement of time-lapse release of hazardous trace elements (Grathwohl et al., 2009); however, paramount challenges of this method include installation of a column, entailing well-functioning equipment, skilled techniques, and thoroughly standardized operating conditions.

Sequential leaching tests (SLT), which are classified as an intermediate between batch and column tests, are currently being employed to investigate the long-term leaching of hazardous trace elements (Cappuyns and Swennen, 2008; Tabelin et al., 2014b). These tests allow comparison of the results with those of column tests by plotting the concentrations of contaminants against increasing cumulative liquid-tosolid ratios (L/S), instead of time (Grathwohl and Susset, 2009; Li et al., 2016). Although SLT experiments cannot simulate actual field conditions due to factors such as mechanical friction, centrifugation and filtration (Grathwohl and Susset, 2009), their simple design, low cost, short operating time, and easy standardization nevertheless render great advantages. Our previous study investigated the long-term release of As by SLT, which led to proposal of a new concept, the pollution potential index (PPI) (Li et al., 2016). The PPI value is defined as the cumulative L/S ratio at which the average As concentration (the ratio of cumulative released As (mg kg⁻¹) to cumulative L/S value (L kg⁻¹)) in the leachate equals the Japanese environmental standard (10 μ g L⁻¹) in Japan (Li et al., 2016). This concept could provide new insights into the assessment of the pollution threat posed by As leaching; however, the applicability and extensibility of this concept to other excavated soils needs further investigation. Because the initial terminology (pollution potential index (PPI)) did not specifically highlight leaching behavior, it has been changed to the pollution potential leaching index (PPLI). In this study, leaching of As from excavated soils was investigated using SLT. Based on these results, PPLI values were obtained to assess the water leaching risk of As in these soils. In addition, sequential fractionation was used to evaluate changes of As in different fractions after SLT.

2. Materials and methods

2.1. Sample collection and characterization

Ten excavated urban soils (N1–N10) were obtained from construction projects at different sites on Honshu Island, Japan. During construction, the excavated soils were temporarily stored at interim sites before their final disposal. Samples were randomly collected at these sites. After collection, samples were placed in sealed polyethylene bags and transported to the laboratory, where they were air-dried, gently crushed and sieved through a 2 mm sieve to remove gravel. A portion of each < 2 mm air-dried soil was used for analysis of selected properties, while another portion was ground to pass through a 0.15 mm sieve for total content determination and leaching tests. The measurements of selected physicochemical properties are given in Supplementary information and the results are shown in Table 1.

2.2. Sequential leaching tests

Sequential leaching tests, in which the soil is successively extracted by water, were performed with L/S ratios of 10 and 20 L kg⁻¹ at each step (Grathwohl and Susset, 2009). According to Notification No.46 of the Environment Agency (1991), an L/S of 10 (SLT 10) should be used for As leaching. Following this protocol, 2 g of sample was mixed with 20 mL deionized water in a 50 mL polypropylene centrifuge tube. To investigate the effects of L/S on PPLI values, an L/S value of 20 (SLT 20)

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Items		IN	N2	N3	N4	N5	N6	N7	N8	6N	N10
pH (solid: water $= 1:5$)		7.7 ± 0.02	8.5 ± 0.05	9.6 ± 0.05	10.2 ± 0.01	10.0 ± 0.01	11.9 ± 0.03	8.6 ± 0.14	7.7 ± 0.04	8.5 ± 0.01	8.6 ± 0.05
Moisture content (%)		37.6 ± 0.85	35.9 ± 0.13	6.16 ± 0.34	32.5 ± 0.12	6.94 ± 0.54	18.9 ± 0.43	43.2 ± 0.25	2.20 ± 0.09	32.5 ± 0.45	35.4 ± 0.66
Loss on ignition (LOI) (%)		5.02 ± 0.04	4.66 ± 0.03	3.98 ± 0.12	3.74 ± 0.06	3.76 ± 0.10	7.75 ± 0.07	10.3 ± 0.07	3.80 ± 0.01	2.70 ± 0.01	2.90 ± 0.03
Total carbon contents (g kg ⁻¹)		13.0 ± 0.06	15.1 ± 0.11	13.4 ± 0.26	9.80 ± 0.05	13.8 ± 0.15	40.4 ± 0.32	65.0 ± 0.29	14.0 ± 0.17	12.3 ± 0.20	15.6 ± 0.08
Soil texture		Light clay	Light clay	Sandy clay loam	Clay loam	Loam	Sandy loam	Clay loam	Sandy loam	Light clay	Light clay
Particle size distribution (%)	2.0–0.2 mm	14.4	1.50	39.2	4.20	13.9	47.7	4.70	56.4	4.80	19.8
	0.2–0.02 mm	32.0	29.3	30.2	50.5	43.6	29.8	47.4	22.5	32.3	39.8
	0.02-0.002 mm	28.0	37.9	13.9	26.5	28.1	8.90	25.6	9.10	27.7	16.8
	< 0.002 mm	25.6	31.3	16.7	18.8	14.4	13.6	22.3	12.0	35.2	23.6
Chemical composition	Al ₂ O ₃ (wt%)	13.0	11.5	15.4	11.9	14.6	8.30	10.9	10.2	12.1	14.8
	SiO ₂ (wt%)	41.7	38.3	43.2	41.0	47.6	38.0	33.0	49.7	38.0	38.3
	K_2O (wt%)	1.66	2.12	2.58	2.00	4.12	1.42	1.21	2.93	1.85	1.75
	CaO (wt%)	2.30	2.32	5.32	3.72	2.24	18.5	7.36	2.85	2.31	2.02
	TiO ₂ (wt%)	9.13	6.64	8.56	7.41	9.79	7.29	13.4	5.39	7.69	9.62
	MnO (wt%)	0.10	0.03	0.10	0.04	0.05	0.10	0.03	0.10	0.02	0.07
	Fe_2O_3 (wt%)	11.5	8.83	9.26	8.44	69.6	8.32	Ι	4.59	9.40	13.0
	S (wt%)	0.56	0.72	I	I	I	0.50	2.09	I	0.71	1.02
	As $(mg kg^{-1})$	9.23 ± 0.08	9.22 ± 0.39	79.4 ± 1.05	8.81 ± 0.90	6.75 ± 0.44	9.10 ± 0.06	11.7 ± 0.75	9.96 ± 0.52	6.61 ± 0.37	11.4 ± 0.54

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