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Mineralogical and Chemical Properties and Lability of Cadmium in some Soils from Japan and Vietnam

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We collected five Japanese and six Vietnamese soil samples that were polluted with cadmium (Cd) and analyzed them for chemical and mineralogical properties as well as for extractability of Cd in water, 0.01 mol L⁻¹ CaCl₂, 0.1 and 6 mol L⁻¹ HCl. The total Cd content was in a range from 3 to 16 mg kg⁻¹ for the Japanese samples and 0.5 to 1.1 mg kg⁻¹ for Vietnamese samples. The soil samples had mixed clay mineralogy containing both the 1:1 and 2:1 type layer silicate minerals. We noticed no simple relationships among the mode of extractability in four different solutions and soil properties including clay mineralogy. The only exception was the relationship between the effective cation exchange capacity (ECEC) and the extractability for the Japanese samples. The regression analyses suggested that more Cd tends to reside on cation exchange sites of layer silicate minerals as ECEC increases but it does not results in increase of the extractability in the dilute CaCl₂ solution.

INTRODUCTION

High levels of trace metals content are found in many soils and vegetations in areas affected by mining and agricultural activities, emission from metallurgical factories and automobiles. Accurate measurements of the trace metal contents in contaminated soils are important to assess the potential risk of these areas and to select remediation methods. However, total concentration is not a good indicator for metal mobility or bioavailability in soil ecosystems. The measurement of lability or bioavailability of trace metals in soils plays an important role in assessing risks and taking countermeasures to prevent ingestion by plants and animals via food webs (Ross, 1994).

A widely used method for evaluation of bioavailability of heavy metals in soils is the leaching with chemicals, such as weak acids, neutral salts, and chelating agents (Martens, 1968; Lindsay and Norwell, 1978; Haq *et al.*, 1980; Mehlich, 1984; Singh and Narwal, 1984). Both single and sequential extraction procedures have been employed (Beckett, 1989). Although, the sequential extraction provides some useful information about the chemical forms of heavy metals, such as exchangeable, carbonated, oxide-bound and

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organically-bound (Tessier *et al.*, 1979), the procedures are generally tedious and time consuming. The single extraction is more convenient, but the information from it is not satisfactory for evaluating bioavailability of metals.

In the present study, a simple sequential extraction method was devised and applied to characterize the lability of cadmium (Cd) in some polluted soils from Japan and Vietnam. The soils were also analyzed for basic properties including clay mineralogy. The results of the sequential extraction was examined in search of any relationship among the mode of distribution of Cd in various chemical forms and soil properties. If such relationships are present, one can broadly estimate the lability of Cd in a soil from the total Cd content and soil properties, which is available from databases in soil survey institutions.

MATERIALS AND METHODS

Soil samples

Cadmium-contaminated surface soil samples were taken from a 0–10 cm layer of arable lands in Japan and Vietnam (Table 1). The Omuta samples had been received air-born dust containing Cd salts from a neighboring smelting plant. The Kyowa and Kadzuno samples were contaminated by the tailing from metal smelting works located at upstream of the river from which irrigation water was taken. These samples were collected in Japan.

The Thanhtri, Gialam and Donganh samples were collected in the Red river delta in northern part of Vietnam. These soils were contaminated with cadmium mostly through irrigation. The sources of cadmium in the irrigation water were the manufacturers of dry cell, clock and paint, etc. In addition, the application of sediment that was dredged up from the irrigation canal may have contributed the pollution (Huy and Wada, 2004).

Table 1. Selected properties of soil samples.

Sample name	pH	TOC	ECEC						Particle size distribution			
			Exchangeable cations (cmol/kg)	(cmol/kg)	Total Cd	g kg ⁻¹						
		g kg ⁻¹	Ca	Mg	Na	K	mg/kg	Clay	Slit	Sand	>2mm	
Omuta-1	5.81	25.7	7.05	2.15	2.15	1.51	12.87	16.27	275	318	310	98
Omuta-2	5.73		4.20	1.01	0.52	0.55	6.28	7.95	256	285	351	108
Hakuba	5.50	67.4	2.10	1.18	0.11	0.54	3.93	3.06	151	248	321	279
Kyowa	5.59	69.7	4.12	0.94	0.26	0.87	6.19	6.55	316	234	213	237
Kadzuno	5.82	106.5	3.36	0.96	0.25	1.03	5.60	3.40	122	187	249	441
Donganh-1	6.71	16.1	1.47	0.52	0.15	0.46	2.61	0.77	86	142	722	50
Donganh-2	8.60	37.3	10.51	0.86	0.14	0.42	11.94	0.82	108	129	416	348
Donganh-3	6.16	77.5	2.77	0.85	0.17	0.33	4.12	0.52	234	272	418	76
Thanhtri-1	8.34	64.2	10.83	1.26	0.38	0.99	13.45	1.14	297	313	239	151
Thanhtri-2	8.42	95.8	10.13	1.16	0.29	0.74	12.32	1.09	164	262	245	329
Gialam	7.51	51.2	4.91	0.97	0.22	0.37	6.47	0.67	219	89	646	45

Experimental methods

The soil samples were air dried, ground to pass through a 2-mm sieve, homogenized and stored in polyethylene containers. The soil pH (H_2O) was measured at a soil to water ratio of 1 : 2.5 (kg/L) with a glass electrode. Exchangeable cations were extracted with 1 mol L^{-1} ammonium acetate solution following the method described by Wada and Umegaki (2001). Total cadmium was determined with the method developed by Asami and Kato (1977) but after a preliminary digestion of the soil samples with 7% H_2O_2 to remove the humic substance. The atomic absorption spectroscopy was used to determine Cd and exchangeable cations. Total organic carbon was analyzed by Shimadzu TOC-5000A dissolved carbon analyzer. Particle size distribution was determined by sieving and pipette method after removing organic matter with 30% H_2O_2 and dispersion by sonication. The clay fraction ($<2\mu\text{m}$) was separated from the silt and sand fractions following the method described by Hai *et al.* (2002). The mineralogy of sand, silt and clay fractions was analyzed by X-ray diffraction (XRD) with Ni-filtered Cu K_α radiation at a scanning speed of 2°min^{-1} over a 2θ range of 3 to 30° . The XRD analyses were made on air-dried and glycerol-solvated specimens for the Mg-saturated clays and on air-dried and heated (at 300°C and at 550°C for 2 h) specimens for the K-saturated clays (Hai *et al.*, 2002).

To characterize the lability of Cd, the soil samples were extracted successively with water, 0.01 mol L^{-1} CaCl_2 , 0.1 mol L^{-1} HCl and hot 6 mol L^{-1} HCl. A two gram portion of each soil sample and 20 mL of water were placed in a 85-mL polycarbonate centrifuge tube and shaken on a reciprocal shaker for 2 h. The tube was centrifuged and the extract was collected in a polyethylene bottle after filtration. In a similar way, residual soil was extracted successively with 20 mL of 0.01 mol L^{-1} CaCl_2 for 1 h and 0.1 mol L^{-1} HCl for 1 h. The HCl extraction is a modification of the method proposed by Marten, (1968), which was developed for extracting Cu, Zn in contaminated soils. The shaking time was determined after preliminary experiments. After each extraction, the soil samples were rinsed with 10 mL of water by shaking and centrifugation and the supernatant was combined with the extract. All the experiments were carried out in triplicate and the results were averaged.

Finally the residual soil sample after the HCl extraction was transferred to a polytetrafluoroethylene beaker and digested with hot 6 mol L^{-1} HCl following the procedure of Asami and Kato (1977).

The extracts were stored in polyethylene bottles with fitted stoppers at 4°C before analysis. The Cd concentration in the extract was analyzed by atomic absorption spectroscopy after adjusting background HNO_3 concentration to 0.1 mol L^{-1} .

RESULTS AND DISCUSSION

The basic soil properties of Vietnamese and Japanese soil samples were shown in Table 1. The soil pH ranged from 5.5 to 8.4 and the total organic carbon content was in a range from about 27 to 106 g kg^{-1} . The lowest effective cation exchange capacity, i. e. the summed amount of exchangeable cations, was about 2.61 cmol. kg^{-1} for the Donganh1 sample and 13.45 cmol. kg^{-1} for the Thanhtril sample. The range of clay content was also wide, from 85.5 to 316.2 g kg^{-1} . As arable soil, the selected soil samples are quite diverse

in terms of the selected properties.

The total Cd content ranged from 0.67 mg kg⁻¹ for the Gialam sample to 16.27 mg kg⁻¹ for the Omuta-1 sample. The average Cd contents of the earth crust and soil are 0.1 and 0.35 mg kg⁻¹, respectively (Sposito, 1984). Table 1 shows the Cd contents of the used soil samples were well above the averages for soil and earth crust. In general, the Cd content of the Japanese soil samples is significantly higher than that of Vietnamese soil samples. All the Japanese soil samples that were used in the present study were polluted with Cd through mining and smelting activities. Huy and Wada (2004) reported that the major sources of Cd in the Vietnamese soils were irrigation water and irrigation canal sediment that were polluted by Cd-containing wastes from factories. In Japanese soils the major chemical forms of Cd that reached soils would have been soluble Cd-salts, particulates of sulfides. And so would have been in Vietnamese soils. The significant difference in the total Cd content between Japanese and Vietnamese soil samples would simply be due to difference in the amount of Cd-compounds that were incorporated in the soils (Nguyen, 2002).

The X-ray diffraction (XRD) patterns of representative soil samples are shown in Fig. 1, in which the diffractograms are arranged from upper to lower in the following order; Mg-saturated and air-dried, Mg-saturated and glycerol solvated, K-saturated and air-dried, K-saturated and heated at 300°C, K-saturated and heated at 550°C. Numbers indicated at peaks are d-spacings in nm. The diffraction patterns of the Omuta-2 sample resembled those of Omuta-1 sample, the Dongganh-1 and Donganh-2 samples resembled the Donganh-3 sample, the Thanhtri-2 and Gialam samples resembled the Thanhtri-1 samples.

Since the amount of clay samples mounted on the glass slide was approximately the same, the peak intensity in the diffraction patterns may be a measure of the content of crystalline clay minerals. Among the 11 soil samples the heights of peaks in the Kyowa and Kadzuno samples, particularly in the Kyowa sample, were apparently lower than those of any other soil, suggesting that the content of noncrystalline components is higher in these soil samples. These soil samples showed diffraction peaks at 1.4, 1.0 and 0.7 nm irrespective of the treatment and the peak intensity at 0.7 nm decreased significantly after heating at 550°C. In addition the 1.4 and 1.0 peaks poorly resolved and the 1.0 nm peak intensified after 550°C heating with diminish of the peak at 1.4 nm. These characteristics show that the major layer silicate minerals in these soils are vermiculite-chlorite intergrade, illite, kaolin minerals and some chlorite.

On the contrary, the diffraction peaks of the Hakuba sample were the sharpest. It exhibited strong peaks at 1.4, 0.94, 0.7 nm, with an accessory peak at 1.0 nm. For this sample, the intensity of the peaks at 1.4 and 0.7 nm did not change significantly, suggesting that the major clay minerals are chlorite. The characteristic peak at 0.94 nm indicates the presence of talc.

The Omuta-1 sample also showed peaks at 1.0 and 0.7 nm and the 0.7 nm peak diminished significantly after 550°C heating. The 1.4 nm peak in Mg-saturated and air-dried specimen almost completely shifted to 1.7 nm. These indicate that the major layer silicate minerals in the Omuta-1 sample are smectite, illite and kaolin minerals with small amount of chlorite.

The major layer silicate clay minerals are illite, kaolin minerals and some chlorite for

Fig. 1. X-ray diffractograms of clay fraction of sample soils.

Table 2. Summary of mineralogical analyses.

Soil sample	Layer silicate minerals	Active iron content / g kg ⁻¹
Omuta-1	Smectite, Kaolin, Illite, chlorites	4.11
Omuta-2	Smectite, Kaolin, Illite, chlorites	4.40
Hakuba	Chlorite, Talc	2.58
Kyowa	Vermiculite-chlorite intergrade, Illite, Kaolin	6.95
Kadzuno	vermiculite-chlorite intergrade, illite, kaolin	2.12
Donganh-1	Kaolin, Illite and Vermiculite-chlorite intergrade	0.92
Donganh-2	Kaolin, Illite and Vermiculite-chlorite intergrade	4.58
Donganh-3	Kaolin, Illite and Vermiculite-chlorite intergrade	2.73
Thanhtri-1	Illite, Kaolin, Chlorite	4.06
Thanhtri-2	Illite, Kaolin, Chlorite	4.26
Gialam	Illite, Kaolin, Chlorite	17.05

Thanhtri-1 sample, and kaolin mineral, illite and vermiculite-chlorite intergrade for Donganh-3.

The results of the XRD analysis were summarized in Table 2. In addition to the layer silicate clay minerals, most of the samples contained quartz and feldspars, but they are omitted from Table 1 because they are inert with regard to Cd adsorption. Mineralogy of silt and sand fraction was also examined by X-ray diffraction. The major minerals in these fractions were feldspars and quartz and there was no distinctive characteristics. Table 2 also lists the contents of noncrystalline iron oxides and hydroxides that was determined by oxalate-oxalic acid extraction.

The results of the sequential extraction were tabulated in Table 3. The sum of the successively extracted amounts of Cd was fairly close to the amount of total Cd for all soil samples (Table 1), indicating that the magnitude of the experimental errors involved in the multi-step extraction procedure is not serious. On the basis of the data in Table 3, percentage of each fraction was calculated and graphed in Fig. 2. Table 3 and Fig. 2 clearly show that there is a big difference in the lability of Cd between Japanese and Vietnamese soil samples. The percentage of the 0.0 mol L⁻¹ CaCl₂ extractable fraction is in a range from about 2% to 7% for the Vietnamese samples whereas it is from 17% to 38% for the Japanese samples. The trend is quite opposite for the 6 mol L⁻¹ HCl extractable fraction and its percentage was from 53% to 86% for Vietnamese soils and from 8 to 7% for Japanese soil samples. The amount of water-extractable fraction was negligibly low for all soil samples except for the Omuta-1 sample.

The interpretation of the form of the water-extractable Cd is straightforward, whereas it is difficult to relate the CaCl₂ and HCl extractable Cd to specific chemical forms. Generally, heavy metal cations that can be extracted with neutral salt solutions are labeled as exchangeable cations (Tessier *et al.*, 1979). And those extractable in dilute acid are categorized as cations that are surface complexed on oxide minerals. However, recent studies indicated that at least a part of Cd ions that are released from cation exchange site by neutral salts are re-adsorbed on co-existing oxide minerals (Kheboian and Bauer, 1987). Thus, the Cd found in neutral salt extracts is actually a fraction of

Fig. 2. Distribution of water-, 0.01 mol L⁻¹ CaCl₂-, 0.1 mol L⁻¹ HCl- and 6 mol L⁻¹ HCl-extractable Cd in sample soils.

Table 3. Results of sequential extraction.

Sample	Cd extracted by				Total
	Water	0.01 mol L ⁻¹ CaCl ₂	0.1 mol L ⁻¹ HCl	6 mol L ⁻¹ HCl	
	mg/kg				
Omuta1	0.95	2.51	10.68	1.25	15.39
Omuta2	<0.01	2.96	3.95	1.03	7.67
Nagano	<0.01	1.17	1.08	1.32	3.57
Kyowa	<0.01	1.53	3.68	1.21	6.42
Kadzuno	<0.01	1.08	1.70	1.09	3.87
Donganh1	<0.01	0.05	0.11	0.99	1.15
Donganh2	<0.01	0.07	0.08	0.81	0.96
Donganh3	<0.01	0.04	0.09	0.45	0.58
Thanhtri1	<0.01	0.10	0.50	0.88	1.48
Thanhtri2	<0.01	0.05	0.45	0.58	1.08
Gialam	<0.01	0.01	0.13	0.65	0.79

exchangeable Cd retained on permanent negative charges on layer silicate minerals. Similarly the Cd extracted by dilute acid from specifically adsorbing sites on oxide minerals is partly held on layer silicate minerals (Mori and Wada, 2003). Therefore, complete speciation of heavy metals in soils by successive extraction is in a sense an illusion.

The sequential extraction procedure employed in the present study does not aim at chemical speciation of Cd in the soil samples. Extracting solutions and extraction condi-

Fig. 3. Pair-wise scatter plot among fractions of three forms of Cd and pH, Organic carbon content (TOC), effective cation exchange capacity (ECEC) and oxalate soluble Fe (Active Fe).

tions are selected to obtain a measure of lability of Cd under different chemical conditions. The 0.01 mol L⁻¹ CaCl₂ solution has an ionic strength that is prevailing in many arable soils during plant growth. The 0.01 mol L⁻¹ CaCl₂ extractable Cd can be regarded as a measure of lability of Cd under normal soil conditions (Houba *et al.*, 1990). With regard to Cd, the 0.1 mol L⁻¹ HCl is strong enough for quantitative extraction of both the exchangeable and surface complexed Cd (Mori and Wada, 2003) and this fraction would be a measure of Cd susceptible to soil acidification. And it would be reasonable to regard the hot 6 mol L⁻¹ HCl extractable fraction as non-labile Cd in soils.

The percentage of each Cd fraction was plotted against measured values of chemical

and mineralogical properties in Fig. 3. In this plot, the percentages were used as dependent variables because the absolute amount of Cd in soils depends mostly on anthropogenic factors, not on nature of soils. Here, again, there is a big difference in trends between the Japanese and Vietnamese soil samples. As a whole the percentages of three Cd fractions seems rather indifferent to the selected soil properties except for to the ECEC and soil pH.

Due to the limited sample size it is difficult to generalize but the relationships between percentages of three Cd fractions and ECEC seems meaningful particularly for the Japanese soil samples. The percentage of the 6 mol L⁻¹ HCl extractable Cd decreased as the ECEC increased (Fig. 3). This suggests that more Cd is distributed over cation exchange sites on layer silicate minerals as ECEC increases. The percentage of the 0.01 mol L⁻¹ CaCl₂ extractable Cd was also negatively correlated with ECEC for the Japanese soils. Although these trends are confusing, they are reasonable because the 0.01 mol L⁻¹ CaCl₂ solution extract only a part of the exchangeable Cd at the present soil : solution ratio and its extent is expected to inversely correlate with the ECEC. The positive correlation between the percentage of the 0.1 mol L⁻¹ HCl extractable Cd and the ECEC is also understandable because 0.1 mol L⁻¹ of strong acid extracts most part of both the exchangeable and surface complexed Cd (Mori and Wada, 2003).

As already discussed above, the Vietnamese soil samples contained vermiculite–chlorite intergrades. Saha *et al.* (2001) synthesized a hydroxyaluminum–montmorillonite complex and carried out a series of heavy metal adsorption experiments. According to their data, the adsorption percentage for Cd increased as solution pH increased and reached 100% at around pH 7 and the adsorbed Cd was not extracted by washing with a 0.01 mol NaClO₄ solution. The low percentage of the 0.01 mol CaCl₂ extractable Cd in the Vietnamese soil samples may partly due to the adsorption onto the hydroxyaluminum interlayer of the vermiculite–chlorite intergrades.

CONCLUSIONS

The total Cd content was about 3 to 16 mg kg⁻¹ for the Japanese samples and 0.5 to 1.1 mg kg⁻¹ for Vietnamese samples and the extent of pollution is much less in the Vietnamese soils examined. There was also a big difference between the Japanese and Vietnamese soil samples with regard to the extractability of Cd in 0.01 mol L⁻¹ CaCl₂ and 0.1 and 6 mol L⁻¹ HCl. More than 60% of the total Cd was extractable in 0.01 mol L⁻¹ CaCl₂ and 0.1 mol L⁻¹ HCl for the Japanese soils, whereas the percentage of Cd that were extractable in these solution was below 50% for Vietnamese soils. This indicates that Cd in the Vietnamese soils is much less labile in normal and acidified soil conditions. Graphical analyses of the correlation among the extractability of Cd and soil properties including pH, TOC and ECEC indicated that no single property governs the chemical forms and extractability. Contribution of hydroxyaluminum interlayer to the Cd adsorption was suggested but further mineralogical research is needed.

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