

## **Concentration, Soil-to-Plant Transfer Factor and Soil-Soil Solution Distribution Coefficient of Selenium in the Surface Environment -9106**

Shigeo Uchida, Keiko Tagami, Nao Ishikawa

Office of Biospheric Assessment for Waste Disposal, National Institute of Radiological Sciences,  
Anagawa 4-9-1, Inage-ku, Chiba 263-8555, Japan

### **ABSTRACT**

Of the major radioactive selenium isotopes, Se-79, a beta emitter with a half-life of about 1.1 million years, is of special interest because it is one of the most important radionuclides for the long-term dose assessment of radioactive waste disposal. This radionuclide can reach human beings through several transfer paths in the environment. To predict Se-79 behavior from the environment to human beings, it would be useful to obtain the following information: stable Se concentration in environmental samples; soil-soil solution distribution coefficient ( $K_d$ ); and soil-to-plant transfer factor (TF). In the present study, stable Se concentrations in river water, soil and crop samples collected in Japan,  $K_d$ s and TFs were obtained. The results showed that geometric mean (GM) concentrations of river water, soil and crops were 0.057  $\mu\text{g/L}$  (range: <D.L.-1.17  $\mu\text{g/L}$ ), 0.43 mg/kg (range: 0.068-1.56 mg/kg-dry), and 0.015 mg/kg-dry (range: <D.L.-0.24 mg/kg-dry), respectively. GMs of  $K_d$ s for paddy field soil and upland field soil samples were 116 and 67, respectively, whereas GMs of TFs for brown rice and upland field crops were 0.066 and 0.024, respectively. Probably due to longer growing period and different water management in the paddy fields for brown rice compared to those for upland field crops, the TF would be high in brown rice.

### **INTRODUCTION**

Selenium is an essential nutrient for animals; however, at high concentrations the element becomes toxic to animals. Of the major radioactive selenium isotopes, Se-79, a beta emitter with a half-life of about 1.1 million years, is of special interest because it is one of the most important radionuclides for the assessment of radioactive waste disposal in deep underground sites [1, 2]. This radionuclide can reach human beings through several transfer paths in the environment. The principal mechanism for the dispersion of radionuclides from radioactive wastes disposed underground is dissolution of the radionuclides into the ground water. The groundwater would transfer radionuclides to the surface environment, such as river water, where the water would be used as irrigation water for crops and drinking water for human beings and animals. In this way, Se-79 could be taken into the human body by ingestion of food and water.

To predict Se-79 transfer behavior from the environment to human beings, it would be useful to obtain following three information: (1) concentration of stable Se in environmental sample as natural analogue of Se-79; (2) soil-soil solution distribution coefficient ( $K_d$ ); and (3) soil-to-plant transfer factors (TF). Se abundance in the natural environment has been reported [3-7]. Average crustal abundance of Se is 0.05 mg/kg, and its concentration in coal and other organic rich deposits can be high. Se concentrations in soil are typically in the range of 0.01-2 mg/kg with a world average of 0.4 mg/kg [3]. In natural waters, the data on Se concentrations are scarce. The concentration in river water is estimated to be 0.07  $\mu\text{g/L}$  [4] and that in atmospheric precipitations is 0.04-1.4  $\mu\text{g/L}$  [5]. For plant samples, the average concentration in various crop types collected in the USA ranged from 0.015-0.407 mg/kg dry weight [3] and was 0.02

mg/kg dry weight in a reference plant [6]. Most plants contain around 0.025 mg/kg [7]. However, knowledge on mobility in soil and plant uptake behavior of Se in agricultural fields is still limited. In the present study, stable Se concentrations in river water, soil and crop samples collected in Japan, TFs of stable Se and  $K_d$  s of Se-75 were reported.

## EXPERIMENTAL

### River Water Samples

#### *Sample collection*

We selected 45 major Japanese rivers as shown in Fig.1. Only 2-3 days were spent in collection at any one river, because river conditions can be affected by the weather and the season. Ten samples per river were collected from the upper stream to the river mouth. If it rained heavily within 3-5 days before the intended sampling dates, then the sampling was rescheduled to avoid a dilution effect from the rain. However if the water flow did not dramatically increase because only a small amount of rain fell, then we collected samples. Even if it rained on sampling dates, we also collected the samples if the water was not apparently different from its typical condition.



**Fig.1. Forty-five major Japanese rivers.**

The uppermost limit of the upstream river section was selected as being where a car was able to go and the lowermost limit of the upstream river section was selected as 5 to 10 km from the river estuary. Ten sampling points were selected by dividing the distance between the uppermost and lowermost limits into nine equal parts and then finding suitable bridges not far from the selected points. However we avoided sampling of muddy flows, places where another stream or river joined the sampled river and places near drainpipes. At places where a tributary flowed into the river, samples were taken downstream

where river water was well-mixed (at a distance equal to ten times the river width). And when there were several parallel flows, samples were collected in the largest flow. In principle, collection points were selected in the center of the river width and samples were taken from bridges. If a suitable bridge was not found near the collection point, the river water was directly collected while standing on the riverbank.

### ***Analytical methods***

Polypropylene bottles, 500 mL and 100 mL, were used for sampling and storage to avoid sorption of elements on the bottle wall. All bottles were cleanly washed with nitric acid, rinsed with Milli-Q quality water ( $>18\text{ M}\Omega$ ) and finally dried under clean air conditions.

The water temperature, pH, and electric conductivity (EC,  $\mu\text{S}/\text{cm}$ ) were measured on-site. River water samples were collected in the clean-washed 100 mL polypropylene bottles. Each 100 mL bottle was filled with pretreated river water which was later directly used for laboratory measurements of Se by inductively coupled plasma mass spectrometry (ICP-MS). The pretreatment was done on-site as follows. About 150 mL of a collected river water sample were filtered using a disposable filter unit with a  $0.45\ \mu\text{m}$  pore size membrane filter. A small portion of the filtrate was used to wash the sample storage bottle, then the rest of the filtrate, ca 100 mL (weighed sample), was transferred into this washed bottle and 1 mL of concentrated nitric acid (Tama Chemical Industries., TAMAPURE AA-100) was added. After completing the pretreatment, these samples were also transported to our institute under cool condition ( $5^\circ\text{C}$ ).

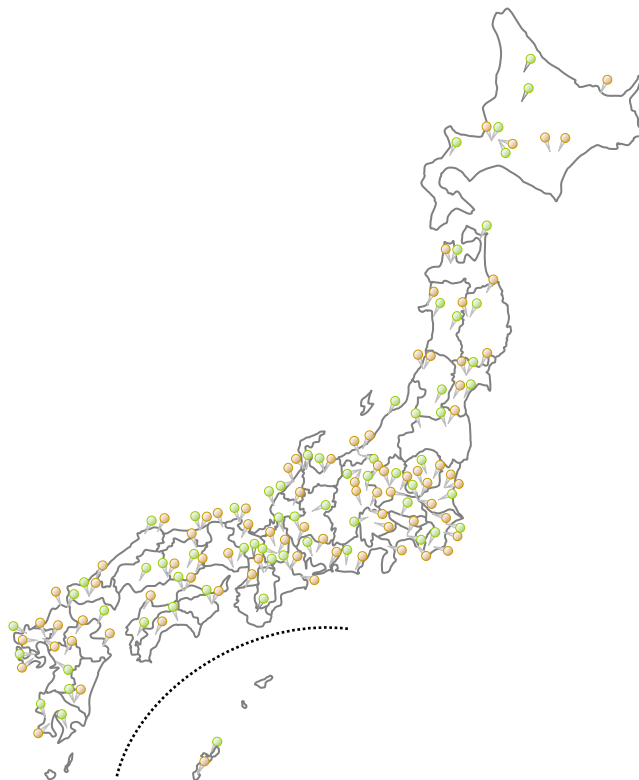
At the laboratory, dissolved amounts of Se were determined by ICP-MS (Yokogawa, Agilent 7500c). Each measurement was made twice on different days and the average value was taken as a quantitative value. We used 2-3 multi-element standard solutions (SPEX, XSTC) and single element standard solutions (Merck, CertiPrep series); they were mixed and diluted with 2%  $\text{HNO}_3$  accordingly to measure target elements. Accuracy of the measurements was confirmed as needed by using standard samples purchased from National Institute of Standards and Technology (NIST) and National Institute for Environmental Studies.

### **Soil and Crop Samples**

#### ***Sample collection and pretreatment***

Seventy-nine upland field and 63 paddy field soil samples (plowed soil layer: up to ca. 20 cm depth), were collected nationwide in 2002-2006 as shown in Fig.2. From one sampling field, 5 sub-samples, approximately 1 kg on fresh weight basis each, were collected in the harvesting season and these sub-samples were mixed well. About 15 kg amounts (on a fresh weight basis) of edible parts of crops were also collected. Crops were 63 brown rice samples (hulled rice), 18 green vegetable samples (cabbage, Chinese cabbage, spinach, lettuce and so on), 11 tuber samples (potato, sweet potato and taro), 12 allium samples (leek and onion), 7 legume samples (bean and pea), 18 fruit vegetable samples (cucumber, tomato, sweet pepper and so on), 10 root vegetable samples (carrot and Japanese radish) and 9 cereal samples (wheat and barley).

Three kg of the soil samples were air-dried and passed through a 2-mm mesh sieve. For crop samples, edible parts were washed with deionized water at least 3 times, and the removal of dust and soil particles was completed. The washed parts were paper-towel dried, chopped and freeze-dried. For leek, green and white parts were separated. Leaves of carrot and Japanese radish are also edible so that roots and leaves were separated and elemental compositions of both of them were measured. Finally, samples of 142 soil samples and 148 crop parts were obtained and they were separately and thoroughly ground into fine powders. The powders were transferred into glass vials and stored at room temperature.



**Fig.2. Soil and crop sampling sites. Green: paddy fields, Orange: upland fields.**

### ***Measurements***

The pH (H<sub>2</sub>O) of the soils was measured at a soil:water ratio of 1:2.5. Cation exchange capacity was determined by the semi-micro Schollenberger's method [8] using 1 mol/L of neutral ammonium acetate. The contents of total carbon were analyzed with CHN analyzer (Euro Vector, EuroEA3000) using 1-1.5 mg of crop samples and 10 mg of soil samples. Electric conductivity (EC) was measured at a soil:water ratio of 1:5. Soil particle size distribution was also analyzed.

For the measurement of Se, samples were prepared as follows. The soil samples, 100 mg each, were digested with mineral acids (a mixture of HNO<sub>3</sub>, HF and HClO<sub>4</sub>) using a microwave digester (CEM, Mars 5). For crop samples, 500 mg amounts were used. To them were added 10 mL HNO<sub>3</sub> and 4 mL HF. The mixtures were heated for 10 h at 80°C to decompose organic matter. After that, microwave digestion was carried out. Following their digestion, both soil and crop samples were evaporated to near dryness at 140°C. The residue was dissolved with 1 mL of conc. HNO<sub>3</sub> and 0.5 mL of H<sub>2</sub>O<sub>2</sub> and evaporated again. Finally, the residue was dissolved with 1 mL of 40% HNO<sub>3</sub> and diluted with deionized water. The digestion samples were made in duplicate. All the acids used were ultra-pure analytical grade (Tama Chemicals, AA-100). Water (>18.1MΩ) which was treated using a Milli-Q water system (Millipore Co.) was used throughout the work.

After diluting the acid solutions to a suitable concentration, elements in both crop and soil samples were measured using ICP-MS. Standard reference materials, such as SRM-1573a (NIST, tomato leaves), GBW-07603 (Institute of Geophysical and Geochemical Exploration, bush twigs and leaves), JB-3 (Geological Survey of Japan, igneous rock) were also analyzed together with the samples to check the accuracy of the method.

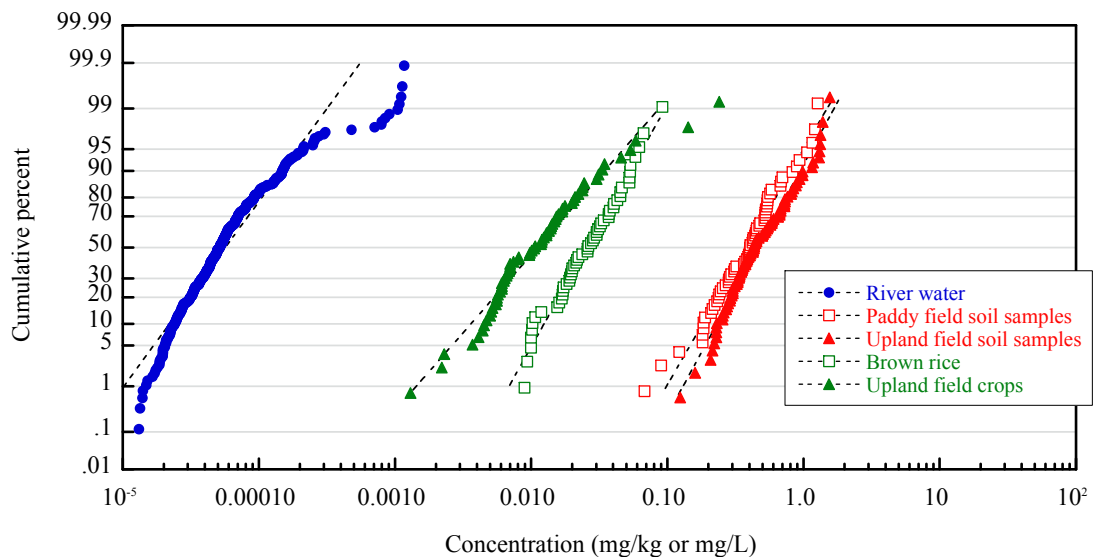
### Batch Sorption Test for $K_d$

The  $K_d$  values of Se were obtained by means of separate batch sorption tests using Se-75 (half-life: 119.8d) as a tracer. Each soil sample and deionized water (solid/liquid ratio, 1 g dry weight per 10 mL or 3g dry weight per 30 mL) [9, 10] were mixed in a plastic bottle, and initially shaken for 24 h at 23°C. A carrier free Se-75 solution (chemical form: selenite) was added to the well-mixed solution. Previously, we had confirmed that sorption equilibrium was reached within 7 days [9]. Thus, all samples were collected after shaking the mixture for 7 days. The suspension was centrifuged at 3000 rpm (Hitachi, Himac CT5L) for 10 min, and the supernatant was filtered through a 0.45- $\mu$ m membrane filter (Millex-LH). Radioactivity of the radionuclide in the filtrate was measured with an NaI scintillation counter (Aloka, ARC-380).

## RESULTS AND DISCUSSION

### Selenium Concentrations in River Water, Soil and Crop Samples

The cumulative probability distributions of Se in river water, soil and crop samples were close to the log-normal lines as shown in Fig.3. Their geometric mean (GM) concentrations were calculated as 0.057  $\mu$ g/L (range: <D.L.-1.17  $\mu$ g/L), 0.43 mg/kg (range: 0.068-1.56 mg/kg-dry), and 0.015 mg/kg-dry (<D.L.-0.24 mg/kg-dry), respectively. World averages for Se in river water, soil, and plants are 0.07  $\mu$ g/L, 0.4 mg/kg and 0.025 mg/kg, respectively; thus, the samples collected in Japan contained almost the same Se concentration as the world average values. Concentrations of Se in river waters at each sampling sites are listed in Table I. The GM concentration in the Abe River was one order of magnitude higher than GM of all samples, but the value was 10 times lower than the WHO international drinking water quality guidelines value for Se of 10  $\mu$ g/L [11]. Concerning the concentration distribution pattern of Se from the upper stream to the river mouth of each river, it was usually narrow (data not shown). Since Se concentration was low in the water samples, the observed concentrations would mainly be originated from weathering of bedrock.



**Fig.3. Probability distributions of Se concentrations in Japanese river water, agricultural fields and crop samples.**

Table I. Concentrations of Se, pH and Electric Conductivity (EC) in River Water Samples

River Name	Sample number of Detected / Used	pH Range	EC (mS/m) Range	Se ( $\mu\text{g/L}$ )	
				Range	Geometric mean
Teshio	10 / 10	6.9 - 7.5	4.0 - 8.2	0.034 - 0.052	0.043
Yubetsu	10 / 10	6.0 - 6.7	3.1 - 6.7	0.025 - 0.066	0.054
Ishikari	10 / 10	6.5 - 7.6	3.1 - 31.0	0.042 - 0.107	0.070
Shiribetsu	10 / 10	6.3 - 7.0	3.3 - 8.4	0.025 - 0.047	0.035
Saru	10 / 10	7.1 - 7.5	5.7 - 10.5	0.028 - 0.068	0.044
Tokachi	10 / 10	6.7 - 7.3	6.0 - 11.0	0.052 - 0.115	0.072
Mabuchi	10 / 10	6.0 - 6.8	5.1 - 9.8	0.035 - 0.060	0.045
Kitakami	10 / 10	6.5 - 6.9	9.2 - 14.7	0.034 - 0.091	0.057
Abukuma	10 / 10	7.5 - 8.0	11.9 - 24.0	0.039 - 0.089	0.070
Yoneshiro	10 / 10	6.5 - 6.9	6.7 - 13.1	0.043 - 0.207	0.096
Omono	10 / 10	6.5 - 7.1	5.2 - 11.5	0.022 - 0.053	0.036
Mogami	10 / 10	4.7 - 7.7	10.5 - 18.7	0.044 - 0.073	0.060
Naka	10 / 10	7.4 - 7.6	13.4 - 17.0	0.081 - 0.144	0.099
Tone	10 / 10	6.6 - 8.8	2.1 - 30.0	0.053 - 0.276	0.136
Ara	9 / 9	7.2 - 8.9	12.8 - 39.0	0.078 - 0.149	0.099
Sagami	10 / 10	7.1 - 9.1	10.1 - 16.1	0.040 - 0.152	0.114
Agano	10 / 10	6.7 - 7.3	5.3 - 11.4	0.020 - 0.046	0.031
Shinano	10 / 10	7.1 - 8.2	4.7 - 19.5	0.022 - 0.092	0.067
Seki	10 / 10	6.5 - 8.7	8.3 - 28.0	0.024 - 0.076	0.043
Shou	10 / 10	6.8 - 7.7	3.0 - 10.9	0.028 - 0.042	0.033
Fuji	10 / 10	6.9 - 7.8	8.5 - 17.5	0.033 - 0.193	0.082
Abe	10 / 10	7.6 - 8.7	17.2 - 25.0	0.707 - 1.170	0.945
Ooi	10 / 10	7.5 - 8.2	8.3 - 15.9	0.092 - 0.308	0.176
Tenryu	10 / 10	7.1 - 9.2	9.4 - 15.7	0.057 - 0.092	0.073
Kiso	7 / 9	7.1 - 8.7	6.0 - 8.3	0.013 - 0.031	0.019
Yura	9 / 9	6.0 - 6.7	4.2 - 14.0	0.018 - 0.057	0.028
Yodo	7 / 7	7.6 - 7.7	18.2 - 22.0	0.046 - 0.060	0.053
Kako	10 / 10	7.1 - 7.6	6.2 - 14.6	0.028 - 0.056	0.048
Kino	10 / 10	7.7 - 8.1	9.9 - 15.8	0.033 - 0.052	0.042
Kumano	10 / 10	6.4 - 6.8	4.6 - 6.0	0.024 - 0.128	0.076
Kuzuryu	10 / 10	6.6 - 7.9	3.0 - 11.4	0.020 - 0.062	0.040
Hii	10 / 10	6.6 - 7.0	4.5 - 9.7	0.020 - 0.036	0.026
Gouno	9 / 10	7.1 - 8.5	7.2 - 12.9	0.014 - 0.023	0.018
Yoshii	9 / 9	7.5 - 7.8	5.4 - 16.4	0.021 - 0.045	0.031
Takahashi	10 / 10	7.3 - 8.8	6.7 - 13.0	0.014 - 0.044	0.025
Oota	0 / 10	6.2 - 7.0	4.0 - 6.2		<D.L.
Yoshino	10 / 10	7.3 - 8.0	4.0 - 10.0	0.022 - 0.064	0.043
Hiji	10 / 10	7.0 - 7.5	10.5 - 16.3	0.055 - 0.085	0.069
Niyodo	10 / 10	7.0 - 7.6	2.8 - 7.5	0.031 - 0.052	0.043
Shimanto	10 / 10	6.4 - 6.7	6.6 - 10.4	0.024 - 0.053	0.029
Onga	10 / 10	7.4 - 8.1	7.9 - 32.0	0.017 - 0.052	0.033
Chikugo	10 / 10	7.4 - 8.2	11.7 - 26.0	0.020 - 0.048	0.028

Kuma	10 / 10	6.2 - 6.9	6.2 - 11.4	0.034 - 0.062	0.048
Ohno	10 / 10	6.3 - 6.9	3.1 - 21.0	0.036 - 0.300	0.144
Ooyodo	10 / 10	6.8 - 8.6	12.9 - 21.0	0.134 - 0.479	0.176
All	430/ 443	4.7 - 9.2	2.1 - 39.0	0.013 - 1.170	0.057

### Soil-Soil Solution Distribution Coefficient ( $K_d$ )

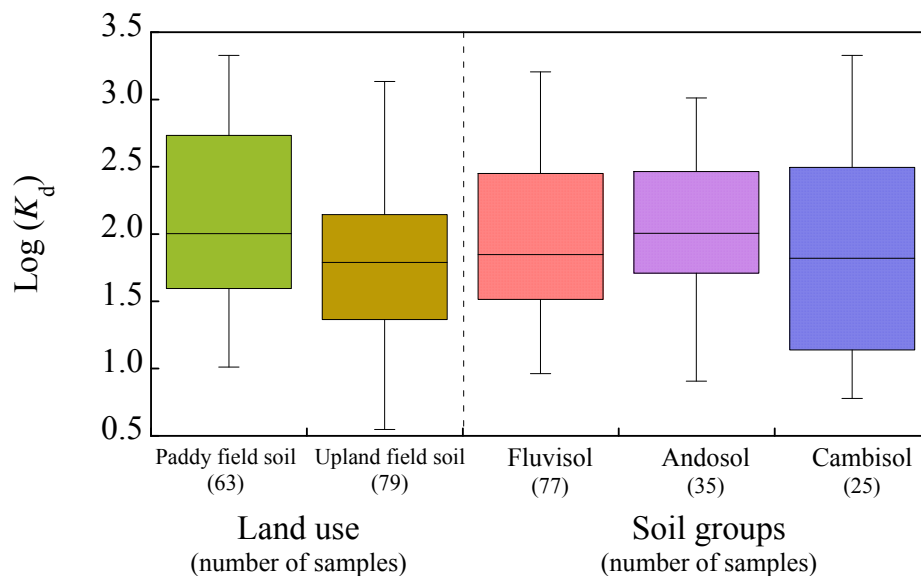
The  $K_d$  value (L/kg) was calculated using the following equation:

$$K_d = (C_i - C_e) * W_l / (C_e * W_s)$$

where  $C_i$  (Bq/L) is the initial radionuclide activity,  $C_e$  (Bq/L) is the radionuclide activity in the liquid phase after shaking for 7 days with the tracer,  $W_l$  is the solution volume (L), and  $W_s$  is the soil dry weight (kg).

During the batch process, selenite, the initially added form, would be changed to selenate by oxidation or to elemental Se by reduction which may affect the  $K_d$ -Se. Thus information on Se chemical form during the  $K_d$  measurement was checked. During the experimental period, pH did not change with time, whereas the solution remained under oxic conditions. Under these pH and oxic conditions, selenite would be stable. Indeed, we previously found that Se in the filtrate was collected by an anion exchange resin [9]. These results implied that the predominant chemical form of Se in the soil solution was selenite.

The  $K_d$ -Se values are listed in Tables II and III for paddy field soil and upland field soil samples, respectively and values are summarized in Fig.4. The cumulative probability distribution of  $K_d$ -Se was on the log-normal line so that GM was calculated; the value was 85 L/kg (range: 3.5-2100 L/kg). The  $K_d$  values for Japanese agricultural soils were close to the  $K_d$  range expected for soils in temperate environments, 150-1800 L/kg [12]. Comparison of the  $K_d$  values between soil uses showed the values differed (t-test,  $p < 0.05$ ); for example, GMs of the  $K_d$ s for paddy field soil and upland field soil samples were 116 and 67, respectively. Previously, we reported that acid oxalate extractable Al and Fe were the major adsorbents of Se in Japanese agricultural soils [9]. These acid oxalate extractable Al and Fe concentrations might have an effect in the present study. However,  $K_d$  values did not differ significantly (t-test,  $p > 0.05$ ) among the soil groups such as Andosol, Cambisol and Fluvisol.



**Fig. 4. Box-and-whisker plot of  $K_d$  values of the land uses and soil groups.**Table II. Soil-Soil Solution Distribution Coefficient,  $K_d$ , and Several Soil Properties in Paddy Field Soil Samples

No.	Soil classification FAO-UNESCO	$K_d$ (L/kg)	PH (H <sub>2</sub> O)	CEC (meq/100g)	EC (1:5) ( $\mu$ S/cm)	Total C (g/kg)	Particle size distribution (%)		
							Sand	Silt	Clay
P-1	Fluvisol	111	5.0	11	129	37	28	32	41
P-2	Andosol	159	6.1	16	96	85	52	31	17
P-3	Andosol	74	6.1	12	112	28	31	22	47
P-4	Fluvisol	298	5.6	14	112	25	48	25	28
P-5	Fluvisol	864	6.0	12	108	20	21	29	50
P-6	Cambisol	84	5.2	13	118	28	43	21	36
P-7	Fluvisol	74	5.6	<D.L.	71	35	58	22	20
P-8	Fluvisol	44	6.0	11	72	21	60	18	22
P-9	Fluvisol	33	6.8	13	98	20	53	22	26
P-10	Fluvisol	125	5.3	<D.L.	253	19	53	20	27
P-11	Fluvisol	55	6.7	13	95	21	36	35	29
P-12	Fluvisol	671	5.6	13	81	35	17	38	45
P-13	Fluvisol	1034	5.0	10	87	26	42	40	18
P-14	Andosol	292	6.7	23	109	108	50	33	17
P-15	Cambisol	590	5.5	14	123	19	22	28	51
P-16	Fluvisol	611	5.7	11	142	16	25	35	40
P-17	Fluvisol	833	5.4	14	92	23	25	36	38
P-18	Fluvisol	287	6.1	16	75	30	46	29	25
P-19	Fluvisol	108	6.7	19	75	16	52	29	20
P-20	Fluvisol	541	5.3	13	92	18	53	20	28
P-21	Fluvisol	208	6.0	<D.L.	57	16	51	29	20
P-22	Fluvisol	506	5.5	15	75	22	35	41	25
P-23	Fluvisol	602	5.3	<D.L.	72	16	56	18	26
P-24	Fluvisol	127	6.0	<D.L.	114	14	61	24	16
P-25	Cambisol	842	4.8	13	89	25	50	25	26
P-26	Fluvisol	578	5.3	13	96	33	47	29	24
P-27	Fluvisol	956	5.7	11	74	47	30	42	29
P-28	Fluvisol	1606	5.5	13	63	23	48	25	28
P-29	Fluvisol	1194	5.5	13	65	43	49	33	19
P-30	Fluvisol	156	5.5	<D.L.	81	16	52	25	23
P-31	Cambisol	1337	5.6	12	64	17	51	18	32
P-32	Fluvisol	570	5.8	13	49	29	59	23	18
P-33	Fluvisol	101	6.7	12	170	30	48	33	20



P-34	Fluvisol	105	6.2	10	61	16	75	12	14
P-35	Fluvisol	110	5.8	10	54	17	64	19	18
P-36	Cambisol	2130	5.3	13	89	23	21	35	44
P-37	Andosol	142	5.9	13	65	25	42	22	36
P-38	Fluvisol	18	5.5	17	65	19	72	14	14
P-39	Fluvisol	15	5.4	16	93	30	45	25	30
P-40	Histosol	15	6.2	15	61	30	73	15	11
P-41	Fluvisol	18	5.1	14	88	19	70	18	11
P-42	Gleysol	15	5.7	13	58	19	69	14	17
P-43	Andosol	36	5.8	14	108	125	50	35	15
P-44	Fluvisol	13	5.6	13	60	15	63	22	16
P-45	Fluvisol	13	6.2	11	132	19	72	14	14
P-46	Fluvisol	14	5.2	12	128	29	54	28	18
P-47	Fluvisol	10	5.8	12	95	14	63	23	15
P-48	Cambisol	14	5.9	11	53	19	69	17	14
P-49	Fluvisol	18	5.2	11	77	23	68	18	14
P-50	Fluvisol	16	6.1	12	97	28	61	25	14
P-51	Fluvisol	60	5.6	11	72	18	45	28	27
P-52	Fluvisol	80	5.2	27	88	37	40	33	27
P-53	Fluvisol	85	4.8	24	91	35	46	26	28
P-54	Cambisol	66	5.6	24	62	25	50	20	30
P-55	Andosol	58	5.9	10	94	22	63	27	10
P-56	Gleysol	137	5.8	26	69	58	49	34	17
P-57	Fluvisol	38	6.9	19	67	19	56	26	19
P-58	Fluvisol	46	6.2	9	82	34	65	20	15
P-59	Fluvisol	65	5.6	12	81	45	65	22	13
P-60	Fluvisol	79	5.2	5	57	17	43	31	26
P-61	Cambisol	72	5.3	13	72	18	49	25	26
P-62	Fluvisol	39	6.3	22	129	15	54	22	24
P-63	Fluvisol	66	6.3	6	109	16	38	35	28

Table III. Soil-Soil Solution Distribution Coefficient,  $K_d$ , and Several Soil Properties in Upland Field Soil Samples

No.	Soil classification FAO-UNESCO	$K_d$ (L/kg)	PH (H <sub>2</sub> O)	CEC (meq/100g)	EC (1:5) ( $\mu$ S/cm)	Total C (g/kg)	Particle size distribution (%)		
							Sand	Silt	Clay
F-1	Andosol	140	5.7	12	175	59	60	24	17
F-2	Andosol	99	6.4	16	93	75	67	22	12
F-3	Fluvisol	52	6.2	12	214	17	34	38	28
F-4	Andosol	113	5.3	13	179	94	60	25	15

WM2009 Conference, March 1-5, 2009, Phoenix, AZ

F-5	Andosol	190	7.0	16	157	25	71	17	11
F-6	Fluvisol	66	5.8	15	67	28	17	33	51
F-7	Andosol	70	5.4	11	70	28	65	15	20
F-8	Cambisol	32	5.7	<D.L.	43	9	57	19	24
F-9	Cambisol	23	6.5	11	53	14	66	14	20
F-10	Andosol	97	5.0	14	251	63	22	49	30
F-11	Fluvisol	22	6.9	14	107	23	57	19	24
F-12	Andosol	1028	5.6	10	113	17	65	21	14
F-13	Fluvisol	283	6.4	14	76	24	15	41	44
F-14	Andosol	582	6.5	16	118	27	33	33	33
F-15	Cambisol	113	6.7	16	194	41	54	22	24
F-16	Fluvisol	560	6.8	13	105	32	27	47	27
F-17	Fluvisol	1364	6.4	17	110	59	53	31	16
F-18	Andosol	358	5.7	14	124	36	37	35	28
F-19	Andosol	368	6.0	17	201	68	53	26	22
F-20	Regosol	4	6.9	<D.L.	34	3	98	<0.1	2
F-21	Fluvisol	141	5.2	15	211	20	47	32	21
F-22	Cambisol	63	6.2	14	444	23	58	21	21
F-23	Cambisol	37	6.1	13	559	21	61	11	28
F-24	Fluvisol	21	6.6	14	141	22	64	14	22
F-25	Fluvisol	62	7.1	16	137	23	44	28	28
F-26	Fluvisol	33	5.6	13	906	31	69	15	16
F-27	Fluvisol	32	7.4	18	192	18	10	39	51
F-28	Fluvisol	140	6.7	18	132	19	23	39	38
F-29	Cambisol	313	5.7	16	82	29	33	32	35
F-30	Andosol	636	5.7	12	73	59	43	38	20
F-31	Andosol	846	6.7	12	152	67	54	30	16
F-32	Andosol	669	6.0	12	637	56	68	20	12
F-33	Fluvisol	35	7.6	11	256	36	50	27	22
F-34	Cambisol	978	7.3	14	464	24	72	16	12
F-35	Fluvisol	70	7.1	12	66	30	70	13	18
F-36	Andosol	304	6.2	13	70	36	50	27	23
F-37	Fluvisol	124	4.3	13	478	22	55	23	22
F-38	Cambisol	19	7.2	12	117	24	69	9	22
F-39	Cambisol	606	5.4	14	55	44	50	23	27
F-40	Fluvisol	35	7.8	12	169	32	57	24	19
F-41	Fluvisol	851	5.3	11	76	11	45	16	39
F-42	Fluvisol	51	7.0	12	126	16	51	21	28
F-43	Fluvisol	318	6.1	21	95	35	68	19	14
F-44	Cambisol	116	5.2	16	289	8	30	30	40
F-45	Andosol	101	5.7	<D.L.	971	76	65	17	18
F-46	Andosol	22	6.0	17	134	28	83	7	10
F-47	Cambisol	8	6.8	20	2635	26	59	23	18

F-48	Andosol	16	7.4	16	95	31	84	7	9
F-49	Fluvisol	17	6.3	14	72	17	49	29	22
F-50	Andosol	8	6.4	14	263	13	66	19	15
F-51	Gleysol	11	6.1	14	189	27	61	19	20
F-52	Andosol	36	6.8	15	92	41	80	8	13
F-53	Andosol	23	6.6	21	300	73	80	9	11
F-54	Andosol	39	6.2	15	181	25	80	8	12
F-55	Fluvisol	10	6.7	11	49	15	73	14	12
F-56	Cambisol	8	6.5	11	193	21	79	8	13
F-57	Cambisol	6	6.2	12	675	31	75	13	12
F-58	Fluvisol	9	6.5	12	81	23	74	13	13
F-59	Cambisol	11	5.4	11	307	16	72	14	13
F-60	Fluvisol	17	5.1	12	107	26	57	20	23
F-61	Cambisol	10	6.3	20	141	69	71	15	13
F-62	Fluvisol	13	8.1	20	487	17	32	43	24
F-63	Cambisol	119	4.6	13	196	31	51	26	22
F-64	Fluvisol	98	4.9	19	91	24	36	36	28
F-65	Fluvisol	42	5.7	10	592	20	48	29	24
F-66	Andosol	55	5.9	21	314	54	39	51	10
F-67	Andosol	51	6.9	9	160	22	63	19	18
F-68	Andosol	48	6.8	30	90	71	52	39	9
F-69	Fluvisol	41	6.8	7	24	6	78	10	12
F-70	Fluvisol	60	6.3	8	107	15	45	29	27
F-71	Andosol	73	5.8	24	149	51	45	30	25
F-72	Fluvisol	68	6.8	13	77	17	32	34	35
F-73	Andosol	56	6.3	17	442	28	32	36	32
F-74	Cambisol	13	6.9	6	146	14	63	16	21
F-75	Fluvisol	91	6.1	13	54	52	32	60	8
F-76	Fluvisol	27	6.5	21	972	50	54	36	11
F-77	Andosol	190	6.5	28	115	100	48	41	11
F-78	Andosol	129	6.2	16	64	43	45	36	19
F-79	Andosol	121	5.7	14	69	47	61	27	13

### Soil-to-plant Transfer Factor (TF)

The TF value was calculated using the following equation:

$$TF = C_p / C_s$$

where  $C_p$  (mg/kg-dry) is the elemental concentration in plant and  $C_s$  (mg/kg-dry) is its concentration in soil. In this study, we calculated TFs of stable Se from the results for crop and soil samples. The results are shown in Tables IV and V for brown rice and upland field crops, respectively. The GM of TFs for brown rice, wheat, leafy vegetables, beans, fruit vegetables, tubers, and root vegetables were 0.066, 0.038, 0.033, 0.030, 0.017, 0.016 and 0.015, respectively. Brown rice showed the highest TFs though Se

concentrations in the soil samples were almost the same for most soils.

Previously reported TF values were 0.64 for soybean [13] and 3.7-79.5 in 5 crops [14]. These values were obtained with soil freshly contaminated with Se radioisotopes, thus the added Se would be more mobile than we found in the fields. It was pointed in ref. [3] that the bioavailability of Se to plants is determined by other factors including pH and redox conditions, speciation, soil texture and mineralogy, organic matter content, and the presence of competing ions. A high Se content soil would show selenium deficiency if the amount of bioavailable Se is low. As clear from Fig. 3, there were no Se concentration differences in paddy field and upland field soil samples, though  $K_d$  values were statistically higher in paddy field soil samples. From these results, probably due to the longer growing period and different water management in the paddy fields for rice compared to those for upland field crops, the TF might be high in rice.

Table IV. Concentrations of Se in Agricultural Soil and Crop Samples, and TF Values on Dry Weight Basis (Brown Rice).

No.	Se in soil (mg/kg)	Se in crop (mg/kg)	TF ( $\times 10^{-3}$ )	No.	Se in soil (mg/kg)	Se in crop (mg/kg)	TF ( $\times 10^{-3}$ )	No.	Se in soil (mg/kg)	Se in crop (mg/kg)	TF ( $\times 10^{-3}$ )
P-1	0.55	<D.L.	-	P-22	0.39	0.016	41	P-43	0.94	0.009	10
P-2	1.16	0.054	46	P-23	0.22	0.040	182	P-44	0.09	0.012	131
P-3	0.42	0.092	218	P-24	0.28	0.024	84	P-45	0.22	0.018	85
P-4	0.25	<D.L.	-	P-25	0.68	0.010	15	P-46	0.23	0.033	141
P-5	0.43	<D.L.	-	P-26	0.83	0.043	51	P-47	0.07	0.020	287
P-6	0.31	<D.L.	-	P-27	0.68	0.019	28	P-48	0.18	0.040	220
P-7	0.52	<D.L.	-	P-28	0.52	0.022	42	P-49	0.26	0.020	78
P-8	0.18	<D.L.	-	P-29	1.21	0.062	51	P-50	0.37	0.031	85
P-9	0.45	<D.L.	-	P-30	0.21	0.017	80	P-51	0.38	0.021	56
P-10	0.18	<D.L.	-	P-31	0.12	0.011	88	P-52	0.57	0.045	80
P-11	0.41	<D.L.	-	P-32	0.51	0.034	66	P-53	0.45	0.022	49
P-12	0.53	0.037	71	P-33	0.37	0.016	42	P-54	0.29	0.017	60
P-13	0.70	0.021	29	P-34	0.19	0.010	53	P-55	0.40	0.028	70
P-14	1.27	0.029	23	P-35	0.24	0.017	72	P-56	1.06	0.038	36
P-15	0.31	0.019	62	P-36	0.45	0.032	70	P-57	0.18	0.009	51
P-16	0.48	0.046	97	P-37	0.40	0.026	67	P-58	0.29	0.010	35
P-17	0.53	0.028	52	P-38	0.54	0.053	99	P-59	0.54	0.026	48
P-18	0.90	0.045	50	P-39	0.47	0.030	64	P-60	0.24	0.030	126
P-19	0.40	0.020	49	P-40	0.44	0.037	84	P-61	0.27	0.017	63
P-20	0.28	0.010	36	P-41	0.40	0.053	130	P-62	0.58	0.058	100
P-21	0.32	0.026	81	P-42	0.54	0.067	124	P-63	0.43	0.053	123

Table V. Concentrations of Se in Agricultural Soil and Crop Samples, and TF Values on Dry Weight Basis (Upland Field Crops)

No.	Se in soil (mg/kg)	Crop name	Se in crop (mg/kg)	TF ( $\times 10^{-3}$ )	No.	Se in soil (mg/kg)	Crop name	Se in crop (mg/kg)	TF ( $\times 10^{-3}$ )
F-1	0.82	Potato	<D.L.	-	F-38	0.23	Nozawana	0.015	66
F-2a	0.98	Leek (green part)	<D.L.	-	F-39	0.57	Japanese radish	0.012	21
F-2b	0.98	Leek (white part)	<D.L.	-	F-40	0.23	Taro	0.004	18
F-3	0.23	Cabbage	<D.L.	-	F-41	0.30	Cabbage	0.011	36
F-4	1.15	Cabbage	<D.L.	-	F-42	0.21	Onion	<D.L.	-
F-5	0.44	Chinese cabbage	0.143	321	F-43	0.34	Wheat	0.017	51
F-6	0.76	Cabbage	<D.L.	-	F-44	0.35	Potato	0.017	48
F-7a	0.45	Leek (green part)	0.054	119	F-45	0.89	Sweet pepper	<D.L.	-
F-7b	0.45	Leek (white part)	<D.L.	-	F-46	0.50	Carrot	0.005	10
F-8	0.22	Chinese cabbage	<D.L.	-	F-47	0.39	Tomato	0.002	6
F-9a	0.30	Carrot (leaves)	0.059	194	F-48	0.67	Japanese radish	0.016	24
F-9b	0.30	Carrot	<D.L.	-	F-49	0.40	Wheat	0.020	49
F-10a	0.68	Japanese radish (leaves)	<D.L.	-	F-50	0.27	Chinese cabbage	0.012	45
F-10b	0.68	Japanese radish	<D.L.	-	F-51	1.31	Japanese radish	0.021	16
F-11	0.25	Spinach	<D.L.	-	F-52	0.37	Chinese cabbage	0.006	15
F-12	0.59	Wheat	0.005	8	F-53	0.94	Cucumber	0.025	26
F-13	0.60	Soybean	0.015	25	F-54	0.35	Spinach	0.014	41
F-14	1.34	Potato	0.035	26	F-55	0.30	Cabbage	0.006	21
F-15	0.72	Cucumber	0.007	10	F-56	0.28	Japanese radish	0.007	23
F-16	0.42	Barley	0.018	42	F-57	0.34	Cabbage	0.004	11
F-17	0.81	Peanut	0.014	17	F-58	0.25	Potato	0.004	18
F-18	0.65	Sweet potato	0.007	11	F-59	0.33	Egg plant	0.007	21
F-19	0.73	Lettuce	0.006	8	F-60	0.34	Wheat	0.006	17
F-20	<D.L.	Sweet potato	<D.L.	-	F-61	0.38	Cucumber	0.010	25
F-21	0.29	Taro	0.006	22	F-62	0.50	Bitter cucumber	0.046	91
F-22	0.23	Tomato	0.008	35	F-63	0.54	Soybean	0.013	25
F-23	0.43	Tomato	0.006	13	F-64	0.39	Soybean	0.021	53
F-24	0.12	Onion	<D.L.	-	F-65	0.33	Cucumber	0.012	36

F-25	0.32	Potato	0.007	21	F-66	0.59	Tomato	0.007	11
F-26	0.27	Lettuce	0.004	17	F-67	0.41	Wheat	0.024	58
F-27	0.29	Onion	0.005	18	F-68	0.88	Japanese radish	0.030	34
F-28	0.31	Barley	0.013	42	F-69	0.22	Egg plant	0.006	27
F-29	0.84	Potato	0.010	12	F-70	0.37	Wheat	0.013	35
F-30	1.38	Japanese radish	0.014	10	F-71	1.18	Tomato	0.010	8
F-31	1.56	Wheat	0.240	154	F-72	0.36	Soybean	0.031	88
F-32	1.30	Sweet pepper	0.007	5	F-73	0.36	Tomato	0.006	17
F-33	0.43	Cabbage	0.005	13	F-74	0.16	Onion	0.002	14
F-34	0.52	Egg plant	0.012	23	F-75	0.67	Soybean	0.010	15
F-35a	0.44	Leek (green part)	0.024	55	F-76	0.55	Egg plant	0.005	9
F-35b	0.44	Leek (white part)	0.008	18	F-77	1.33	Carrot	0.006	5
F-36a	0.73	Leek (green part)	0.033	46	F-78	0.53	Soybean	0.015	28
F-36b	0.73	Leek (white part)	0.022	30	F-79	0.47	Sweet potato	0.001	3
F-37	0.26	Egg plant	<D.L.	-					

## ACKNOWLEDGEMENTS

We thank Mr. T. Takamura and Ms. I. Hirai (Tokyo Nuclear Co., Ltd.) for their technical support. This work was partially supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan.

## REFERENCES

1. JAPAN NUCLEAR CYCLE DEVELOPMENT INSTITUTE, "H12: Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan, Supporting Report 3", JNC TN1400 99-020, Japan Nuclear Cycle Development Institute (1999).
2. THE FEDERATION OF ELECTRIC POWER COMPANIES OF JAPAN and JAPAN NUCLEAR CYCLE DEVELOPMENT INSTITUTE, "Second Progress Report on Research and Development for TRU Waste Disposal in Japan -Design, Safety Assessment and Means of Implementation in the Generic Phase-", The Federation of Electric Power Companies of Japan (2005). [in Japanese] (English version is in press).
3. J. A. PLANT, D. G. KINNIBURGH, P. L. SMEDLEY, F. M. FORDYCE, and B. A. KLINCK, "Arsenic and selenium", In "Treatise on Geochemistry" (H. D. Holland, K. K. Turekian Eds.), Elsevier, Amsterdam. 9: 17-66 (2005).
4. J. GAILLARDET, J. VIERS and B. DUPRE, "Trace elements in river waters", In "Treatise on Geochemistry" (H. D. Holland, K. K. Turekian Eds.), Elsevier, Amsterdam. 5: 225-272 (2004).
5. Y. HASHIMOTO and J. W. WINCHESTER, "Selenium in the atmosphere," Environ. Sci. Technol. 1, 338-340 (1967).
6. B. MARKERT, "Instrumental Element and multi-element analysis of plant samples", John Wiley & Sons, Chichester (1996).

WM2009 Conference, March 1-5, 2009, Phoenix, AZ

7. A. KABATA-PENDIAS and A. B. MUKHERJEE, "Trace elements from soil to human", Springer, Berlin (2007).
8. C. J. SCHOLLENBERGER and R. H. SIMON, "Determination of exchangeable capacity and exchangeable bases in soils", *Soil Sci.* 59, 13-24 (1945).
9. Y. NAKAMARU, K. TAGAMI and S. UCHIDA, "Distribution coefficient of selenium in Japanese agricultural soils", *Chemosphere* 58, 1347-1354 (2005).
10. N. K. ISHIKAWA, S. UCHIDA and K. TAGAMI, "Soil-soil solution distribution coefficient for radionuclides in Japanese agricultural soils", In "Proceedings of 16<sup>th</sup> Pacific Basin Nuclear Conference", Aomori, Paper ID P161123. (2008).
11. WORLD HEALTH ORGANIZATION, "Guidelines for Drinking-water Quality: incorporation first addendum to third edition, Volume 1 Recommendations", WHO, Geneva (2006).
12. INTERNATIONAL ATOMIC ENERGY AGENCY, "Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments", Technical Report Series No.364, IAEA, Vienna (1994).
13. S. AMBE, Y. OHKUBO, Y. KOBAYASHI, M. IWAMOTO, H. MAEDA and M. YANOKURA, "Multitracer study on transport and distribution of metal ions in plants", *J. Radioanal. Nucl. Chem.*, 195, 305-313 (1995).
14. C. MUNIER-LAMY, S. DENEUX-MUSTIN, C. MUSTIN, D. MERLET, J. BERTHELIN and C. LEYVAL, "Selenium bioavailability and uptake as affected by four different plants in a loamy clay soil with particular attention to mycorrhizae inoculated ryegrass", *J. Environ. Radioactiv.* 97, 148-158 (2007).