Transfer Factors of Technetium-99 for Various Plants in Forests

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Introduction

Technetium belongs to the Mn group and it has no stable isotopes. Among its radioactive isotopes, ^{99}Tc is of potential long-term importance in the environment, because ^{99}Tc is a pure beta emitter (E_{max}=0.29 keV) and its half-life is 2.11 x 10^5 y. It is produced in the fissions of ^{235}U and ^{239}Pu at relatively high ratios of ca. 6%, which is similar to the values for ^{137}Cs and ^{90}Sr. Technetium-99 is widely distributed in the environment as a result of fallout from nuclear weapons testing and discharges from nuclear facilities. The quantity of ^{99}Tc produced by atmospheric nuclear weapon explosions was estimated to be 140 TBq using an equal fission yield to that of ^{137}Cs and actual ^{137}Cs deposition data[1)]. The release of ^{99}Tc by nuclear industries through 1986, as estimated by Luykx[2)], was of the order of 1000 TBq. This was mainly from nuclear fuel reprocessing and most of the ^{99}Tc was discharged into the sea and ^{99}Tc concentration in the terrestrial environment is very low[3)]. The amount introduced via nuclear medical use of ^{99m}Tc (half-life: 6.01 h) is negligible. For example, the amounts from ^{99}Mo-^{99m}Tc generators and ^{99m}Tc used during 2004 in Japan were 173 TBq and 325 TBq, respectively[4)], which corresponds to ca. 1.6 MBq of ^{99}Tc generation in total.

Due to very low concentration and analytical difficulties for determination of ^{99}Tc in environmental samples, there is a general lack of data on the levels in the literature. Therefore, the behavior of ^{99}Tc in the terrestrial environment is not well understood. However, it is necessary to obtain transfer parameters under natural conditions for a realistic long-term radiological assessment such as soil-to-plant transfer factors (TFs, concentration in plant/concentration in soil). Thus, we focused on the Chernobyl site where ^{99}Tc concentration levels have already been shown to be higher than those from global fallout Tc[5)]. The concentrations of ^{99}Tc in plant leaves collected in three forests

near the Chernobyl site were determined. The samples were leaves of raspberry, strawberry and pink plants, black alder, birch, cowberry and oak trees, and ferns. Since TFs are usually high in leafy vegetables and grass as reported in Reference 6, leaves of raspberry, strawberry and pink plants are of the most interest in this study for comparison purposes.

Experimental

A Tc-selective chromatographic resin (Eichrom Industries Inc., TEVA resin) was used for Tc purification. Deionized water (>17.6 MΩ) was used throughout the work. Technetium-95m, which was obtained from an Nb foil using the reaction ^{93}Nb (a, 2n) ^{95m}Tc, was applied to determine the recovery of ^{99}Tc in the samples during the chemical separation procedure. The ^{95m}Tc was made at Cycrotron and Radioisotope Center, Tohoku University[6]. A standard ^{99}Tc solution available from Amersham (Solution TCZ.44) was used for calibrating the ICP-MS.

Leaves of 27 plant samples were collected at the Chernobyl site in 1994 and 1995 by the Federal Office for Radiation Protection, Germany, assisted by Moscow State University, Russia, and the Research and Industrial Association 'Pripyat', Ukraine. The sampling was carried out in three forests, D1, D3 and K2, around the Chernobyl Nuclear Power Plant. Forests D1 and D3 were 28.5 km and 26 km to the south of the reactor, respectively, while forest K2 was 6 km to the southeast. Soil samples were also collected at the same sampling sites and the details were reported previously[5].

A simple wet digestion method in combination with an TEVA resin separation method was applied for ^{99}Tc. After incineration of the soil samples at 450˚C, a certain amount of ^{95m}Tc was added and they were mixed uniformly. Then, Tc was extracted with 4M HNO_3 while heating the samples at 100˚C in a glass beaker covered with a watch glass. The residue was removed by filtration and the solution was diluted to obtain the acidity of ca. 0.1M HNO_3 and passed through a TEVA resin column to purify and concentrate Tc isotopes. Technetium adsorbed on the resin was eluted with 5 mL of 8 M HNO_3 solution. The volume of the ^{99}Tc fraction from the TEVA column solution was reduced to near dryness (<70˚C) and then dissolved in 5mL of 2% HNO_3 solution. Radiochemical recoveries of Tc plant samples were monitored with ^{95m}Tc activities. Then the solution was introduced into an ICP-MS (Yokogawa, PMS-2000).

Results and Discussion

Total chemical recoveries throughout the method with ^{95m}Tc ranged from 0.48 to 0.92 with an average of 0.76. No relation was found between sample amount and recovery. Three replicates of samples K2-94F-1 and D3-94G-1 were used in amounts of 1 - 2.6 g and 1.8 - 10.2 g, respectively, and there were no differences between their recoveries. However, the recoveries of two sub-samples of K2-94G-2 and D1-94G-1 differed by about 0.1 and 0.2, respectively; when sample amounts increased, the recoveries decreased. Possibly, plant matrices, such as K, Ca and Cl, would affect the ^{99}Tc recovery, especially at the TEVA resin separation steps.

Technetium-99 concentrations in all samples are listed in Table 1 and they ranged from <0.006 to 6.0 mBq g^{-1}. For 11 samples, especially in the tree group, it was hard to determine ^{99}Tc, because of its low concentration.

TF is defined as the ratio of activity concentration in plant (in Bq g^{-1} dry weight (DW)) to activity concentration in soil (in Bq g^{-1} DW). It has been noted that forest under story plants are expected to take up radionuclides mainly from organic layers and therefore, concentration of ^{99}Tc in organic soil would be important for estimation of its uptake. Thus, TFs were calculated in terms of activity concentration in a plant relative to activity concentration in organic soil. Table 1 also shows the TFs of Tc for all the samples. They ranged from <0.016 to 0.28 for Fern, 0.009 to 0.47 for Herb and <0.006 to 0.05 for Tree groups based on the ^{99}Tc contents of the organic layers. The highest TF was found in the leaves of raspberry. The observed-TFs were lower than the IAEA-compiled values of 8.1 - 2600 for leafy vegetables, fodder and grass[7]; other reported values are as listed in Table 2. Comparing the TF for Tc reported for various plants grown in soil, the present results were almost the same as the data observed in reclaimed land[10], although the data from pot experiments[11] were much higher.

From these results, we concluded Tc bioavailability in the natural environment would be lower than the bioavailabilities obtained from laboratory studies that used TcO_4^- to obtain Tc TFs. It is known that the most plant-available form of Tc is TcO_4^-, and other forms have less availability to plants. Thus, TcO_4^- was not the only Tc chemical from in these forests. Possibly most Tc was in less soluble form, such as organically bound forms, sesquioxide bound and lower oxidation forms[12].

Concerning the Herb group, the TFs were 0.17-0.47 at K2, 0.20-0.32 at D1 and 0.009-0.086 at D3. The TFs for the samples collected at D3 were lower than those at D1 and K2. This might be due to the fact that D3 was a so-called wet forest and reducing

conditions might possibly exist. The bioavailable Tc was presumably influenced by the soil redox conditions: Tc would be transformed from to a lower oxidation and less available form under a relatively low redox condition. Due to the relatively low redox conditions of the D3 site, the TFs were lower than those in K2 and D1 sites.

Thus, the low TFs we observed in this study implied that 8-9 years after ^{99}Tc release from the Chernobyl Nuclear Power Plant, Tc should have been transformed to insoluble or less plant-available forms. Technetium would not be in readily available form to plants in the natural environment.

References

1) Aarkrog A., Dahlgaard H., Hallstadius L., Holm E., Mattson S., Rioseco J., Technetium in the Environment (Desmet G. and Myttenaere C. eds.), Elsevier Appl. Sci. Pub., London (1986) 69.
2) Luykx F., Technetium in the Environment (Desmet G. and Myttenaere C. eds.), Elsevier Appl. Sci. Pub., London (1986) 21.
3) Tagami K., Uchida S., J. Nucl. Radiochem. Sci. **3** (2002) 1.
4) Japan Isotope Association, Statistics on the Use of Radiation in Japan (2005) 38.
5) Uchida S., Tagami K., Wirth E., Rühm W., Chemosphere **39** (1999) 2757.
6) Sekine T., Konishi M., Kudo H., Tagami K., Uchida S., J. Radioanal. Nucl. Chem. **239** (1999) 483.
7) IAEA, Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments, Technical Reports Series No. 364. IAEA, Vienna, (1994) 14.
8) Vandecasteele C.M., Dehut J.P., Van Laer S., Deprins D., Myttenaere C., Health Phys. **57** (1989) 247.
9) Hoffman F.O., Garten C.T., Jr., Lucas D.M., Huckabee J.W., Environ. Sci. Technol. **16** (1982) 214.
10) Green N., Wilkins B.T., Davidson M.F., Hammond D.J., J. Environ. Radioactiv. **27** (1995) 35.
11) Echevarria G., Morel J.L., Florentin L., Leclerc-Cessac, E. J. Environ. Radioact. **70** (2003) 85.
12) Tagami K., Uchida S., Toxicol. Environ. Chem. **56** (1996) 235.

Table 1. Chemical recovery of ^{95m}Tc, concentration of ^{99}Tc and transfer factor in plant samples collected in forests within the 30-km zone around the Chernobyl Nuclear Power Plant.

Plant type	Place		Tc-95m Recovery	Tc-99 mBq g^{-1} dry	Transfer factor
Fern	K2	94Fern-1a	0.847	2.42 ± 0.18	
		94Fern-1b	0.895	2.37 ± 0.29	
		94Fern-1c	0.884	2.31 ± 0.26	0.276 ±0.019
		95Fern-1	0.829	0.603 ± 0.045	0.045 ±0.003
		95Fern-2	0.749	3.79 ± 0.35	0.283 ±0.026
	D3	94Fern-1	0.661	< 0.040	< 0.016
		95Fern-2	0.483	< 0.031	< 0.018
		95Fern-3	0.844	< 0.069	< 0.040
Glass	K2	94Glass-1	0.599	4.04 ± 0.42	0.470 ±0.051
		94Glass-2a	0.747	1.30 ± 0.17	
		94Glass-2b	0.852	1.57 ± 0.16	0.168 ±0.015
		95Glass-1	0.739	6.02 ± 0.52	0.450 ±0.039
		95Glass-2	0.762	3.61 ± 0.33	0.269 ±0.025
	D1	94Glass-1a	0.596	0.254 ± 0.022	
		94Glass-1b	0.824	0.451 ± 0.090	0.318 ±0.042
		94Glass-2	0.563	0.222 ± 0.017	0.200 ±0.015
		95Glass-1	0.901	0.257 ± 0.021	0.231 ±0.019
	D3	94Glass-1	0.868	0.215 ± 0.016	0.092 ±0.006
		94Glass-2	0.706	0.094 ± 0.008	0.039 ±0.003
		94Glass-3	0.553	0.128 ± 0.011	0.053 ±0.004
		95Glass-1	0.915	0.016 ± 0.003	0.009 ±0.002
Tree	D1	94Tree-1	0.898	< 0.006	< 0.006
		95Tree-1	0.883	0.013 ± 0.001	0.012 ±0.001
	D3	94Tree-1	0.878	< 0.035	< 0.014
		94Tree-2	0.652	< 0.040	< 0.016
		94Tree-3	0.850	< 0.056	< 0.023
		94Tree-4	0.633	< 0.028	< 0.011
		95Tree-1	0.685	< 0.028	< 0.016
		95Tree-2	0.543	< 0.023	< 0.013
		95Tree-3	0.856	< 0.028	< 0.016
		95Tree-4	0.843	0.094 ± 0.011	0.055 ±0.007

Table 2. Transfer factors for Tc reported for various plants grown in soil.

Crop	TF	Conditions	Reference
Grass pasture mixture	6	Lysimeter	8
Herbaceous vegetation	6.7-22	Silt loam, Field	9
Grass	0.91-3.02	Reclaimed land, Field	10
Rye grass	44-371	Cambisol, Pot	11
Herbaceous vegetation	0.009-0.47	Podzol, Peat-gley, Field	This study