Relationship between Radiocesium and Stable Cesium in Plants and Mushrooms Collected from Forest Ecosystems with Different Contamination Levels

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INTRODUCTION

Many studies on forest ecosystems were carried out mainly in the European forests after the Chernobyl accident (1-5). Japanese forest sites have also extensively been investigated, although the radionuclide deposition originating from the accident was low (6-11). Radiocesium contamination of forest products is still high in contrast to agricultural products, even more than 10 years after the Chernobyl accident. High concentrations of radiocesium were observed especially in mushrooms. Since the removal of radiocesium from a contaminated forest is not feasible on a large scale, studies on the distribution and transfer of radiocesium in forest ecosystems are important from radiation protection viewpoint.

The experimental investigations in forest ecosystems have provided information for the development of models which explain radiocesium transport through several compartments in a forest ecosystem. These models were reviewed by Schell et al. (12, 13). However, discussions during recent workshop on forest radioecology (14) and preliminary results of the IAEA BIOMASS Forest Working Group model validation studies show that the long-term fate of radiocesium in forest ecosystems is still difficult to predict, mainly because the vertical profile of radiocesium in soil is still changing with time. Other reasons may be the time-dependent availability of radiocesium in soil, notably during the first few years after deposition, and the variability of ecological characteristics for different forest sites. As the chemical behavior of radiocesium is expected to be almost identical to that of stable Cs, analyses of stable Cs and related stable elements should be useful to understand the long-term behavior of radiocesium and its equilibrium distribution.

Myttenaere et al. (15) summarized the relationship between radiocesium and K in forests, and suggested the possible use of K behavior for the prediction of radiocesium behavior. There are many studies on the behavior of major nutrient elements such as K, Mg and Ca in forests (16, 17) because these elements are directly related to the forest growth. Effect of acid deposition on the behavior of these elements was also studied in the last two decades (18-20). However, the data for trace alkali and alkaline earth elements including stable Cs in forest ecosystems are limited.

In this study, the concentrations of stable Cs and related alkali and alkaline earth elements in mushrooms, plants and soils were determined by inductively coupled plasma-mass spectrometry (ICP-MS) or inductively coupled plasma-atomic emission spectrometry (ICP-AES). Samples were collected in forests with different contamination levels in Japan, Germany, Finland, Italy, Ireland and Belarus. Data of stable elements were compiled together with those of radiocesium.

SAMPLING AND MEASUREMENTS

Sampling of mushrooms, plants and soils

Twenty-nine mushroom samples belonging to 17 species were collected from a Japanese pine forest on sandy soil near the coast in Tokai-mura, Ibaraki from 1989 to 1991. In this forest, most of the radiocesium originates from atmospheric nuclear weapons testing. Eight plant samples were collected in the same forest in November 1990. After removing attached soil and humus, samples were freeze-dried and pulverized. Soils from different depths were also sampled in the forest. Twenty-two mushroom samples belonging to 3 species were collected from forests in Alavus, Kirkkonummi and Kullaa in Finland from 1989 to 1993. A detailed study on the transfer of radiocesium and stable Cs from soil to fungal fruit bodies and green plants was performed at a Norway spruce stand in Hochstadt, Germany. This site has been under investigation since 1987, and the soil layers from which certain species of mushrooms take up radiocesium were estimated by using the $^{137}Cs/^{134}Cs$ ratios (21). Totally 25 soil profiles were randomly taken within an area of about 100 x 100 m^2 of the investigated site in 1993, 1995 and 1996. Mushrooms were collected every year from 1993 to 1996. For the present study, samples of *Clitocybe nebularis*, *Xerocomus badius*, *Hydnum repandum*, and *Russula cyanoxantha* were chosen since it was shown that these species take up radiocesium from different soil horizons. For comparison, samples of a berry plant (*Vaccinium myrtillus*) were also included in this study. All samples were air-dried (70 °C) and milled. Further details of sampling methods in this site are given elsewhere (21, 22).

Tree samples were collected in a mixed forest in Italy and commercial stand in Ireland. One representative tree was selected for each forest and different parts of the tree were sampled. 90 years old Norway Spruce (*Picea abies*) was sampled in September 1996 in Italy, and 43 years old Sitka Spruce (*Picea sitchensis*) was sampled in 1998 in Ireland. The samples were dried in an oven at 105 °C and milled.

Determination of 137Cs

Each dried sample was placed in a plastic bottle and concentrations of ^{137}Cs and ^{134}Cs were determined by counting with a Ge-detector. The decay correction was made as to May 1986. Details for the radiocesium determination have been described by Muramatsu et al. (6) and Rühm et al. (21, 22).

Determination of stable elements

Mushroom and plant (0.2 - 0.4 g) and soil (0.1 g) samples were digested in Teflon™ PFA pressure decomposition vessels with acids (HNO_3, HF and $HClO_4$). A microwave digester (CEM, MDS-2000) was used for heating the samples. After digestion, the samples were evaporated to dryness. Then, the residues were dissolved in 1 - 2% HNO_3 to yield the sample solutions. Trace elements (Rb, Cs, Sr and Ba) were measured by ICP-MS (Yokogawa Analytical Systems, PMS-2000). Major elements (Na, K, Mg and Ca) were determined by ICP-AES (Seiko Instruments, SPS7700). Duplicate sample preparation and measurement have been done for each sample. Standard reference materials, such as Tomato Leaves (1573a) issued by the National Institute of Standards & Technology and JB-1a (basalt) issued by the Geological Survey of Japan, were used to validate the analytical procedure. Details for the analyses have been described by Yoshida and Muramatsu (23).

RESULTS AND DISCUSSION

Difference between mushrooms and plants

The analytical results of alkali and alkaline earth elements and ^{137}Cs in 29 mushroom and 8 plant samples collected at a pine forest in Tokai-mura, Japan, are compiled by Yoshida and Muramatsu (24). The highest median concentration in mushrooms was found for K (27200 mg/kg-dry) followed by Mg (1050 mg/kg-dry), Na (1000 mg/kg-dry), Ca (389 mg/kg-dry), Rb (87.6 mg/kg-dry), Ba (5.12 mg/kg-dry), Sr (2.88 mg/kg-dry) and Cs (1.01 mg/kg-dry). For plants, the highest median value was found for Ca (16300 mg/kg-dry) followed by K (8860 mg/kg-dry), Mg (1800 mg/kg-dry), Na (802 mg/kg-dry), Sr (70.8 mg/kg-dry), Ba (11.1 mg/kg-dry), Rb (9.35 mg/kg-dry) and Cs (0.043 mg/kg-dry). The median concentrations of ^{137}Cs were 135 Bq/kg-dry for mushrooms and 3.8 Bq/kg-dry for plants. In comparison with the elemental composition of plants, the mushroom composition could be characterized by the high ^{137}Cs, Cs and Rb concentrations and low Ca and Sr concentrations. Higher accumulations of Cs and Rb, and lower accumulation of Sr in mushrooms than those in plants were also observed from cultivation experiments in flasks using radiotracers (25, 26).

Relationship between 137Cs and stable Cs

Correlations between ^{137}Cs and stable Cs for mushrooms collected from 6 different forests in Finland, Germany and Japan are summarized in **Figure 1**. The data for mushrooms collected from a pine forest in Rokkasho-mura, Aomori, Japan reported by Tsukada et al. (11) are also plotted in the figure. A good correlation between ^{137}Cs and stable Cs was observed for each site independently, although several different species of mushrooms are included. This finding suggests that mushrooms take up ^{137}Cs together with stable Cs. The $^{137}Cs/Cs$ ratios were fairly constant for samples collected at the same site. The results for different sites, however, showed different degrees of variability. The highest ratio was obtained at Kullaa ($^{137}Cs/Cs$ = 5100 ± 1900 Bq/mg), followed by Alavus (4000 ± 800 Bq/mg), Hochstadt (990 ± 470 Bq/mg), Kirkkonummi (800 ± 260 Bq/mg), Rokkasho-mura (430 ± 120 Bq/mg) and Tokai-mura (150 ± 40 Bq/mg). The difference between the ratios of different sites is partly attributable to the different deposition levels of ^{137}Cs. The estimated total deposition of ^{137}Cs from Chernobyl accident is the highest in two Finnish forests, Kullaa (18.2 kBq/m^2) and Alavus (25.7 kBq/m^2) and the lowest in Japanese forests (Rokkasho-mura and Tokai-mura). A positive correlation between the total deposition of ^{137}Cs and the $^{137}Cs/Cs$ ratio was observed for the different sites. However, different concentrations of stable Cs in soil and different forest types (e.g. vegetation, geology, soil type) also affect the $^{137}Cs/Cs$ ratio.

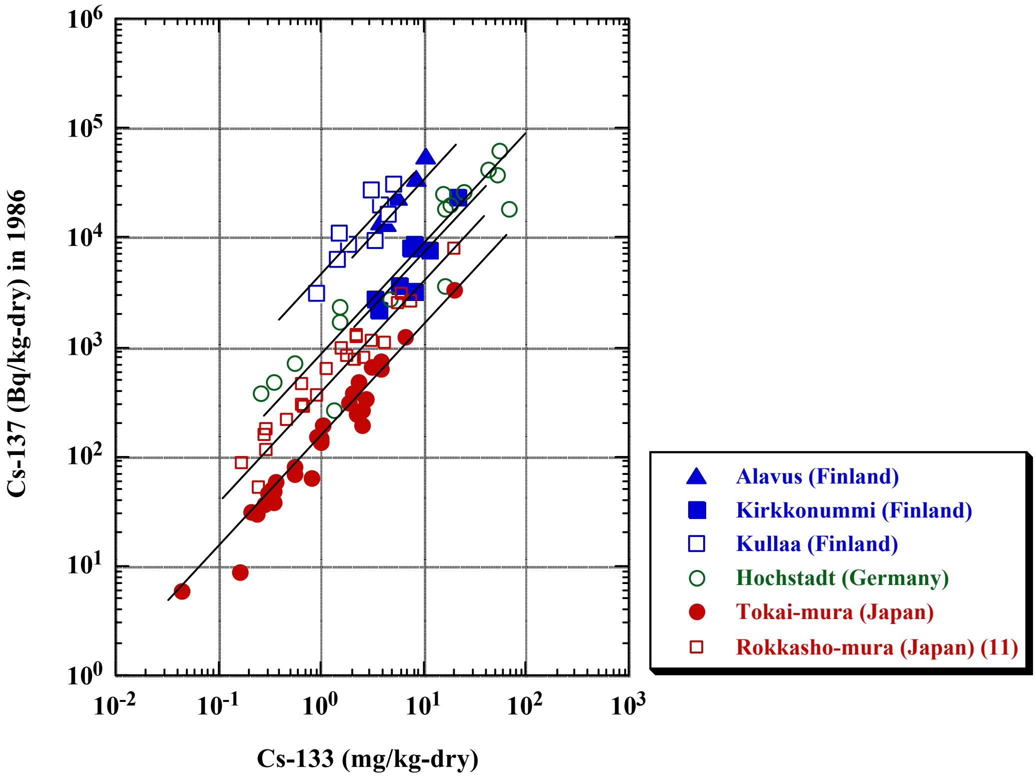

Figure 1. Relationship between stable Cs and ^{137}Cs in mushrooms collected from 6 different forests in Finland, Germany and Japan. Data for Rokkasho-mura were reported in Tsukada et al. (11).

The ^{137}Cs/Cs ratio might be a useful criterion for judging the equilibrium of deposited ^{137}Cs to stable Cs in a forest ecosystem. Standard deviation of the ^{137}Cs/Cs ratio was low in Japanese forests (Rokkasho-mura: 28%, Tokai-mura: 27%), in which most ^{137}Cs originated from the global fallout. This finding suggests that ^{137}Cs, mainly deposited onto the forest ecosystem during the 1960s, has already attained a dynamic equilibrium within the soil-mushroom system and is now cycling together with stable Cs. The standard deviation of the ^{137}Cs/Cs ratio was high in Hochstadt, Germany (48%). A detailed study on the transfer of radiocesium from organic soil horizons to mushrooms in Hochstadt showed that the ^{134}Cs/Cs ratio in mushrooms reflected the ratios of those soil layers, from which the corresponding mushroom species takes up ^{134}Cs (27).

A good correlation between ^{137}Cs and stable Cs was also observed in plant samples collected from different parts of trees in Irish and Italian forests as shown in **Figure 2**. An outlier observed for each site is a datum for dead branch or dead wood in which physiological property of Cs might be different from other living parts. The ^{137}Cs/Cs ratios were almost constant for samples collected in the same site, suggesting that the distribution of ^{137}Cs in trees is similar to that of stable Cs more than 10 years after the Chernobyl accident. The higher ^{137}Cs/Cs ratio in Italy compared to Ireland is, at least partly, attributable to the higher deposition of Chernobyl ^{137}Cs at the Italian forest. The estimated total depositions of ^{137}Cs from Chernobyl accident are 37 kBq/m^2 in Italian forest and 8.3 kBq/m^2 in Irish forest.

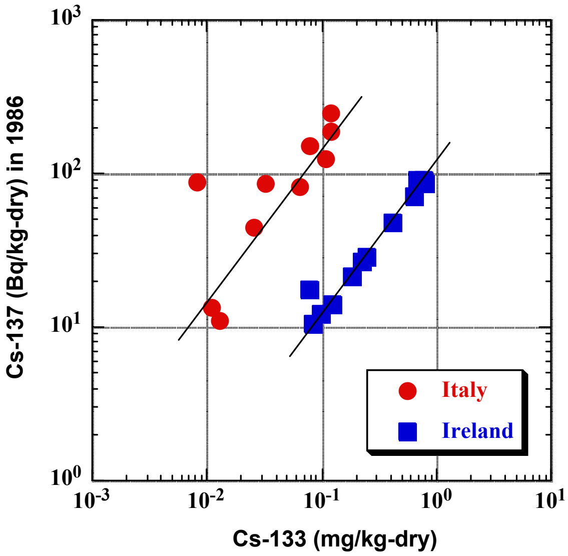

Figure 2. Relationship between stable Cs and ^{137}Cs in plant samples collected from different parts of trees in Irish and Italian forests.

Transfer factors

For mushrooms collected in a Japanese pine forest in Tokai-mura, the tentative transfer factors defined as "median concentration in mushrooms on a dry weight basis" divided by "concentration in the surface 0 - 5 cm soil on a dry weight basis" were calculated both for ^{137}Cs and stable Cs. The values were 3.9 for ^{137}Cs and 0.8 for stable Cs (24). The transfer factor of ^{137}Cs was higher than that of stable Cs. The surface 0 - 5 cm soil in this forest is a mixture of organic materials and minerals (sand). Stable Cs is originally contained in the mineral components and this stable Cs is difficult for plants and mushrooms to take up. Tsukada et al. (11) also reported higher average transfer factor for ^{137}Cs than stable Cs in 21 different species of mushrooms collected in a pine forest in Rokkasho-mura.

On favorable conditions, e.g. in case of organic horizons with a low content of mineral material, the physico-chemical properties of stable Cs and radiocesium are expected to be similar. For the forest at Hochstadt, Germany, transfer factors for ^{134}Cs and stable Cs were calculated for four different mushroom species and one species of green plants (*Vaccinium myrtillus*). Transfer factors were explicitly related to the soil layers, from which the corresponding species takes up radioactive ^{134}Cs and ^{137}Cs (Table 1) (27). Most species take up ^{134}Cs from organic layers (L, Of and/or Oh). The resulting transfer factors for stable Cs were close to the corresponding transfer factors for ^{134}Cs, indicating that bio-availabilities of Chernobyl ^{134}Cs and stable Cs are similar. With this result, it is possible to predict the future contamination of radiocesium in understory vegetation of forest ecosystems; changes will occur due to physical decay of radiocesium, and due to migration of radiocesium to or from the rooting zone of the corresponding species. There will be no significant future change (aging effect) of the bio-availability of radiocesium in the organic layers at the investigated site.

Analyses for stable elements have provided much information on the behavior of elements, which are related to radionuclides in forest ecosystems. The ^{137}Cs/Cs ratio might be useful for judging the equilibrium of deposited ^{137}Cs in different parts of a forest ecosystem. The stable Cs analyses might be also useful to predict the long-term ^{137}Cs contamination of mushrooms and plants. We are currently studying samples collected in 4 different forests with different contamination levels in Belarus. This study is expected to yield comprehensive information of radiocesium and stable cesium and their interrelation in the whole forest ecosystem.

Table 1. Transfer factors for mushrooms and berry plant for stable Cs and Chernobyl ^{134}Cs, both calculated with respect to the layers from which radiocesium is taken up (at Hochstadt, Germany) (27).

Species	Horizon	Cs-134*	Stable Cs
Clitocybe nebularis	L&Of	0.5 ± 0.1	1.0 ± 0.4
Xerocomus badius	L&Of&Oh	21.8 ± 2.3	13.1 ± 2.8
Hydnum repandum	Oh	43.6 ± 7.4	24.2 ± 1.9
Russula cyanoxantha	Oh&Ah	Oh: 2.9 ± 1.9 Ah: 15.3 ± 10.2	Oh: 6.2 ± 3.8 Ah: 10.5 ± 6.5
Vaccinium myrtillus (leaves)	L&Of	2.5 ± 0.5	3.9 ± 1.5

*The decay correction for ^{134}Cs was made as to May 1986.

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