Radioactive contamination of several materials following the Fukushima Daiichi Nuclear Power Station accident

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A R T I C L E   I N F O

A B S T R A C T

Following the 2011 accident at the Fukushima Daiichi Nuclear Power Station (NPS), which is owned by Tokyo Electric Power Co., radioactive nuclides were released into the environment and heavily contaminated the NPS site. In this study, the contamination behavior of radioactive nuclides in accumulated water, rubble, soil, and vegetation is discussed. Activity concentrations are converted to a "transport ratio," which is the activity concentration ratio normalized using the activity in the source term for the nuclides of interest and a selected standard. The transport ratio of Sr to rubble and soil by way of air was approximately 10^{-2} to 10^{-3}, and the successive transport to accumulated water was comparable with Cs (except for the initial release). Transport of Pu, Am, and Cm was lower than for Sr regardless of the transport process (air or water), whereas those of I, Se, and Te were comparable or greater. Contamination with ^{3}H and ^{14}C was independent of ^{137}Cs, ^{90}Sr, and TRU nuclides and was likely a result of different transport process.

1. Introduction

The Great East Japan Earthquake occurred on March 11, 2011. At Fukushima Daiichi Nuclear Power Station (NPS), owned by Tokyo Electric Power Co. (TEPCO), operation of the reactors was successfully halted; however, the resulting tsunami caused fatal damage to the reactors [1]. The damage to the reactor fuels of units #1 through #3 owing to loss of coolant and the subsequent hydrogen explosions in units #1, #3, and #4 owing to the reaction of fuel cladding with steam caused a release of radionuclides contained within the fuels to the environment. The contamination spread across the NPS site (including buildings) and the surrounding areas.

In the course of decommissioning the power plants of the NPS, radioactive waste will be generated. The mass of waste will be much greater than for a case of general reactor decommissioning, and the potential for contamination by nuclear fuel constituents should be carefully considered during waste management. To establish methodologies for waste processing and disposal, obtaining data on the composition and amount of radionuclides in the contaminated materials is essential; therefore, the analysis of various wastes is important and has been previously conducted [2–4].

The analytical data relating to certain contaminated materials such as rubble, vegetation, soil, and water, have been reported to a team responsible for "countermeasures for decommissioning and contaminated water treatment" by Japan Atomic Energy Agency (JAEA) and the International Research Institute for Nuclear Decommissioning (IRID). Some important nuclides for waste disposal safety include "difficult-to-measure" nuclides, which are often not detected owing to their low concentrations. To estimate the activity concentration of these undetected nuclides, it is necessary to know or presume their contamination behavior in various materials. For this purpose, the "transport ratio," which is the activity concentration ratio normalized using the activity in the source term for the nuclides of interest and of a selected standard, is defined, and the contamination behavior of radionuclides discussed.

2. Method

2.1. Analytical data

The concentrations of radionuclides in contaminated materials were taken from publicly available reports.

Below reactor buildings, turbine buildings, and the centralized radioactive waste treatment facility, contaminated water has accumulated. Analytical data for this water sampled after the accident were taken from the study by Nishihara et al. [2]. Post August 2011, analytical data for this water were taken from the meeting report of the secretariat of the team for "countermeasures for decommissioning and contaminated water treatment" including the former organizations [5–8].

TEPCO sampled the soil in 2011 at fixed points across the NPS site. Sampling sites were located WNW, W, and SSW of the stack of units #1 and #2; these three sites are referred to as ground (1), the Forest of Birds (2), and area surrounding a controlled landfill site (3), respectively. Analytical data given in the press releases from TEPCO were used [9].

Data for rubble (mainly concrete) sampled around and inside the reactor buildings and for the paint on the floors and walls inside the reactor buildings were also reported as mentioned above and are used here [10–12].

Concerning vegetation, felled trees from the temporal repository and living trees sampled on the NPS site were analyzed. Data were reported at the abovementioned meeting and are used here [10,13]. Samples of leaf-branch, fallen leaf, and topsoil (often with humus) were collected from selected living trees.

2.2. Transport ratio

The ratio of the activity concentration for a nuclide of interest to that of a standard nuclide was divided by the ratio of activity for the nuclide in the source term, as shown in the following equation. For convenience, this was referred to as the "transport ratio" (T).

$$T_X = (N_{X,sample}/N_{X,source})/(N_{std,sample}/N_{std,source})$$
$$= (c_{X,sample}/c_{std,sample})/(A_{X,source}/A_{std,source}),$$

where N is number of atoms, c is activity concentration, A is activity, and the subscripts X and std are the nuclide of interest and the standard nuclide, respectively. ^{137}Cs was selected as the standard nuclide. Activities were corrected for decay half-life to March 14, 2011.

For the source term, the calculated activity of the reactor fuels of units #1 through #3 was used [14]. T does not provide information on the transport process but merely describes the relative difference between the initial (source term) and resulting (sample) activities.

3. Results and discussion

3.1. Accumulated water

The water accumulated under the reactor buildings of units #1 through #4 was transferred for chemical decontamination treatment, via the turbine buildings, to the Centralized Radioactive Waste Treatment Facility, which comprises the Process Main Building (PM/B) and the High Temperature Incinerator Building (HTI/B). The accumulated water was treated with zeolite to facilitate cesium adsorption and then passed through a reverse osmosis membrane to recover freshwater that contained tritium as the major radionuclide [15]. The freshwater was used as coolant for the damaged fuels; thus, a recycling system for the water was established. The radioactive nuclides that were initially released from the reactor fuels when the accident occurred were already processed and removed into secondary waste and concentrated waste liquid. It is considered that the radioactive nuclides presently observed in the accumulated water come from successive dissolution of the damaged fuel [16].

The radionuclides ^{3}H, ^{60}Co, ^{63}Ni, ^{79}Se, ^{90}Sr, ^{129}I, ^{238}Pu, and $^{239+240}$Pu were detected in the accumulated water sampled at the
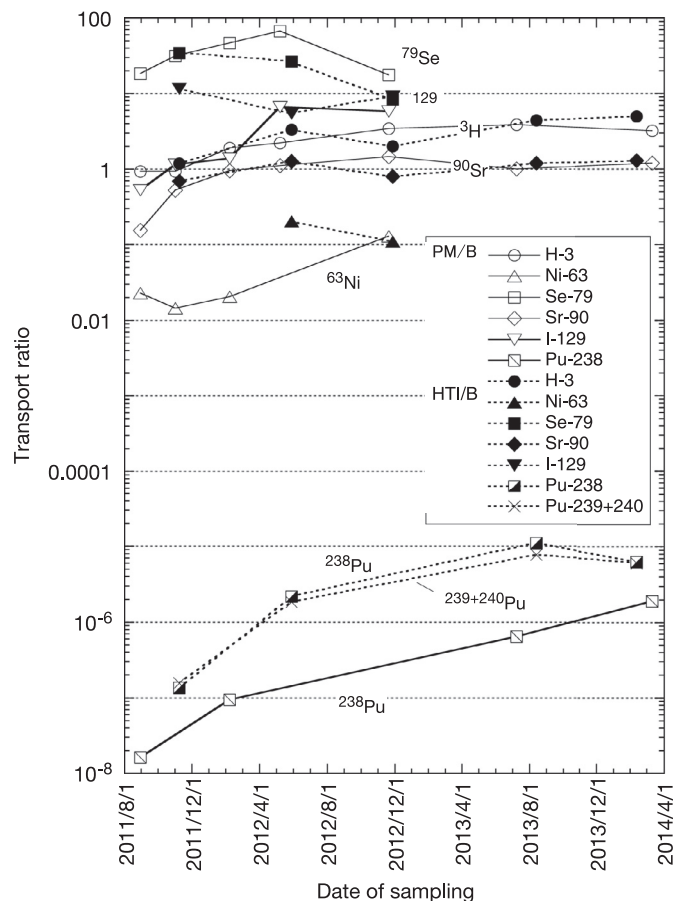

Fig. 1. Transport ratios of selected nuclides to the accumulated water under the Process Main Building (PM/B) and the High Temperature Incinerator Building (HTI/B). The date indicates when the water was sampled.

PM/B and HTI/B. Values of T were calculated with the reported concentrations and their change versus the date of sampling is plotted in Fig. 1. The descending order of T values was Se, I, H, Cs∼Sr, Ni, Pu. T_{Se} decreased from the middle of 2012, whereas T values for ^{3}H and the Pu isotopes seemed to gradually increase. The decrease in T should be affected by differences in the decreases of the nuclides of interest and the standard in the source term, especially when successive release of the nuclides of interest such as Se into the accumulated water is greater than that of standard. T_{Co} was scattered around 0.01–0.1, and a source term other than the damaged fuel is considered likely owing to the less soluble nature of this nuclide.

T_{Sr} shows low values just after the accident, as shown in Fig. 2. T_{Sr} was 10^{-4} for unit #1 and approximately 0.1 for units #2 and #3 (slightly greater for unit #3 than for unit #2), and it gradually increased over time. Treatment of the accumulated water and recycling water was commenced in June 2011, and T_{Sr} increased to around unity in early 2012. During the early stages of the accident (just after the fuel damage), transfer of Sr to the water was suppressed when compared with Cs. It is suggested that when the fuels became damaged, the transfer process for Cs was different than that for Sr, although these gradually became similar.

The concentration of ^{140}Ba was only obtained just after the accident. T_{Sr} was greater than T_{Ba} by several times for units #2 and #3; in contrast, T_{Sr} was somewhat less than T_{Ba} for unit #1.

3.2. Rubble

Concrete and insulator rubble sampled from the first floor of the reactor buildings of unit #1 and #3, and floor paint sampled
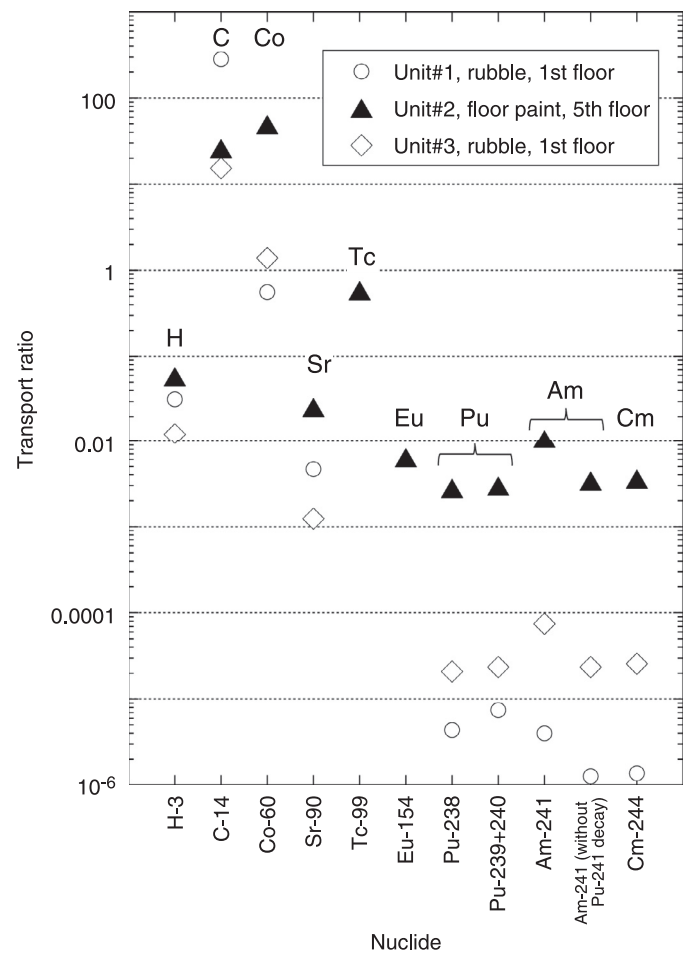
Table 1

Transport ratios calculated for several nuclides to the samples of rubble and paint obtained around and inside the reactor buildings.

Sampling location		Sample	Number of sample	^{3}H		^{14}C		^{60}Co		^{90}Sr	
Unit#1	surroundings	rubble	5	0.042	(5)[1]	<290	(1)[3]	3.5	(2)	0.0046	(5)
	1st floor	rubble	5	0.036	(5)	260	(5)	<0.92	(4)[3]	0.0046	(5)
	1st floor	paint decontaminated	2	<0.4	(0)[2]	<2×10^3	(0)[2]	<300	(0)[2]	0.0030	(1)
Unit#2	5th floor	paint	1	0.066	(1)	23	(1)	74	(1)	0.026	(1)
	1st floor	paint decontaminated	1	0.050	(1)	<100	(0)[2]	<20	(0)[2]	0.014	(1)
Unit#3	surroundings	rubble	5	0.011	(5)	<33	(4)[3]	2.3	(4)	0.00012	(5)
	1st floor	rubble	4	0.014	(4)	<17	(2)[3]	2.3	(3)	0.0012	(4)
Unit#4	surroundings	rubble	5	27	(5)	<2.2×10^5	(4)[3]	<550	(1)[3]	<0.0013	(2)[3]
	fuel pool	gravel, pebble	2					5.2 × 10^7	(2)		

[1] Number of detection, when the number is smaller than the number of samples, the nuclide was not detected for the rest of samples.

[2] No concentration data owing to the detection limits. The T value was calculated using the value of detection limit.

[3] Sample(s) of 'not detected' had lower T value(s) than the samples detected.
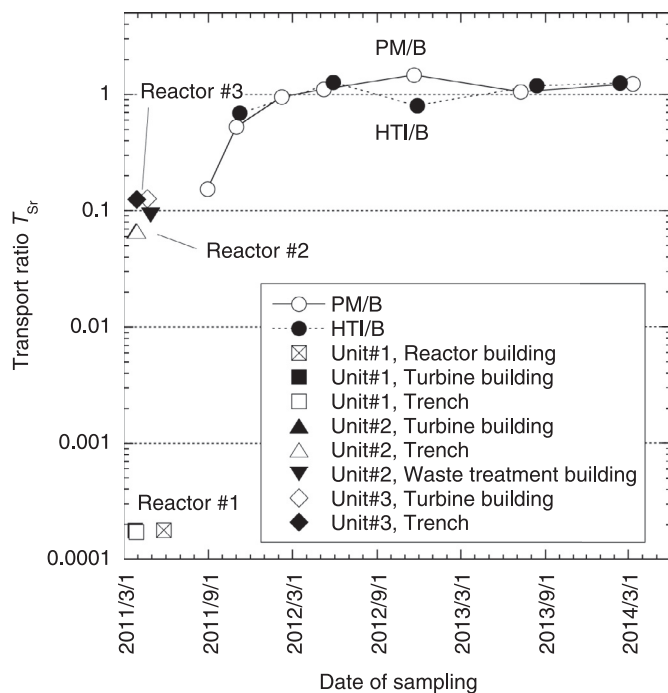
Fig. 2. The transport ratio of ^{90}Sr to the accumulated water under the buildings, including those of units #1 through #3, just after the occurrence of the accident.

Fig. 3. Transport ratio of selected nuclides to the samples of rubble on the first floor of the reactor buildings of units #1 and #3 and floor paint of the 5th floor (operating floor) of the reactor building of unit #2.

from the 5th floor (obtained a few years after the occurrence of the accident), were analyzed. The transport ratios calculated from the reported data are shown in Fig. 3. In the case of plural samples obtained, for the rubble from units #1 and #3, the arithmetic mean was plotted. The decreasing order of T values is C~Co, Cs, Tc, H, Sr, Ln (Eu)~An (Pu, Am, Cm). The values for ^{154}Eu, ^{238}Pu, $^{239+240}$Pu, ^{241}Am, and ^{244}Cm were similar for each unit; therefore, it is suggested that the contamination behavior of TRU and lanthanides was similar. The T value of ^{241}Am was corrected for decay of ^{241}Pu and was observed to be close to that of ^{244}Cm. Consequently, it was suggested that T value was dependent on the unit, with values for unit #2 greater than those for units #1 and #3 (except for ^{14}C).

The reactor buildings of units #1, #3, and #4 experienced hydrogen explosions due to fuel damage; therefore, rubble was scattered around the buildings. The T values of ^{3}H, ^{14}C, ^{60}Co, and ^{90}Sr to the rubble are compared in Table 1. Irrespective of the type of material (rubble or paint) and location of the sampling (inside or outside) the T value of each nuclide and unit was often similar. This suggests that the nuclide composition was similar in each unit; radioactive nuclides presumably contained within particles

were dispersed across the reactor building, maintaining their composition.

The ^{90}Sr concentration of paint on the first floor of units #1 and #2 after decontamination with strippable coating was obtained. It was found that the T values were similar for each unit. Regarding unit #2, the T values of TRU for the paint of the first and fifth floors was different because TRU was not detected on the first floor and the dispersion process near the reactor would be important for non-volatile TRU elements.

Concerning unit #4, T values of ^{3}H, ^{14}C, and ^{60}Co were greater than those for units #1 through #3. The T value of ^{90}Sr was similar
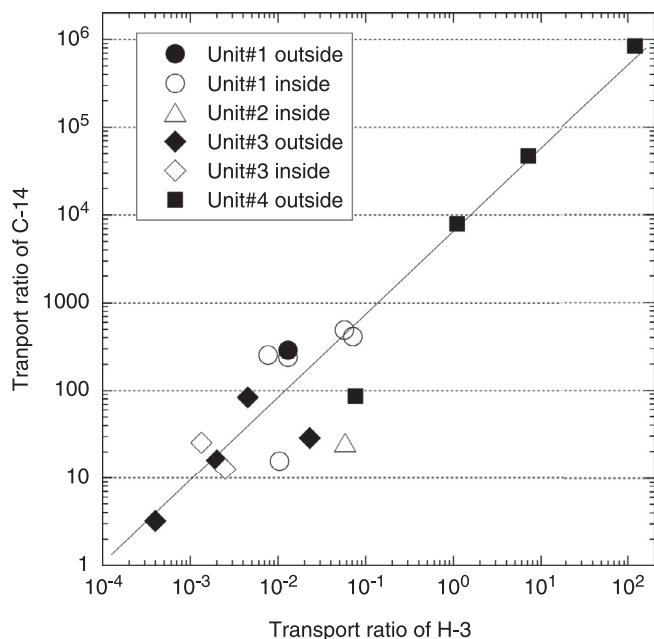
Fig. 4. Correlation between ^{3}H and ^{14}C transport ratios to the samples obtained inside and outside reactor buildings.

to the other units; therefore, the dispersion process of the nuclides was different to that for ^{137}Cs–^{90}Sr. Since ^{60}Co was detected in the gravel and pebble sampled from the spent fuel pool, the high T_{Co} was a result of adsorption of activation products contained in the cooling water. This implies that a source term other than the damaged fuel should be assumed to investigate contamination behavior.

Concerning ^{3}H and ^{14}C contamination, the correlation between T values is shown in Fig. 4. The wide range of values, especially for unit #4, shows that transport of ^{3}H and ^{14}C was dependent on ^{137}Cs, and a different process of dispersion can be suggested. When the accident occurred, the reactor of unit #4 had no fuel and the contamination was owing to influx from unit #3. It is suggested that contamination of unit #4 was a result of a separate influx of ^{3}H–^{14}C and ^{137}Cs–^{90}Sr, and also from a source term related to ^{60}Co within unit #4.

3.3. Soil

When isotopic effects are neglected, the T value should be identical for isotopes of the same element and similar for elements that are chemically similar. The T values of Sr and TRU to the soil at sampling point 1 are plotted versus the date of sampling in Fig. 5 [17]. T values of the strontium isotopes (^{89}Sr and ^{90}Sr) and the TRU nuclides (^{238}Pu, $^{239+240}$Pu, ^{241}Am, and $^{243+244}$Cm) were approximately 0.001 and 2×10^{-5}, respectively. Strontium shows a slightly decreasing tendency; suggesting that it have diffused into the soil while cesium was retained in the surface layer.

The T values for various nuclides to the soil are shown in Fig. 6 [17]. The values for some elements such as I, Sr, and TRU varied across the sampling points, suggesting a dependency on direction from the reactor buildings of unit #1 through #4, and their magnitude followed the order I > Te~Cs > Ag > Sb > Mo > Ru > Ba > Sr > Nb > Pu~Am~Cm. Non-volatile Sr and TRU showed low T values. T_{Sr} values for sampling points 1 through 3 are similar to those for the rubble sampled at reactor buildings #1 and #3 (shown in Fig. 3) and are smaller than those for the accumulated water at steady state. Concerning the relation between the alkaline earth elements Sr and Ba, T_{Ba} was comparable or greater than T_{Sr}.
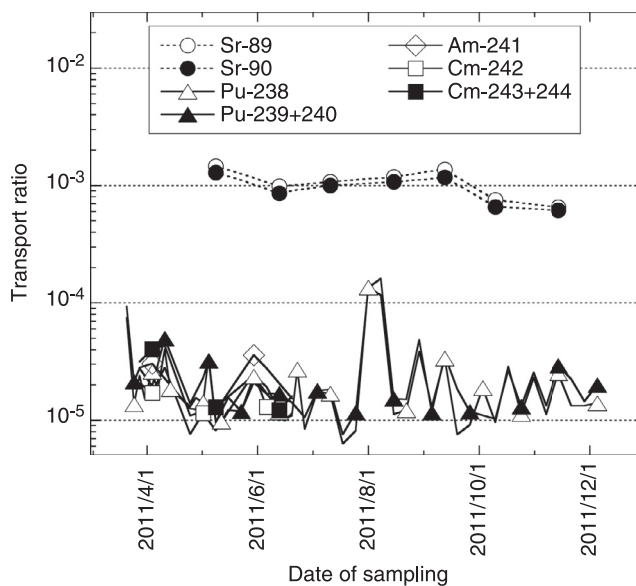

Fig. 5. Time dependence of the transport ratios of Sr and TRU nuclides to the soil sampled at the site of the NPS. The sampling point was in a WNW direction from the stack of units #1 and #2.
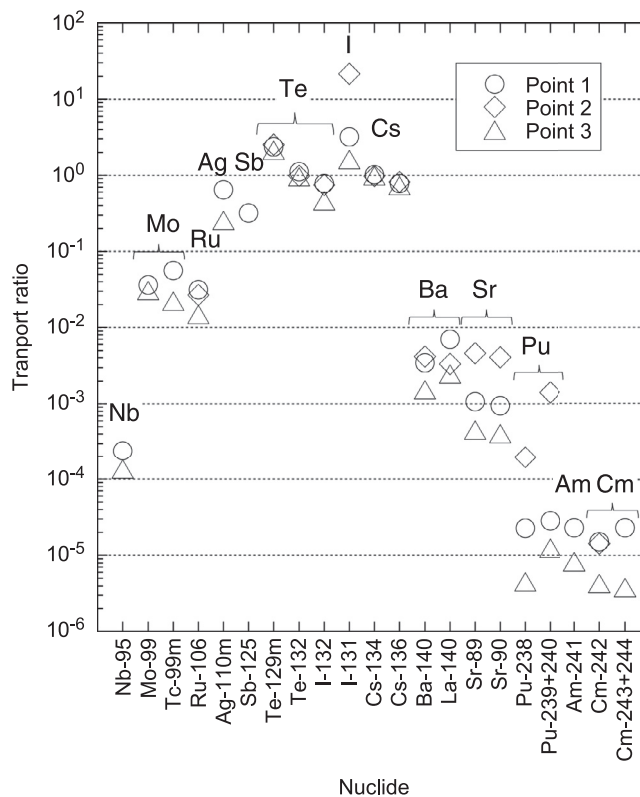

Fig. 6. Transport ratios for selected nuclides to the soil sampled at the three points in the NPS site in 2011. Points 1 through 3 were in the WNW, W, and SSW directions, respectively.

This contrasts with the case of accumulated water in units #2 and #3.

Dependency on distance from the source term should be considered owing to the similarity in T_{Sr} to the soil and rubble sampled inside and outside the reactor buildings. Sr concentration in the soil surrounding the Fukushima Daiichi NPS was reported [18] and concentration values were taken from [19,20] to calculate T values, as shown in Fig. 7 (where the data was plotted ir-
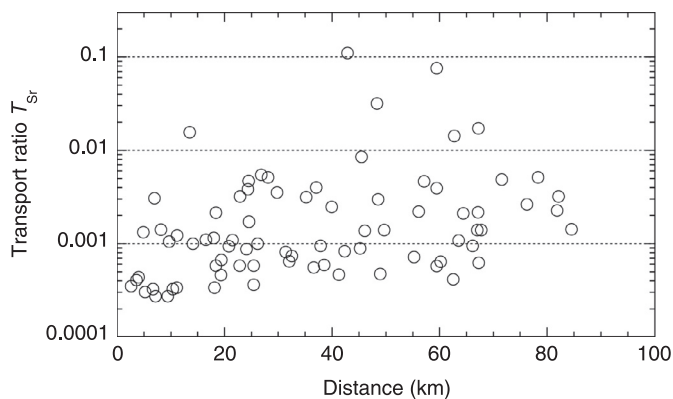
Fig. 7. Distance dependence of ^{90}Sr transport ratio to the soil sampled in the area surrounding Fukushima Daiichi NPS. The data were plotted regardless of the direction from the NPS.
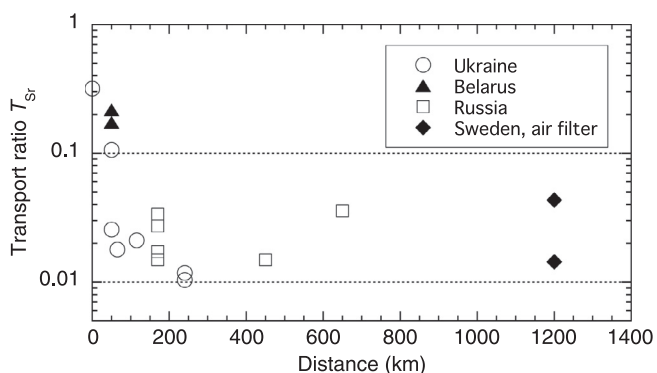

Fig. 8. Distance dependence of ^{90}Sr transport ratio to the soil and air filters obtained in Ukraine, Belarus, Russia, and Sweden after the Chernobyl nuclear power plant accident [21–23].

respective of direction from the NPS because dependency on direction was not apparent). The values were mostly in the range 10^{-2} to 10^{-4}, although a few values exceeded 0.01, and the geometric mean was 0.0015. Because Sr concentration decreased with distance from the NPS and detection of Sr became difficult, T values could not be calculated at distances of 70 km and above.

In the Chernobyl nuclear power plant accident, the contamination spread wider than at Fukushima. The T values of Sr to the soil of Ukraine and other selected countries are plotted (irrespective of direction) in Fig. 8 [21–23]. T_{Sr} decreased to approximate 50 km from the plant and shows a constant value of 0.020 (geometric mean) even beyond 1000 km. This suggests that if radionuclides released from damaged fuel generate particles with a long suspension time in air, they will be transported long distances without a change in radionuclide composition. At Chernobyl, T_{Sr} was one order of magnitude greater than at Fukushima owing to the difference of the source term and the fuel damage process.

Plutonium concentrations in the area surrounding Fukushima Daiichi NPS are so low that the composition of Pu isotopes is affected by the fallout from weapons testing and the Chernobyl accident. Since the contribution of the fallout is significant for $^{239+240}$Pu, ^{238}Pu is important for investigating Pu behavior. The T value of ^{238}Pu was obtained within a distance of approximately 50 km as approximately 10^{-5} [20]. At a distance of 40 km, the concentration ratio of ^{238}Pu to $^{239+240}$Pu was low and the effect of the fallout was recognizable (Table 2).
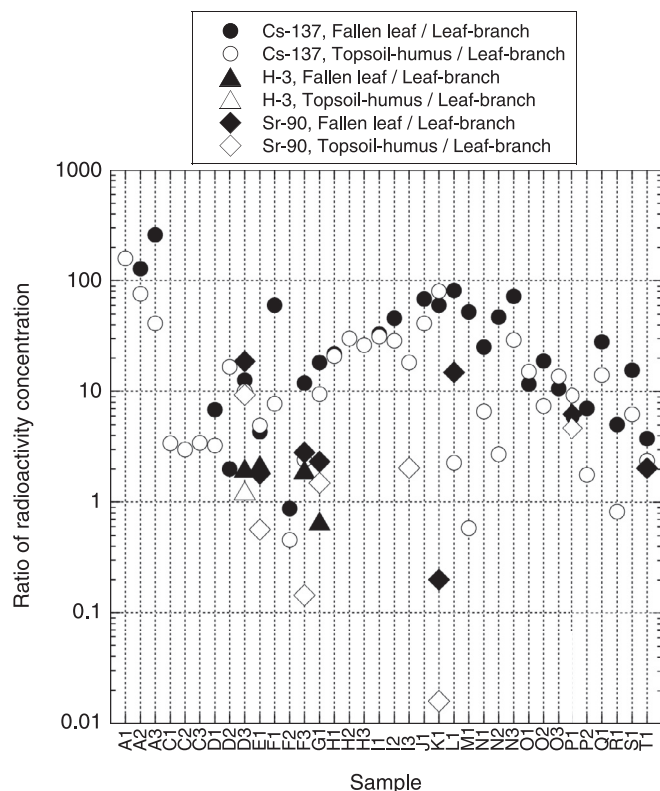

Fig. 9. Concentration ratios of ^{3}H, ^{90}Sr, and ^{137}Cs for vegetation, including fallen leaf and topsoil–humus to those of leaf–branch. Samples were taken across the site and designated as combination of symbols for an arbitrary sectioned area A to T and serial number.

3.4. Vegetation

Living trees sampled two years after the accident were analyzed and the results were reported [13]. The behavior of radionuclides in vegetation should be investigated on consideration with transport of radionuclides within an individual vegetative body. Regarding the concentrations of ^{3}H, ^{90}Sr, and ^{137}Cs, the concentration ratio of fallen leaf and topsoil–humus to leaf–branch is shown in Fig. 9. Many samples exhibited large [^{137}Cs] ratios for fallen leaves and topsoil–humus. The ratios were scattered, suggesting different distributions for individual samples. The concentration ratios for ^{90}Sr were generally lower than those of ^{137}Cs. The ratio of sample K1 was particularly small. As the [^{90}Sr] of the leaf–branch was significantly high, it is likely that ^{90}Sr had been taken up prior to sampling. The ratio for ^{3}H was near unity, indicating that its distribution in the living material was rather uniform. The extent of transport from the leaf–branch to fallen leaf and topsoil followed the order ^{137}Cs > ^{90}Sr > ^{3}H, and it is considered that the transport processes inside the living body were different for the different nuclides.

The T values of ^{3}H and ^{14}C correlate as shown in Fig. 10, and they are independent of Cs transport (similar to the rubble data shown in Fig. 4). As the nuclides were detected in samples obtained 2 years after the accident, it is evident that they were taken up and became fixed within the living material.

4. Conclusions

For contaminated materials generated at the site of the Fukushima Daiichi NPS accident, the behavior of radioactive nuclides was investigated using the "transport ratio." Radionuclides are continuously transported into the accumulated water under

Table 2
The concentrations of plutonium nuclides contained in the soil sample in the neighborhood of the Fukushima Daiichi NPS ad their transport ratio. Concentrations were referred from [20].

| Latitude	Longitude	Distance (km)	Concentration (Bq/m^2)		Concentration ratio	Transport ratio	
			^{238}Pu	$^{239+240}$Pu	^{238}Pu/$^{239+240}$Pu	^{238}Pu	$^{239+240}$Pu
37.4613	141.0099	4.9	0.57	<0.48	>1.2	4.7×10^{-6}	<9.8×10^{-6}
37.4261	140.9739	5.2	0.55	0.66	0.83	3.9×10^{-6}	1.2×10^{-5}
37.5605	140.8238	24	2.3	1.8	1.3	1.9×10^{-5}	3.8×10^{-5}
37.5961	140.7541	31	4.0	1.8	2.2	2.4×10^{-5}	2.7×10^{-5}
37.6640	140.7210	39	0.77	0.60	1.3	4.1×10^{-5}	8.0×10^{-5}
37.7225	140.6881	45	0.82	2.5	0.33	5.2×10^{-5}	3.9×10^{-4}
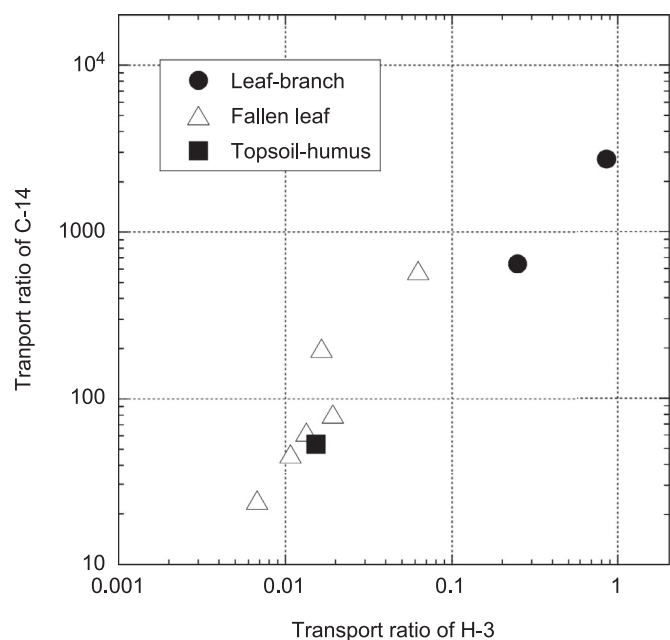
Fig. 10. Correlation between ^{3}H and ^{14}C transport ratios to the samples of vegetation (leaf–branch, fallen leaf, and topsoil–humus).

the reactor buildings. In this process, Sr transport is equivalent to that of Cs, whereas TRU transport is significantly low. In contrast, just after the fuel damage, Sr transport was low and the process might have been different from that of Cs. Transport to the rubble (generated by the hydrogen explosions) and the paint inside the buildings was low for Sr and TRU when compared with Cs. The nuclide composition of contaminants was different among the reactor units. The transport of ^{3}H and ^{14}C did not correlate with ^{137}Cs, suggesting that their transport processes might be different. Transport to the soil at the NPS site was dependent on the direction from the reactor buildings; each damaged reactor exhibited a different release process, contributing to a complex distribution of contamination. Transport of Sr was not dependent on distance from the reactor building. Pu from the accident was detected in the surrounding area. Concerning vegetation, the initial distribution for an individual vegetative body often changed with time. Cs and Sr were transported from leaf–branch to fallen leaf and then to topsoil–humus. In contrast, transport of ^{3}H and ^{14}C was relatively low, and the transport process was not dependent on ^{137}Cs (similar to the case for rubble). Se, Te, and I are volatile and water-soluble, meaning that they were dispersed widely, and their state of stabilization was dependent on the material. The transport ratio defined in this paper is useful for predicting the behavior of elements for which there is no analytical data via the use of data for nuclides of relevant elements.

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