Geographical distribution of radioactive nuclides released from the Fukushima Daiichi Nuclear Power Station accident in eastern Japan

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The geographical distribution of radioactive nuclides released from the Fukushima Daiichi Nuclear Power Station accident in metropolitan areas located in eastern Japan was investigated. The radioactive contamination of environmental samples, including soil and biological materials, was analyzed. The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the soil samples collected from Fukushima City were 122000, 11500 and 14000 Bq/kg on 19th March 2011 and 129000, 11000 and 13700 Bq/kg on 26th March 2011, for the three nuclides respectively. The concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in the soil samples collected from March-June 2011 from study sites ranged from 240 to 101000, 28 to 26200, and 14 to 33700 Bq/kg, respectively. In Higashiosaka City, it began to detect those radioactive nuclides in the atmospheric airborne dust from 25th March. Radioactive fission products ^{95}Zr-^{95}Nb were detected on 18th April 2011. Biological samples collected from Tokyo Bay were studied. The maximum concentrations of ^{134}Cs and ^{137}Cs detected in the biological samples were 12.2 and 19.2 Bq/kg, which were measured in goby. ^{131}I was not detected in the biological samples however, trace amounts of the short half-life nuclide ^{110m}Ag were found in the shellfish samples.

Key Words: *Fukushima Daiichi Nuclear Power Station Accident, Radioactive Nuclide, Radioactive Pollution, Soil, Airborne Dust*

1. Introduction

The radioactive pollution in the metropolitan areas in eastern Japan was received by the Fukushima Daiichi Nuclear Power Station (FDNPS) accident. In order to investigate contamination due to radioactive nuclides released from FDNPS, measurements have been carried out on various environmental samples collected from the surrounding areas.[1, 2)] The detailed measurements from metropolitan areas in eastern Japan have, however, not yet been performed. The geographical distribution of the radioactive pollution in eastern Japan has been investigated by monitoring the dose of radioactive nuclides in the air.[3)] Measurements of the radioactivity of soil and biological samples are scare. Quantifying the radioactivity of the soil samples is important when evaluating environmental contamination from airborne radioactive nuclides released from FDNPS, since airborne nuclides deposit soon after. In addition, measurements on biological samples are necessary to assess exposure of the

food chain to radioactive pollution.

In this study, soil and biological samples taken from metropolitan areas in eastern Japan are analyzed. The geographical distribution of the radioactive pollution from the FDNPS accident is studied. The behavior of radioactive nuclides in the environment is discussed in relation to the resulting findings.

2. Experimental Procedures

The radioactivity of soil and biological samples was determined by gamma-ray spectrometry using a HPGe detector (ORTEC LO-AX 30P) connected with a 4096 ch MCA. The gamma-ray spectrum was measured for energies ranging from 20 to 800 keV. The efficiency of the detector and the geometrical efficiency (for a given sample weight) were corrected using the NIST Environmental Radioactivity Standards SRM 4350B (River Sediment), SRM 4354 (Freshwater Lake Sediment), and 4357 (a blend of ocean sediments collected off the coast of the Sellafield and Chesapeake Bay). The detection limits of ^{134}Cs and ^{137} Cs were 0.5 Bq/kg.[4)]

3. Results and Discussion

(1) Airborne dust from Higashiosaka City

The measurements of radioactivity of airborne dust in Higashiosaka City commenced on 15th March 2011. Higashiosaka City is located 600 km west of FDNPS. The data is shown in Figure 1. The volatile radioactive nuclide ^{131}I was observed first in measurements carried out on 25th March 2011, while ^{134}Cs and ^{137}Cs were seen from 30th March 2011. The concentrations of ^{131}I, ^{134}Cs and ^{137}Cs initially increased rapidly, however, a sudden decrease is observed after the rainfall event on 7th April 2011.[5)] This suggested that radioactive nuclides in the atmosphere were washed out by the rain and, most likely, deposited in the soil. The refractory radioactive fission products ^{95}Zr-^{95}Nb were seen in measurements taken on 18th April 2011. The presence of fission products suggests that the nuclear fuel was involved in the meltdown at FDNPS. The gamma-ray spectrum is shown in Figure 2 (Red: Fission Product, Black: Natural Nuclide). The concentrations of radioactive nuclides in the airborne dust in Higashiosaka City returned to its background level after 25th April 2011.

(2) Soil samples from Fukushima City

The two separate soil samples were collected from the same site in Fukushima City on both 19th March 2011 and 26th March 2011. The gamma-ray spectrum of the sample collected on 19th March 2011 is shown in Figure 3. The radioactive fission products ^{129}Te, ^{129m}Te, ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, and ^{140}Ba were seen in the spectrum. The concentrations and inventories of ^{131}I, ^{134}Cs, and ^{137}Cs found in the soil samples collected on 19th and 26th March 2011 were almost the same (Table 1). It can therefore be concluded that no additional pollution from FDNPS during this period.

(3) Soil samples from the metropolitan area

Radioactive contamination of the soil in the metropolitan area, due to the deposition of an atmospheric radioactive polluted plume, was occurred after rainfall events on 22nd April 2011 and 23rd April 2011.[5)] The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the soil samples collected from the metropolitan area and the Kanto area are given in Table 1. This data presented herein represent the first measurement of the radioactive contamination from the FDNPS accident in these areas.[6)]

The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the soil collected from the outer garden at the Imperial Palace on 10th April 2011 were 18700, 800, and 960 Bq/kg, respectively. The corresponding inventories were 243000, 10400, and 12500 Bq/m^2. Radioactive cesium was seen in 1 cm depth layer in the soil. It has been suggested that radioactive cesium is strongly absorbed by soil due to ion exchange with clay mineral.[7)] The highest measured concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the Kanto area in the soil collected from Kameido on 27th April 2011 were 30400, 2860, and 3590 Bq/kg. The corresponding inventories of the nuclides were 395000, 37100, and 46700 Bq/m^2, respectively. In Kashiwa City, located 20 km northeast of the Imperial Palace, the concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the gutter sludge soil collected on 11th June 2011 were 101000, 26200 and 33700 Bq/kg, respectively. The corresponding inventories of the nuclides were 1310000, 341000, and 438000 Bq/m^2. The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were seen to be higher in soil samples taken from Kashiwa City than in soil samples taken from the other metropolitan areas (Table 1). The high level of contamination here might result from radioactive nuclides moving within the environment of Kashiwa City.

(4) Biological samples from Tokyo Bay

The concentrations of radioactive nuclides in the biological samples collected from Tokyo Bay are given in Table 2. The mullet (*Mugil chephalus*) samples were collected on 20th August 2011. The concentrations of ^{134}Cs and ^{137}Cs measured in the edible and the viscera parts of the fish respectively were 1.1 and 1.5 Bq/kg, and 10.9 and 14.6 Bq/kg. The goby (*Acanthogobius flavimanus*) samples were collected on 21st October, 7th December and 18th December 2011. The concentration of ^{134}Cs ranges from 3.7 to 12.2 Bq/kg and of ^{137}Cs ranges from 5.3 to 19.2 Bq/kg. The short half-life nuclide ^{110m}Ag was seen in the quahog (*Mercenaria mercenaria*) samples collected on 30th October 2011 at the concentration of 0.4 Bq/kg. This nuclide was also seen in the viscera of geoduck clam (*Panopea Japonica*) samples collected on 1st October 2011, at the concentration of 2.1 Bq/kg. Trace amounts of ^{110m}Ag were observed in collected shellfish samples, which were thought to concentrate the transition elements in their viscera organs.[8)]

(5) Environmental samples from the Gunma prefecture

The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the samples collected from the Gunma prefecture are given in Table 3. The radioactive cesium was concentrated to mushroom, fallen leaf and bark in order to well known. The leaf litters were sampled on 29th April 2011 and 14th August 2012. The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs measured in the leaf litter collected on 29th April 2011 were 3210, 3900, and 4740 Bq/kg, respectively. The data measured in 14th August 2012 also showed the similar results. The activity of ^{131}I in the leaf litter collected on 14th August 2012 was extinguished by the disintegration (Table 3). The concentrations of these radioactive nuclides in the soil sample taken from underneath the leaf litter were very low, because the radioactive nuclides were still remained on the leaves and not transferring to the soil.

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Table 1. Concentrations and inventories of ^{131}I, ^{134}Cs, and ^{137}Cs in the soil samples collected from the various locations in eastern Japan.

Site	Depth cm	Sampling Date	Concentration, Bq/kg			Inventory, Bq/m^2		
			^{131}I*	^{134}Cs	^{137}Cs	^{131}I*	^{134}Cs	^{137}Cs
Fukushima City A	0 - 1	March 19, 2011	122000 (72800)**	11500	14000	1590000 (946000)**	150000	182000
Fukushima City B	0 - 1	March 26, 2011	129000 (32200)**	11000	13700	1680000 (419000)**	144000	178000
Tokyo A (Tsukiji)	0 - 1	April 10, 2011	10300	480	580	134000	6250	7530
Tokyo B (Kameido)	0 - 1	April 27, 2011	30400	2860	3590	395000	37100	46700
Tokyo C (outer garden at the Imperial Palace)	0 - 1	April 10, 2011	18700	800	960	243000	10400	12500
Tokyo C (outer garden at the Imperial Palace)	0 - 1	April 28, 2011	11500	940	1170	171000	12900	16800
	1 - 3		810	28	42			
	3 - 6		nd	nd	14			
Chiba City A	0 - 1	April 11, 2011	12200	2030	2510	159000	26400	32600
Chiba City B	0 - 1	April 12, 2011	1900	150	190	24800	1880	2440
Kashiwa City	0 - 1	June 11, 2011	101000	26200	33700	1310000	341000	438000
Gunma Pref. Naganohara Town	0 - 1	March 26, 2011	240	130	160	3160	1740	2120
Asaka City	0 - 1	April 10, 2011	6190	200	250	80500	2550	3280
Chiba Pref. Katori Country	0 - 1	April 20, 2011	6720	410	500	87400	5260	6490
Chiba Pref. Tateyama City	0 - 1	April 20, 2011	1400	50	70	18200	640	900
Nagano Pref. Karuizawa (Alt.1000m)	0 - 1	April 28, 2011	3410	583	753	44400	7580	9790

* These values were decay corrected. The concentration shown is the corrected value for 16th March 2011.

** These values were detected concentration on the corresponding date.

Table 2. Concentrations of ^{134}Cs, ^{137}Cs, and ^{110m}Ag in the biological samples collected from Tokyo Bay.

Species	Position	Sampling Date	Concentration, Bq/kg		
			^{134}Cs	^{137}Cs	^{110m}Ag
Mullet (*Mugil chephalus*)	edible portion	August 20, 2011	1.1	1.5	nd
	viscera		10.9	14.6	nd
Goby (*Acanthogobius flavimanus*)	whole body	October 21, 2011	5.5	6.8	nd
		December 7, 2011	3.7	5.3	nd
		December 18, 2011	12.2	19.2	nd
Quahog (*Mercenaria mercenaria*)	whole body	October 30, 2011	0.4	1.0	0.4
Geoduck Clam (*Panopea japonica*)	edible portion	October 1, 2011	0.4	0.7	nd
	viscera		1.6	2.3	2.1

Table 3. Concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs in the environmental samples collected from the Gunma prefecture.

Samples	Sampling Date	Concentration, Bq/kg		
		^{131}I	^{134}Cs	^{137}Cs
Polypore (*Polyporaceae*)	April 29, 2011	nd	2610	3190
Bark (Cherry Tree) outside	Augst 14, 2011	nd	2510	3300
inside		nd	nd	nd
Leaf litter	April 29, 2011	3210*	3900	4740
Soil underneath leaf litter		nd	3	27
Leaf litter	August 14, 2012	nd	2420	4470
Soil underneath leaf litter		nd	39	86

* The concentration is the corrected value for 16th March 2011. nd: not detected.

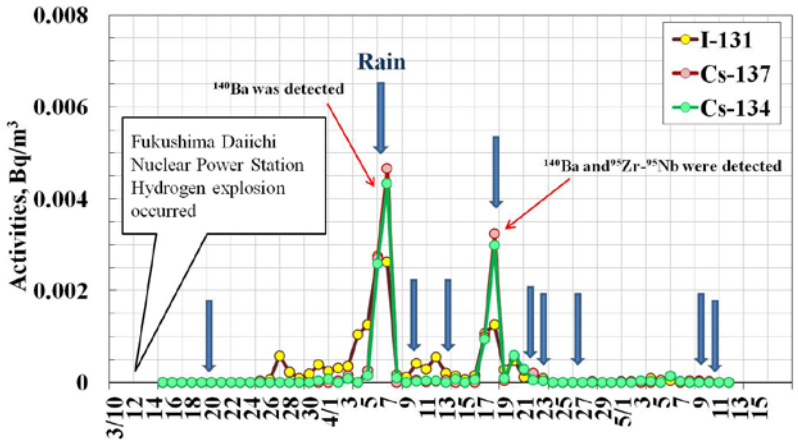

Figure 1. Time variation of the concentration of radioactive nuclides in the airborne dust sample collected in Higashiosaka City.

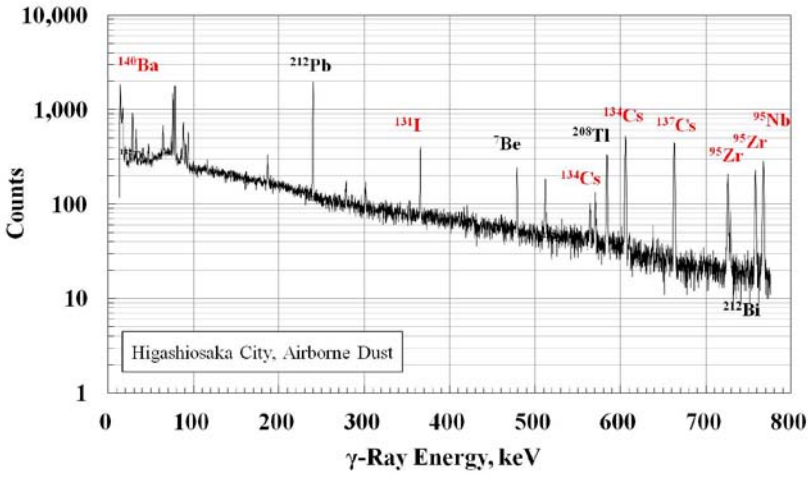

Figure 2. Gamma-ray spectrum of the airborne dust collected in Higashiosaka City on 18th April 2011.

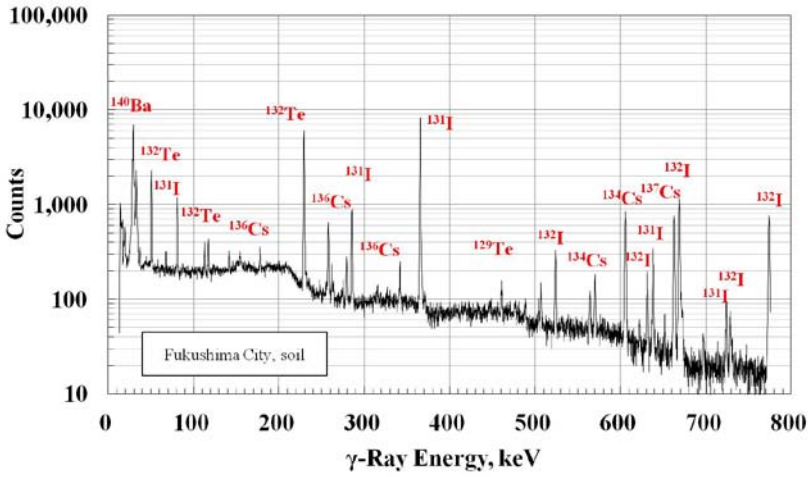

Figure 3. Gamma-ray spectrum of the soil sample collected in Fukushima City on 19th March 2011.