

Sr-90 Deposition Observed in Central and Northeast Honshu Island, Japan, after the Fukushima Dai-ichi Nuclear Power Plant Accident

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Abstract

Monthly depositions of anthropogenic radionuclides, which directly reflect atmospheric processes including emission from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), are an important probe to re-construct FDNPP accident sequence. Japanese radioactivity survey network monitored monthly ^{90}Sr deposition after the FDNPP accident. The highest monthly ^{90}Sr deposition ($470 \pm 17 \text{ Bq m}^{-2}$) was observed in March 2011 at Futaba near the FDNPP site, which is the same order of magnitude as that due to global fallout observed in 1963. The higher ^{90}Sr deposition in March 2011 occurred in the Kanto and inland of the Tohoku areas. The monthly ^{90}Sr deposition decreased with time, as did ^{137}Cs . Atmospheric effect of the FDNPP-derived ^{90}Sr continued more than 1 year. $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in March 2011, which were in the range from 0.99×10^{-4} to 3×10^{-4} , showed no large spatial variation. This result allowed us to estimate total atmospheric emission of ^{90}Sr from the FDNPP accident from the total atmospheric release of ^{137}Cs . The total release amount of ^{90}Sr at an early stage of accident was calculated to be 2.7 – 3.6TBq.

Introduction

On 11 March 2011, the Richter scale 9.0 magnitude earthquake, so called “the 2011 Great East Japan Earthquake”, and Tsunami struck the northeast coast of Japan, resulting in widespread injury and loss of life. These natural disasters caused severe accident in the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) [1]. As a result, large amounts of radionuclides have been released in the environment [1,2]. Japanese government, research institute and university have continuously monitored the FDNPP-derived radionuclides in the atmospheric samples [3-8]. Just after the FDNPP accident, ^{131}I , ^{134}Cs and ^{137}Cs were detected as dominant FDNPP-derived radionuclides in atmospheric samples such as atmospheric dust and deposition, whereas refractory radionuclides such as ^{103}Ru and ^{95}Zr could not be detected [3-5]. In contrast to volatile radionuclides such as radiocesium, there is only limited information on atmospheric behaviors of FDNPP-derived radiostrontium (^{89}Sr (half life: 50.53 d) and ^{90}Sr (half life: 28.79 y))[8] because radiostrontium was a minor release component due to un-volatile properties of oxide [9]. Total atmospheric emissions of ^{89}Sr and ^{90}Sr from the FDNPP accident were estimated to be about one and two orders of magnitude lower than ^{137}Cs , respectively[1]. Although radiostrontium is a minor component within the FDNPP-derived radionuclides, knowledge on radiostrontium derived from the FDNPP is important to have better understanding of characteristics of the FDNPP accident.

The monthly ^{90}Sr deposition have been monitored by Japanese Radioactivity Survey project since the late 1950s, in which major part has been performed by Meteorological Research Institute (MRI) [10-13]. After the FDNPP accident, the monthly ^{90}Sr deposition observed at Tsukuba was reported [8], in which the monthly ^{90}Sr deposition in March 2011 increased to $5.2 \pm 0.1 \text{ Bq m}^{-2}$, being more two orders magnitude greater than that in the late 2000s. However, there is no report about atmospheric dispersion of the FDNPP-derived ^{90}Sr in Japan.

Activity ratios of ^{89}Sr and ^{90}Sr are an important indicator to identify freshly released ^{90}Sr using ^{89}Sr with a short half-life. Although after the FDNPP accident ^{89}Sr was detected in airborne dust ($^{89}\text{Sr}/^{90}\text{Sr}$: ~15) [8] and seawater ($^{89}\text{Sr}/^{90}\text{Sr}$: 11.8 ± 0.8) [14], there is very limited

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information on ^{89}Sr in environmental samples because of complicated and time-consuming radiochemical procedure. Activity ratios of ^{90}Sr to ^{137}Cs are a useful tool to identify sources of radioactive releases and to have better understanding of environmental behaviors of these radionuclides [15,16]. The annual mean $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios of deposition samples in the period of 1980 to 1985 were a range from 0.43 to 0.77, which is due to radioactive fallout from the 26th Chinese atmospheric nuclear test conducted in October 1980 [17]. In May 1986 just after the Chernobyl accident, the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in a Tsukuba deposition sample decreased to 0.0094. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in deposition samples varied in the 1990s and 2000s due to effects of resuspension and terrestrial processes [13,18,19]. In March 2011 just after the FDNPP accident, the marked low $^{90}\text{Sr}/^{137}\text{Cs}$ ratio (about 2×10^{-5}) was observed in the deposition sample collected at Tsukuba [8]. To have better understanding of environmental behaviors of the FDNPP-derived radionuclides and the FDNPP accident sequence, therefore, it is important to examine the spatial and temporal variations of the $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in deposition samples.

In this paper, we describe spatial distributions and temporal variations of the monthly ^{90}Sr deposition observed in the central and eastern Honshu Island, Japan, after the FDNPP accident, and discuss total atmospheric emission of ^{90}Sr from the FDNPP.

Sampling and Method

Monthly ^{90}Sr deposition data recorded in Environmental Radioactivity database[20] were used for analysis of its temporal variation. Monthly radioactivity deposition samples have been collected at radioactivity monitoring stations by Meteorological

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Research Institute (MRI, Tsukuba) and local government laboratories. The monitoring stations in central and eastern regions in Japan (latitudinal range: 35°N – 40°N, longitudinal range: 138°E – 141°E, Figure 1) are as follows; Akita (39.72°N, 140.13°E), Morioka (39.68°N, 141.13°E), Yamagata (38.25°N, 140.33°E), Futaba (37.40°N, 140.99°E), Utsunomiya (36.60°N, 139.94°E), Hitachinaka (36.39°N, 140.43°E), Maehashi (36.40°N, 139.10°E), Tsukuba (36.1°N, 140.1°E), Tokyo (35.71°N, 139.70°E), Ichihara (35.53°N, 140.07°E), Chigasaki (35.33°N, 139.38°E), Shizuoka (35.00°N, 138.39°E), Kofu (35.67°N, 138.55°E), Nagano (36.64°N, 138.18°E), Niigata (37.85°N, 138.94°E), and Toyama (36.70°N, 137.10°E). Monthly deposition samples (rainwater and falling dust) were collected by rainwater samplers with surface areas of 0.5 m²(4 m² for MRI), which are usually installed on the roof of main monitoring building in each monitoring station. Monthly rainwater and falling dust samples was collected in appropriate bottles on the first day morning of every month.

We briefly describe the method of sample preparation and ⁹⁰Sr measurements in detail described elsewhere [21]. ⁹⁰Sr analysis has been performed by MRI and Japan Chemical Analysis Center (JCAC). Water sample was dried on a hot plate. The resultant residues were weighted after drying in an oven at 110°C and then transferred to a plastic container. Dried residue sample in a plastic container was subjected by gamma spectrometry. After the gamma survey, the deposition samples collected by local government laboratories were transported to JCAC. A part of the deposition sample was subjected to ⁹⁰Sr measurements. The deposition samples were decomposed with conc. HNO₃ and H₂O₂, and dissolved in HNO₃ solution. To separate Sr fraction from other natural and anthropogenic beta-emitters, carbonate, oxalate, chromate and iron hydroxide precipitation techniques were performed successively, and the Sr fraction was finally purified by fuming HNO₃ technique or ion exchange technique basically according to MEXT manual [21,22]. After that, Sr was precipitated as carbonate and fixed on a 5C filter paper, which was subjected to beta counting. After the activity measurement sources were left for several weeks to achieve ⁹⁰Sr-⁹⁰Y radioequilibrium, their beta-activities were measured using a low-background 2π gas-flow counter (Tennelec LB5100) and determined with a calibration curve, which was made from a relationship between known amounts of ⁹⁰Sr activity and SrCO₃. Counting efficiency (⁹⁰Sr-⁹⁰Y) was in a range of 50 to 70%. Chemical yield was determined from recovery of added stable Sr. In this case, treatment to remove contribution of ⁸⁹Sr has been carried out [8]. The ⁹⁰Sr activity was decay-corrected at mid-sampling. The detection limits of ⁹⁰Sr were approximately 3.5 mBq m⁻² (MRI) and approximately 50 mBq m⁻² (JCAC), respectively.

Results and Discussion

The spatial distributions of monthly ⁹⁰Sr deposition in March 2011 are depicted in Figure 1A. The monthly ⁹⁰Sr depositions in the sites within 300 km apart from the FDNPP, which were in the range from 0.89 ± 0.023 Bq m⁻² to 470 ± 17 Bq m⁻², are more than one order of magnitude greater than pre-FDNPP accident level [8,13,23]. On the other hand, in contrast of ¹³⁷Cs [5], ⁹⁰Sr activities in some locations of Kofu (35.65°N, 138.57°E; inland site) and the Japan Sea side sites (Niigata: 37.91°N, 139.04°E and Akita: 39.72°N, 140.10°N) were less than the detection limit. The highest monthly ⁹⁰Sr deposition was observed at Futaba about 5 km from the FDNPP, which is higher than the maximum monthly ⁹⁰Sr deposition (170 Bq m⁻²) originating from the 1961-1962 large-scale atmospheric nuclear testing observed at Koenji (Tokyo) in 1963 [10-12]. The relatively high ⁹⁰Sr depositions occurred at Hitachinaka and Tsukuba, corresponding to one of major

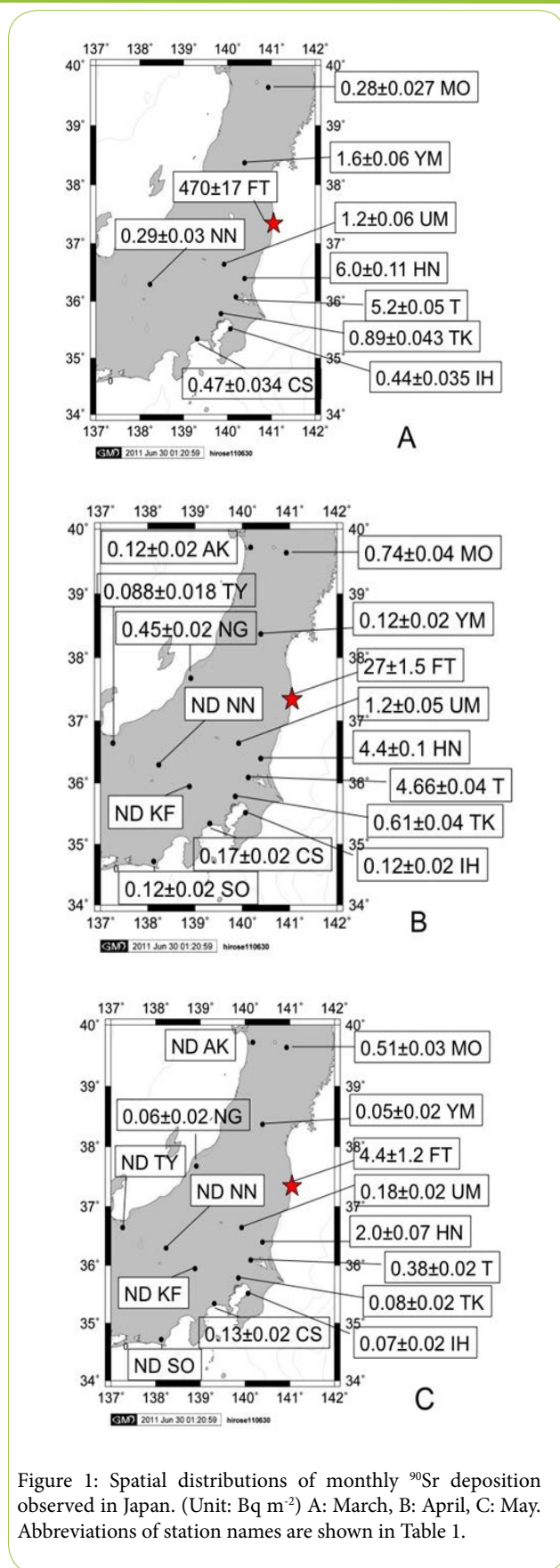


Figure 1: Spatial distributions of monthly ⁹⁰Sr deposition observed in Japan. (Unit: Bq m⁻²) A: March, B: April, C: May. Abbreviations of station names are shown in Table 1.

pathway of the radioactive plume derived from the FDNPP accident [24]. The spatial distribution of the ^{90}Sr deposition in March 2011 roughly corresponded to high ^{137}Cs deposition area depicted by aerial monitoring [1]. These findings suggest that ^{90}Sr detected in the deposition samples collected in March 2011 is derived from the atmospheric emission due to the FDNPP accident, which is supported detection of ^{89}Sr (half life: 50.53 d) in aerosol samples [8]. In April 2011 as shown in Figure 1B, a region detected ^{90}Sr in deposition samples spread to coastal zone of the central Honshu-Island and coastal sites in the Japan Sea, although the monthly ^{90}Sr depositions in the sites detected in March 2011 decreased. The temporal change of the affected area, where the FDNPP-derived ^{90}Sr was observed, is similar to that of ^{137}Cs [5]. In May 2011 (Figure 1C), the monthly ^{90}Sr deposition in all sites decreased. The spatial distribution of the ^{90}Sr deposition suggests that the atmospheric deposition of the FDNPP-derived ^{90}Sr cannot result in marked enhance of ^{90}Sr deposition density in land surface except area adjacent to the FDNPP. Measurements of ^{90}Sr in soil revealed that a small increase of ^{90}Sr was observed in the exclusion zone near the FDNPP, in which an average ^{90}Sr concentration in surface soil was 17.5 Bq kg^{-1} with a range from 1.4 to 80.8 Bq kg^{-1} whereas out of the exclusion zone there was no significant increase of soil ^{90}Sr having an average ^{90}Sr of 4.6 Bq kg^{-1} with a range from 1.6 to 20.6 Bq kg^{-1} , which is the same level as that in the pre-FDNPP era [25,26].

In order to elucidate depositional behaviors of the Fukushima-derived radionuclides, it is important to examine the temporal change of the monthly ^{90}Sr deposition. Temporal variations of the monthly ^{90}Sr depositions during the period from March 2011 to March 2012 are shown in Figure 2. The monthly ^{137}Cs deposition except Morioka exhibited decrease tendency during the period of March to May. We calculated apparent atmospheric half-lives (AAHL) of the Fukushima-derived ^{90}Sr (Table 1). The AAHLs at Futaba and Yamagata are similar to that of the FDNPP-derived ^{137}Cs during the corresponding period [5], whereas the AAHLs in the Kanto plain (19 -44 days) are longer than that of the FDNPP-derived ^{137}Cs . This finding suggests that the atmospheric behavior and/or emission processes of the FDNPP-derived ^{90}Sr were different from ^{137}Cs . After June 2011, the level of the monthly ^{90}Sr deposition except several sites decreased to less than detection limit. However, the monthly ^{90}Sr deposition at Futaba near the FDNPP with a range of 0.27 to 2.2 Bq m^{-2} showed no decrease until March 2012. The monthly ^{90}Sr deposition at Hitachinaka about 100 km south the FDNPP were measurable until November 2011, although ^{90}Sr was detected in several samples collected in 2012. For Tsukuba, the monthly ^{90}Sr deposition decreased until July 2011 and was kept at a constant level (0.01 to 0.077 Bq m^{-2}), which is about one order of magnitude larger than the pre-FDNPP level (2009-2010: ND to 0.0077 Bq m^{-2}). These findings suggest that the FDNPP-derived ^{90}Sr at least affected in atmosphere of the Kanto plain until early 2012, although its radiological contribution was very small.

$^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples are an effective tool to have better understanding of emission and resuspension processes of the FDNPP-derived radionuclides [13,18,19]. We examine the spatial distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in the deposition samples collected in March 2011. The result is shown in Figure 3. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios, which ranged from 0.88×10^{-4} to 3.5×10^{-4} with a median of 1.8×10^{-4} , are approximately constant spatially. These ratios are similar with the $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in soil of the exclusion zone with a range from 0.57×10^{-4} to 3.0×10^{-4} , although ^{90}Sr in soil contained contribution of ^{90}Sr derived from global fallout [25,27]. The median of

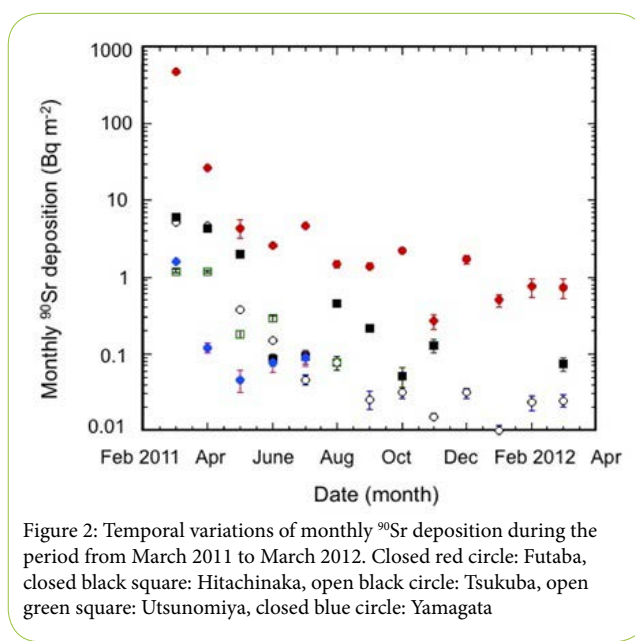


Figure 2: Temporal variations of monthly ^{90}Sr deposition during the period from March 2011 to March 2012. Closed red circle: Futaba, closed black square: Hitachinaka, open black circle: Tsukuba, open green square: Utsunomiya, closed blue circle: Yamagata

Station name	Location	AAHL (days)
MO	39.68°N, 141.13°E	27±19*
YM	38.25°N, 140.33°E	11±2
FB	37.40°N, 140.99°E	11±3
HN	36.39°N, 140.43°E	44±16
UM	36.60°N, 139.94°E	42±21
T	36.10°N, 140.10°E	36±20
MH	36.40°N, 139.10°E	19±1
TK	35.71°N, 139.70°E	30±11
IK	35.53°N, 140.07°E	18±3
CS	35.33°N, 139.38°E	26±6

Table 1: Apparent atmospheric half-lives of FDNPP-derived ^{90}Sr during the period of March 2011 to May 2011.

*AAHL was calculated in the period of April to July because ^{90}Sr maximum was observed in April 2011.

Abbreviations: AAHL: Apparent Atmospheric Half Life, Monitoring stations, AK: Akita, MO: Morioka, YM: Yamagata, FB: Futaba, UM: Utsunomiya, HN: Hitachinaka, T: Tsukuba, TK: Tokyo, IK: Ichihara, CS: Chigasaki, SO: Shizuoka, KF: Kofu, NN: Nagano, NG: Niigata, TY: Toyama, MH: Maehashi.

the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in the deposition samples reflects that in the major radioactive emission into atmosphere just after the FDNPP accident. This finding allows us to estimate the atmospheric total ^{90}Sr emission from total ^{137}Cs release. Many researchers have estimated the total atmospheric release of ^{137}Cs derived from the FDNPP accident because of one of major emission radionuclides and its long physical and environmental half-lives, which has been summarized in several reviews [1,2,28]. Aoyama et al. [29] concluded that the most reliable value of the total atmospheric ^{137}Cs release is $15 - 20 \text{ PBq}$. The total atmospheric ^{90}Sr emission is calculated to be $2.7 - 3.6 \text{ TBq}$ from the median of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios and the total atmospheric ^{137}Cs release, which corresponds to $0.00052 - 0.00069 \%$ of the total core inventory ($5.22 \times 10^2 \text{ PBq}$). This estimated total atmospheric emission of ^{90}Sr is more than one order of magnitude lower than the previous estimate (139 TBq) [1].

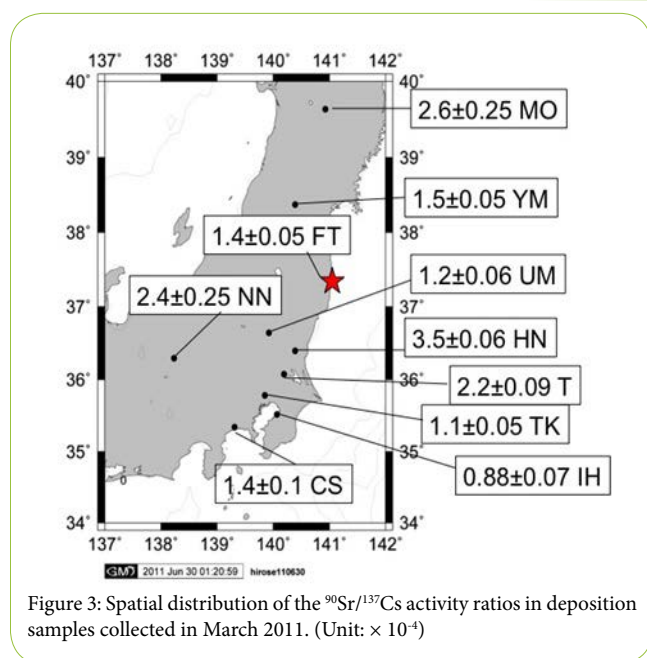


Figure 3: Spatial distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples collected in March 2011. (Unit: $\times 10^{-4}$)

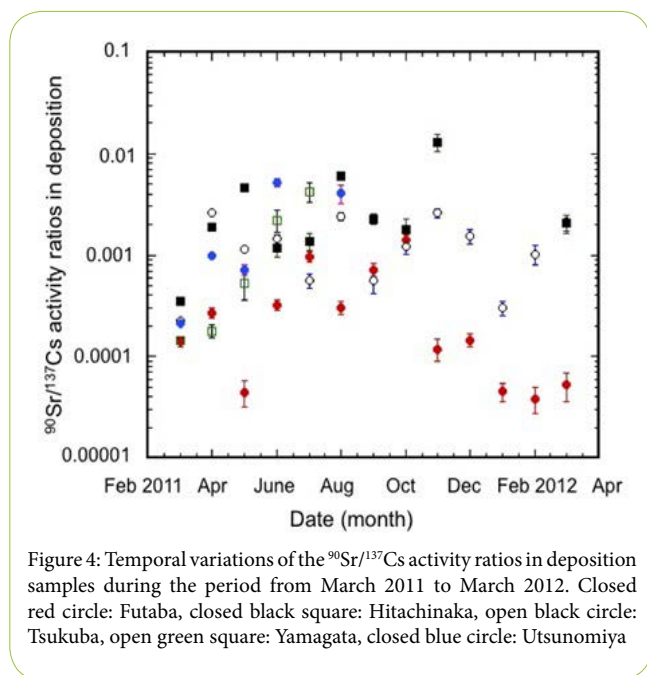


Figure 4: Temporal variations of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples during the period from March 2011 to March 2012. Closed red circle: Futaba, closed black square: Hitachinaka, open black circle: Tsukuba, open green square: Yamagata, closed blue circle: Utsunomiya

The temporal variations of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples collected at Futaba, Hitachinaka, Tsukuba, Utsunomiya and Yamagata are shown in Figure 4. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios at Futaba near the FDNPP showed around 2×10^{-4} , although relatively high and low ratios occurred on August – October 2011 and on January – March 2012, respectively. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios at Hitachinaka, Tsukuba and Utsunomiya, corresponding to the Kanto area, increased from April to June 2011 and have exhibited high values of 0.00055 – 0.013 after April 2011, although its large temporal and spatial variations were observed. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios at Yamagata northwest of the FDNPP increased after May 2011. These findings suggest that atmospheric emission processes and dispersion behaviors of ^{90}Sr and ^{137}Cs after April 2011 differed from the initial release in March 2011. To explain long-term atmospheric effects of the FDNPP-derived radionuclides,

resuspension hypothesis that ^{90}Sr and ^{137}Cs in deposition were supported by local suspension of surface soil particles has been proposed [6,30,31]. However, it is difficult to explain long-term variations of the $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in the deposition by this hypothesis because ^{90}Sr and ^{137}Cs in local soil are too low to support their deposition levels [6]. One of other possible processes is atmospheric emission from highly contaminated area including the FDNPP [7]. The relatively high ^{90}Sr concentrations (up to 1 kBq kg^{-1}) were observed in surface soil near the FDNPP collected in the period of December 2011 to July 2012, which correspond to higher $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in the major emission [32]. The post accident sporadic release of radionuclides may be contained more enriched ^{90}Sr than in the initial release because TEPCO implemented selective removal of radiocesium in contaminated water at the first stage. In fact, one major sporadic release of radionuclides as a late emission was observed on August 19, 2013, in which higher ^{90}Sr contamination and higher $^{90}\text{Sr}/^{137}\text{Cs}$ ratio (0.04) in a soil sample corresponding to the simulated plume line [33]. However, factors controlling the complicated temporal variations of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios are still unknown as mentioned by Igarashi et al. [8].

Conclusion

We summarized monitoring results of radiostrontium after the Fukushima Daiichi Nuclear Power Plant accident. Radiostrontium (^{89}Sr and ^{90}Sr) was released in the atmosphere due to the FDNPP accident. Since the maximum deposition on March 2011 just after the FDNPP accident, the monthly ^{90}Sr deposition in the sites decreased with time. The late effects of ^{90}Sr in deposition continued more than one year, although the area affected by ^{90}Sr is limited comparing with ^{137}Cs . For the initial emission in March 2011, the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples with a median of 1.8×10^{-4} were approximately constant. The total atmospheric emission of ^{90}Sr is estimated to be 2.7 – 3.6 TBq from the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio and the total release amount of ^{137}Cs . The $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratios in deposition samples showed large spatial and temporal variability, which reflects complicated atmospheric behaviors of ^{90}Sr and ^{137}Cs including post-accident sporadic releases from the FDNPP site.

Competing Interests

The author declares no financial competing interest.

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