



Radiation Emergency Medicine 2015 Vol. 4 No. 1 57-61

Note

# Radiocesium Inventory on the Campus of Nihon University, Koriyama, Fukushima, Japan

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Received 8 September 2014; revised 27 November 2014; accepted 2 December 2014

We have collected surface soil samples on the campus of Nihon University, Koriyama, Fukushima, Japan, after the Fukushima Dai-ichi Nuclear Power Plant accident for three years and measured their radiocesium ( $^{134}$ Cs and  $^{137}$ Cs) concentrations to understand the behavior of deposited radiocesium on soil surfaces in the campus. In 2011, the inventory of  $^{134}$ Cs and  $^{137}$ Cs ranged from 73 to 164 kBq m<sup>2</sup> with the arithmetic mean value of 110 ± 33 kBq m<sup>2</sup>, and from 83 to 203 kBq m<sup>2</sup> with the arithmetic mean value of 134 ± 42 kBq m<sup>2</sup>, respectively. In 2012 and 2013, we found radiocesium inventories were rapidly decreasing at most sampling stations because of university decontamination work programs to remove the surface soil to a depth of 5 cm. Slightly increasing radiocesium inventories were observed at some stations, however, where soil and grass covered soil areas met. We considered that the elevated radiocesium inventories were affected by resuspension of soil dust. The  $^{137}$ Cs residual rate at areas that had not been decontaminated was estimated at approximately 50% during 2012 to 2013.

Key words: Fukushima Dai-ichi Nuclear Power Plant accident, radiocesium inventory, Koriyama City

## 1. Introduction

On 11 March 2011, the Great East Japan Earthquake (magnitude 9.0) and its unexpectedly high tsunami caused widespread damage<sup>1)</sup>. Facilities of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) of Tokyo Electric Power Company (TEPCO) were damaged seriously by this disaster, and large amounts of radionuclides were released from the plant reactors due to the events that

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followed<sup>2, 3)</sup>. The major radionuclides released that must be considered for radiation protection of the public's health were radioiodine (<sup>131</sup>I: half-life of 8.01 d) and radiocesium (<sup>134</sup>Cs: half-life of 2.06 y, <sup>137</sup>Cs: half-life of 30.1 y). Total amounts of released <sup>131</sup>I and <sup>137</sup>Cs from the FDNPP to the atmosphere were estimated to be  $1.5 \times 10^{17}$ and  $1.3 \times 10^{16}$  Bq, respectively and <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio of radiocesium was estimated as approximately  $1.0^{4)}$ . Some of the radiocesium released into the atmosphere was deposited on the ground surface and has affected the general environment<sup>5.8)</sup>. The Japanese Government surveyed of radionuclides released from the FDNPP that are in the surface soil for the purpose of constructing a contamination map<sup>9, 10)</sup>. The overall soil contamination by

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Fig. 1. Sampling stations on the Koriyama campus of Nihon University.

radiocesium has been revealed from this map. It has been reported that there is a good correlation between <sup>137</sup>Cs inventory in surface soils and the 1 cm dose equivalent rate<sup>11)</sup>. And it is known from experiences in the Chernobyl accident that radiocesium deposited on the ground surface affects atmospheric radiocesium concentrations and internal exposure by inhalation<sup>12)</sup>. Therefore, it is important to understand the distribution and inventory of radiocesium continuously.

The College of Engineering of Nihon University is located in Koriyama City which is in the central area of Fukushima Prefecture, and is approximately 60 km west from the FDNPP. The campus area is approximately 380,000 m<sup>2</sup> and about 4,800 students are enrolled on this campus. Since the FDNPP accident, radiation dose measurements and decontamination work programs have been carried out continuously<sup>13)</sup>. The half-life of radiocesium is longer than that of the other radionuclides released from the FDNPP. The inventory of radiocesium around the campus was reported by the Japanese Government<sup>9, 10)</sup>. However, the observation results in the vicinity of the campus are limited. It is important to understand the details of the radiocesium inventory on the campus to ensure the safety of students and staff.

We determined radiocesium inventory on the Koriyama campus of Nihon University from 2011. In this paper, we have reported the distribution and annual change of radiocesium inventory on the campus for 3 years.



Fig. 2. Variation of monthly average 1 cm dose equivalent rate at the main gate of the campus and the athletics stadium.

## 2. Materials and Method

All samples were collected on the Koriyama campus of Nihon University. Details of the sampling stations are shown in Figure 1 and Table 1. We selected these points where students are not used very often. 1 cm dose equivalent rates were obtained by a pocket survey meter. Continuous results and decontamination activities have been reported on the Nihon University Home Page<sup>13)</sup>. Figure 2 shows the monthly average 1 cm dose equivalent rates at the campus main gate and athletics stadium which correspond to St-1 and St-9, respectively. In June 2011, dose equivalent at the main gate was rapidly decreasing due to paved road cleaning with high-pressure water cleaning tools. In addition, that of the athletics stadium decreased due to decontamination work that involved the removing of surface soil to a depth of 5 cm in February 2012. Moreover, dose equivalent at the main gate was further decreased by surface soil removing decontamination work.

Surface soil samples were collected at 10 sampling station using 100 ml stainless steel sampling tubes (DIK-1801, Daiki, Japan) once a year (Table 1). This type sampling tube is generally used for collecting an undistributed 5 cm deep soil layer. Three soil samples were taken at each sampling station. The three collected samples were placed into a weighed water-vapor-tight laminate bag (AL-30L, Seisannipponsha, Japan), and they were mixed by shaking in the close bag. After mixing and reweighing, drying agent (Z-150, OZO Kagakugiken Co., Ltd., Japan) was also placed in the bag to dry the sample<sup>7</sup>. The samples were dried in this way for about 2 weeks. After drying, the dry weight of samples was determined. The soil samples were then packed into separate plastic containers after removing stones and plant roots by hand picking. The radiocesium (134Cs: 604 keV, 137Cs: 662 keV) concentrations in the samples were determined using a

Doint	Coordinates		Situation	Sampling day		
Point				2011	2012	2013
St-1	37°21'42.02"N	140°22'54.97"E	Near the main gate (Grass covered)	02-Nov	28-Jun	03-Apr
St-2	37°21'38.79"N	140°22'54.78"E	Inside of green belt (Small forest)	02-Nov	28-Jun	03-Apr
St-3	37°21'33.99"N	140°22'50.56"E	Road side (Grass covered)	01-Oct	28-Jun	03-Apr
St-4	37°21'33.33"N	140°22'48.44"E	Edge of running track (Bare soil)	30-Sep	28-Jun	03-Apr
St-5	37°21'31.49"N	140°22'44.71"E	Edge of running track (Bare soil)	02-Nov	28-Jun	03-Apr
St-6	37°21'32.04"N	140°22'53.00"E	Side of building (Grass covered)	02-Nov	28-Jun	03-Apr
St-7	37°21'30.72"N	140°22'54.23"E	Edge of baseball ground (Bare soil)	30-Sep	28-Jun	03-Apr
St-8	37°21'27.83"N	140°22'54.35"E	Edge of baseball ground (Bare soil)	02-Nov	28-Jun	03-Apr
St-9	37°21'33.44"N	140°23'11.01"E	Inside of running track (Bare soil)	02-Nov	28-Jun	03-Apr
St-10	37°21'31.29"N	140°23'11.60"E	Inside of running track (Bare soil)	02-Nov	28-Jun	03-Apr

Table 1. Summary of sampling stations on the campus of Nihon University

high-purity Ge detector (GX-3018, Canberra, USA) for 20,000 to 80,000 s. The efficiency was calibrated by using sealed radioactive standard sources (MX033U8PP, Japan Radioisotope Association, Japan). We checked it using standard soil samples (JASC0471-0473, The Japan Society for Analytical Chemistry) which are similar to our samples (geometry and matrix). Measured values were corrected for radioactive decay to the sampling day.

#### 3. Results and Discussion

Table 2 summarizes radiocesium inventory and <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio at the sampling stations. In 2011, <sup>134</sup>Cs inventory ranged from 73 to 164 kBq m<sup>-2</sup> with the mean value of  $110 \pm 33$  kBq m<sup>-2</sup> and <sup>137</sup>Cs inventory ranged from 83 to 203 kBg m<sup>-2</sup> with the mean value of  $134 \pm 42$  kBg m<sup>-2</sup>. The radiocesium inventory in Koriyama City has been reported for a contamination map<sup>10</sup>. Among mapped values, the radiocesium inventory in the campus vicinity (5 locations) ranged from 46 to 180 kBq m<sup>-2</sup> for <sup>134</sup>Cs, and from 52 to 180 kBq m<sup>-2</sup> for <sup>137</sup>Cs. Our data were similar to the reported values. It is known that mobility of deposited radiocesium by chemical and biological processes is limited for cesium fixed to clay minerals<sup>14, 15)</sup>. We thought that the deposited radiocesium moved only slightly on the campus ground surface by a physical process like wind erosion. The average <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio was 0.82.

After the survey of 2011, decontamination work to remove the surface soil layer to a depth of 5cm was done at St-2, St-4, St-5, St-7, St-8 and St-9 from 19 January to 20 March 2012<sup>13)</sup>. Radiocesium inventories in 2012 were

found to be rapidly decreasing at most sampling stations, <sup>134</sup>Cs and <sup>137</sup>Cs inventories ranged from 1.3 to 89 kBq m<sup>-2</sup> and from 2.0 to 138 kBq m<sup>-2</sup>, respectively. During July to October 2012, decontamination work was done at St-1 and St-313, and radiocesium inventory has continued to decrease. Figure 3 shows the annual variation of <sup>137</sup>Cs inventory on the campus. The decreasing trend varied depending on location and type of decontamination work. However, elevated radiocesium inventories compared with those of last year were observed at some stations (St-2, St-3, St-4, St-5, St-7 and St-8). Radiocesium inventory is controlled by several factors: 1) direct dispersion of radiocesium from the FDNPP, 2) resuspension and erosion of soil particles from the ground surface, and 3) physical decay. After the sampling days of 2011, radiocesium deposition was observed in Fukushima Prefecture<sup>16)</sup>. The results of the <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio in the soil surface layer agreed with the estimated <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio of radiocesium from the FDNPP accident (approximately 1.0)<sup>4)</sup>. Therefore, the effect of new direct dispersion from the FDNPP was excluded. The stations where elevated radiocesium inventories were observed were where soil and grass covered soil areas met and a small forest. We considered that elevated radiocesium inventories were affected by environmental processes like re-suspension of soil dust and surface runoff from other areas.

No decontamination work has been carried out at St-10. We calculated the environmental residual rate of <sup>137</sup>Cs using reported atmospheric deposition data and our inventory data of St-10. Atmospheric radiocesium deposition at Fukushima City has been reported since 0.82

the campus of Nihon University, 2011 to 2013									
	2011								
Point	<sup>134</sup> Cs (kBq m <sup>-2</sup> )	<sup>137</sup> Cs (kBq m <sup>-2</sup> )	<sup>134</sup> Cs/ <sup>137</sup> Cs						
St-1	$8.3E + 01 \pm 1.1E + 00$	$1.0E + 02 \pm 1.2E + 00$	0.83						
St-2	$1.6E + 02 \pm 1.8E + 00$	$2.0E + 02 \pm 2.0E + 00$	0.81						
St-3	$1.0E + 02 \pm 1.3E + 00$	$1.2E + 02 \pm 1.5E + 00$	0.84						
St-4	$1.3E + 02 \pm 1.5E + 00$	$1.5{\rm E}\!+\!02\pm1.7{\rm E}\!+\!00$	0.84						
St-5	$8.1E + 01 \pm 1.2E + 00$	$1.1E + 02 \pm 1.4E + 00$	0.77						
St-6	$9.3E + 01 \pm 1.1E + 00$	$1.2E + 02 \pm 1.2E + 00$	0.80						
St-7	$7.3E + 01 \pm 1.1E + 00$	$8.3{\rm E}\!+\!01\pm1.2{\rm E}\!+\!00$	0.88						
St-8	$1.6E + 02 \pm 1.4E + 00$	$2.0\rm{E}{+}02 \pm 1.6\rm{E}{+}00$	0.81						
St-9	$1.2E + 02 \pm 1.4E + 00$	$1.5{\rm E}\!+\!02\pm1.6{\rm E}\!+\!00$	0.81						
St-10	$8.7{\rm E}\!+\!01\pm1.5{\rm E}\!+\!00$	$1.1\rm{E}{+}02 \pm 1.6\rm{E}{+}00$	0.81						

Ave.  $\pm$  S.D.  $1.1E+02 \pm 3.3E+01$   $1.3E+02 \pm 4.2E+01$ 

Table 2. Radiocesium inventory and <sup>134</sup>Cs/<sup>137</sup>Cs ratio determined on

2012 <sup>134</sup>Cs/<sup>137</sup>Cs Point 134Cs (kBq m-2) <sup>137</sup>Cs (kBq m<sup>-2</sup>) St-1  $6.1E + 01 \pm 1.7E - 01$  $9.2E + 01 \pm 2.1E - 01$ 0.66 St-2  $1.5E + 01 \pm 8.2E - 02$  $2.3E + 01 \pm 1.0E - 01$ 0.67  $8.9E + 01 \pm 2.7E - 01$  $1.4E + 02 \pm 3.4E - 01$ St-3 0.64 St-4  $1.3E + 00 \pm 2.1E - 02$  $2.0E + 00 \pm 2.5E - 02$ 0.66 St-5  $7.9E + 00 \pm 6.0E - 02$  $1.2E + 01 \pm 7.6E - 02$ 0.64  $6.7E + 01 \pm 2.1E - 01$  $1.0E + 02 \pm 2.5E - 01$ St-6 0.66 St-7  $3.3E + 00 \pm 2.3E - 02$  $5.1E + 00 \pm 3.0E - 02$ 0.64 St-8  $2.2E + 00 \pm 3.4E - 02$  $3.4E + 00 \pm 4.3E - 02$ 0.65 St-9  $2.2E + 00 \pm 3.6E - 02$  $3.3E + 00 \pm 4.5E - 02$ 0.68  $5.4E + 01 \pm 1.7E - 01$  $8.4E + 01 \pm 2.1E - 01$ St-10 0.65 Ave.  $\pm$  S.D.  $3.0E + 01 \pm 3.4E + 01$  $4.7E + 01 \pm 5.2E + 01$ 0.65 2013 <sup>137</sup>Cs (kBq m<sup>-2</sup>) <sup>134</sup>Cs/<sup>137</sup>Cs Point 134Cs (kBq m-2) St-1  $1.4E + 00 \pm 5.0E - 02$  $2.6E + 00 \pm 9.2E - 02$ 0.53 St-2  $7.4E + 01 \pm 4.0E - 01$  $1.5E + 02 \pm 8.3E - 01$ 0.50 St-3  $3.3E-01 \pm 3.7E-02$  $6.0E-01 \pm 5.2E-02$ 0.56 St-4  $1.5E + 00 \pm 6.1E - 02$  $3.1E + 00 \pm 1.1E - 01$ 0.50 St-5  $8.2E + 00 \pm 1.1E - 01$  $1.6E + 01 \pm 2.1E - 01$ 0.52 St-6  $2.5E + 01 \pm 2.1E - 01$  $4.8E + 01 \pm 4.2E - 01$ 0.52 St-7  $5.0E + 01 \pm 2.8E - 01$  $9.8E + 01 \pm 5.6E - 01$ 0.51 St-8  $2.8E + 00 \pm 7.4E - 02$  $5.6E + 00 \pm 1.4E - 01$ 0.50 St-9  $9.3E-01 \pm 4.4E-02$  $1.9E + 00 \pm 8.1E - 02$ 0.48 St-10  $1.4E + 01 \pm 7.6E - 02$  $2.8E + 01 \pm 1.5E - 01$ 0.51 Ave.  $\pm$  S.D.  $1.8E + 01 \pm 2.5E + 01$  $3.5E + 01 \pm 5.0E + 01$ 0.51

March 2011<sup>16)</sup>. In Koriyama City, the data have been reported since January 2012. After January 2012, a good correlation was observed in Fig. 4 for the monthly radiocesium deposition of Fukushima and Koriyama Cities (r=0.47, P<0.05). We estimated monthly radiocesium deposition in Koriyama during March to December 2011. And the residual rate of <sup>137</sup>Cs (RE) at the non-decontaminated area was calculated by the following equation:

$$RE = \frac{A_T - (A_P + D)}{A_P + D}$$



Fig. 3. Inventory of <sup>137</sup>Cs on the campus of Nihon University



**Fig. 4.** Relationship of monthly <sup>137</sup>Cs deposition between Fukushima and Koriyama Cities from January 2012 to December 2013.

where  $A_T$  and  $A_P$  are observation data of <sup>137</sup>Cs inventory (Bq/m<sup>2</sup>) in the current year and previous year, D is deposition rate during the time from the soil sampling day of the current year to that of the previous year. As the result, the <sup>137</sup>Cs residual rates are gradually degreasing, and 2012-2013 was approximately 50%.

### 4. Conclusion

We reported radiocesium inventory during 3 years on the Koriyama campus of Nihon University after the FDNPP accident. Radiocesium inventories were generally decreasing at most sampling stations as a result of decontamination work programs. However, slightly increasing radiocesium inventories were observed at some stations. We considered that the elevated radiocesium inventories have been affected by mobility of soil dust as a physical process. The <sup>137</sup>Cs residual rate in 2012-2013 was estimated at approximately 50%. In the future, we will continue our observation of radiocesium inventory on the campus.

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