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Estimation of dry deposition velocities of radionuclides released by the accident at the Fukushima Dai-ichi Nuclear Power Plant

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On the basis of environmental radiation monitoring results at the Nuclear Fuel Cycle Engineering Laboratories, Japan Atomic Energy Agency, following the Fukushima Dai-ichi Nuclear Power Plant accident, the dry deposition velocities of radionuclides on the ground were estimated. As a result, the estimated dry deposition velocities for Iodine-131, Cesium-134, Cesium-136 and Cesium-137 were in the order of 1 mm/s, and were the same order of those estimated in other studies. The estimated dry deposition velocities varied according to the variations of the ratios of the particulate and gaseous forms of their radionuclides and meteorological conditions such as wind speed.

Keywords: *dry deposition velocity; Iodine-131; Cesium-134; Cesium-136; Cesium-137; Fukushima Dai-ichi Nuclear Power Plant accident; Nuclear Fuel Cycle Engineering Laboratories; Tokai-mura; Ibaraki*

1. Introduction

The radioactive substances were released by the accident at the Fukushima Dai-ichi Nuclear Power Plant (1F-NPP) of Tokyo Electric Power Company and deposited on the ground, which caused the increase of an air dose rate. To investigate the dry deposition phenomenon is important in order to clarify the increase of the dose rate, because the air dose rate significantly increased in the south-southwest area of 1F-NPP including Tokai-mura by dry deposition of the high-concentration plume released from the night of March 14 to the morning of March 15 [1]. The process of dry deposition is driven by a variety of physical, chemical and biological factors. Further complication arises from the fact that radionuclides, when released into the atmosphere, exist in various chemical and physical states [2].

The emergency monitoring of environmental radiation including the measurement of the air concentrations of radionuclides from 13 March 2011 to 23 May 2011 and the fallout from 15 March 2011 to 2 May 2011 was performed on the Nuclear Fuel Cycle Engineering Laboratories (NCL), Japan Atomic Energy Agency (JAEA), in addition to the routine monitoring of air dose rate, meteorological condition and so on.

In this paper, the dry deposition velocities of radionuclides, which were defined as the fallout divided by the time integrated air concentration, were estimated on the basis of their monitoring results when the radioactive plumes would reach the NCL.

2. Materials and method

The air and the fallout samplings in addition to the routine monitoring were performed at the NCL about 115 km south of the 1F-NPP. The location of the NCL is shown in **Figure 1**.

The measuring methods of the air concentrations of radionuclides were the following: The air samples were continuously collected by a portable dust and iodine sampler (model No. PNC-800) installed at the height of 1 m above the ground. The dust filter (Type He-40T made by Toyo-roshi Kaisya Ltd., Japan) and charcoal cartridge impregnated by TEDA (tri-ethylene-diamine) (Type CHC-50 made by Toyo-roshi Kaisha Ltd., Japan) were replaced after every 12 h as a matter of principle. However, the interval between the replacements was inversely proportional to the dose rate. As a result, the interval between the replacements was from 3 h to 7 d.

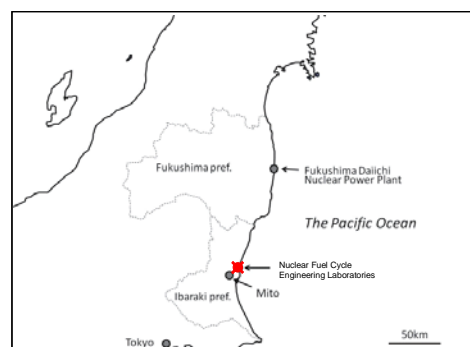


Figure 1. Location of Nuclear Fuel Cycle Engineering Laboratories, JAEA.

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The air concentrations were determined by gamma-ray spectrometry with a Ge detector. The details of the measurement procedures were described in the previous paper [3-4]. The radionuclides collected by the dust filter were considered to be particulate radionuclides, and those collected by the charcoal cartridge were considered to be gaseous radionuclides. The results of the variations of the air concentrations were shown in **Figure 2**.

The measuring methods of the fallout were as follows: The fallout samples were collected using a large bulk sampler covering an area of 0.5 m². The fallout was determined by gamma-ray spectrometry with Ge detector. The results of the variations of the fallout were shown in **Figure 3**.

The routine monitoring of air dose rate, wind speed and direction, rainfall, atmospheric humidity and atmospheric stability has been continuously performed before the accident at the NCL. **Figure 4** showed the variations of the dose rate measured and rainfall observed. The wind speed and direction was observed at the height of 15 m above the ground. **Figure 5** showed the variations of the wind speed and direction. **Figure 6** showed the variations of the atmospheric humidity

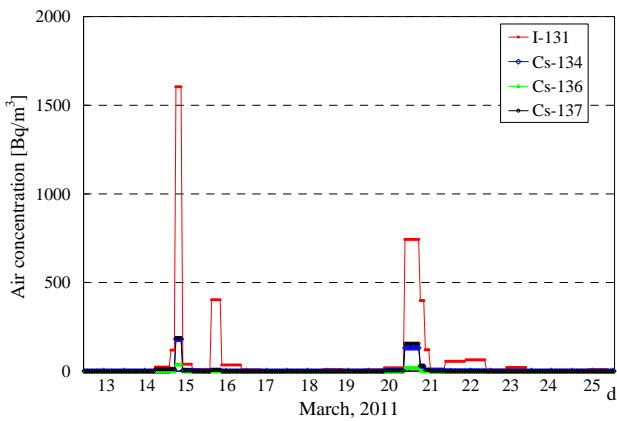


Figure 2. Temporal changes in the air concentrations of radionuclides (sum of particulate and gaseous forms).

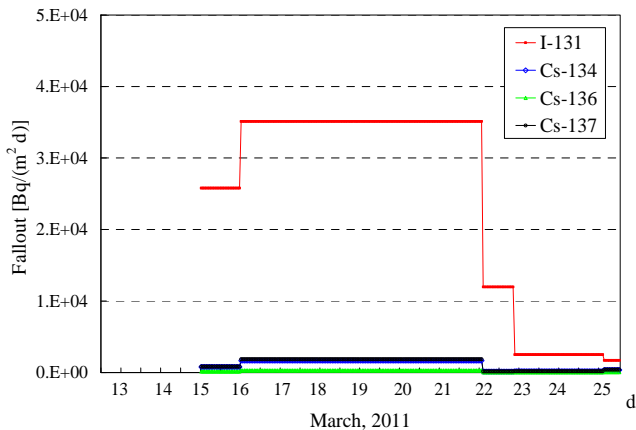


Figure 3. Temporal changes in the fallout of radionuclides.

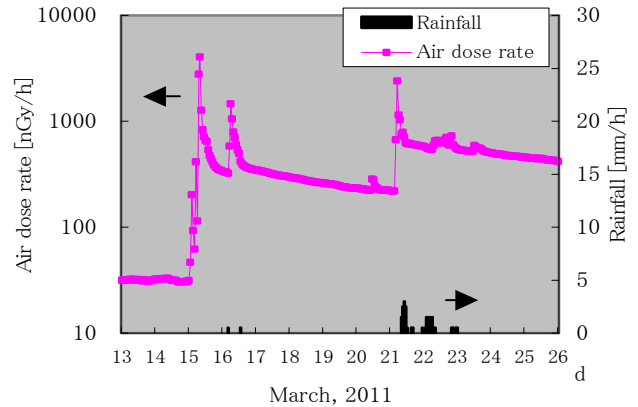


Figure 4. Temporal changes in the air dose rate measured and rainfall observed at the NCL.

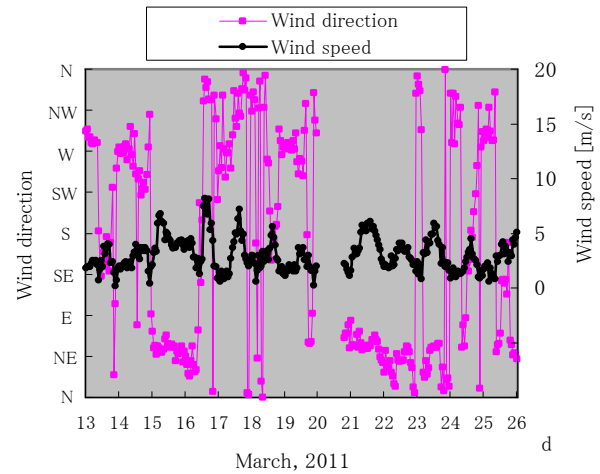


Figure 5. Temporal changes in wind speed and direction observed at the height of 15 m above the ground at the NCL.

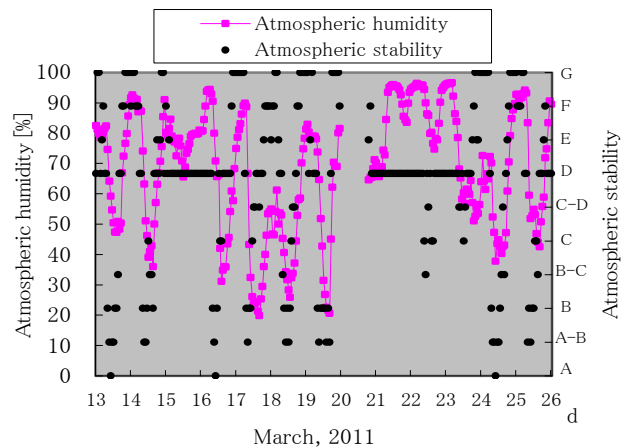


Figure 6. Temporal changes in atmospheric humidity and atmospheric stability at the NCL.

and the atmospheric stability. From Figures 4 and 5, while the wind blew from the north (1F-NPP) on 15, 16, and 21 March, the air dose rate increased with three peaks at 8 JST 15 March, 5 JST 16 March and 4 JST 21 March [3-4]. From this fact, the radioactive plumes reached the NCL at these dates. From the air concentrations and the fallouts measured during the two periods, that is, from 12 JST 15 March to 12 JST 16

March (period 1) and from 12 JST 16 March to 14 JST 22 March (period 2), in which intervals the fallout samples were replaced at around 15, 16 and 21 March, the dry deposition velocities of radionuclides were estimated. The dry deposition velocity (mm/s) is defined as the fallout (Bq/m^2) divided by the time integrated air concentration (Bqh/m^3).

3. Results and discussion

3.1. Estimation of dry deposition velocity

Table 1 showed the time integrated air concentrations, the fallout and the estimated dry deposition velocities for the periods 1 and 2. As shown in Table 1, the dry deposition velocities for Iodine-131 (I-131), Cesium-134 (Cs-134), Cesium-136 (Cs-136) and Cesium-137 (Cs-137) were estimated to be in the order of 1 mm/s for both periods.

Table 1. Time integrated air concentration, the fallout and the estimated dry deposition velocity for each radionuclide.

Period number		1	2
Period		12 JST March 15 - 12 JST March 16	12 JST March 16 - 14JST March 22
I-131	Patculate form	7.9E+02	4.6E+03
	Gaseous form	2.0E+03	5.6E+03
	Sum	2.8E+03	1.0E+04
	Ratio of gaseous to particulate forms	2.6	1.2
	Time integrated air concent- ration (Bqh/m ³)		
Cs-134	Patculate form	4.1E+01	1.3E+03
	Gaseous form	4.6E+00	5.0E+01
	Sum	4.5E+01	1.3E+03
	Ratio of gaseous to particulate forms	0.11	0.04
	Cs-136	Patculate form	7.5E+00
Gaseous form		6.8E-01	6.5E+00
Sum		8.2E+00	2.0E+02
Ratio of gaseous to particulate forms		0.09	0.03
Cs-137		Patculate form	4.9E+01
	Gaseous form	4.5E+00	5.9E+01
	Sum	5.4E+01	1.6E+03
	Ratio of gaseous to particulate forms	0.09	0.04
	Fallout (Bq/m ²)		
Dry deposition velocity (mm/s)	I-131	2.6E+04	2.1E+05
	Cs-134	7.8E+02	1.0E+04
	Cs-136	1.6E+02	1.8E+03
	Cs-137	8.3E+02	1.1E+04
	I-131	2.6E+00	5.7E+00
Cs-134	4.8E+00	2.1E+00	
Cs-136	5.4E+00	2.5E+00	
Cs-137	4.3E+00	1.9E+00	

3.2. Comparison of dry deposition velocities between radionuclides

The dry deposition velocity for I-131 was estimated to be 2.6 mm/s, while those for Cs-134, Cs-136 and Cs-137 were estimated to be about 5 mm/s for the period 1. The dry deposition velocity for I-131 was about half as large as those of Cs-134, Cs-136, and Cs-137. On the other hand, the dry deposition velocity for I-131 was estimated to be 5.7 mm/s, and those for Cs-134, Cs-136, and Cs-137 were estimated to be about 2 mm/s for the period 2. The dry deposition velocity for I-131 was

about three times larger than those of Cs-134, Cs-136, and Cs-137.

Table 1 showed the time integrated air concentrations of particulate and gaseous forms. From Table 1, the time integrated concentrations of the gaseous forms of I-131 were about two and a half times and about the same as large as those of the particulate forms for the periods 1 and 2, respectively. On the other hand, the time integrated concentrations of the gaseous forms of Cs-134, Cs-136 and Cs-137 were few for both the periods 1 and 2.

The gas was composed of unreactive and reactive gases. The dry deposition velocity of unreactive gas is one-tenth smaller than that of aerosol, and that of reactive gas is ten times larger than that of aerosol [5]. When the ratio of unreactive and reactive portions of I-131 varies, the dry deposition velocity of I-131 varies. If the ratio was different between the Periods 1 and 2, the dry deposition velocities of I-131 for these periods I-131 would vary.

The dry deposition velocities for Cs-134, Cs-136, and Cs-137 were the same for the period 1. Also, the dry deposition velocities for these radionuclides were the same for the period 2. This reason seems to be that the chemical forms were almost the same among these radionuclides for each the period.

The dry deposition velocities of Cs-134, Cs-136, and Cs-137 on the period 2 were about half smaller than those of the period 1.

The dry deposition velocity varies according to wind speed and atmospheric humidity [6]. From Figures 5 and 6, the wind blew from the northeast direction, with the wind speed from 1.3 to 4.5 m/s with the mean of 3.6 m/s, the atmospheric stability D (neutral), and the atmospheric humidity of 66.5% to 94.4% m/s with the mean of 80.0% during the period 1. On the other hand, the meteorological conditions varied largely during the period 2 because of the long period. The wind blew from north to south directions. The wind speed ranged from 0.2 to 8.2 m/s with the mean of 3.1 m/s. The atmospheric stability ranged from A to G. The atmospheric humidity ranged from 19.8% to 96.4% m/s with the mean of 64.1%. From Figure 4, the large deposition during the period 2 occurred on 4 JST to 10 JST 21 March (period 2'). During the period 2', the wind blew from northeast direction, with the wind speed from 3.1 to 5.7 m/s with the mean of 4.5 m/s, the atmospheric stability D (neutral), and the atmospheric humidity of 68.9% to 95.5% m/s with the mean of 83.5%. The wind speed during the period 2' was large compared to that during the period 1. The wind direction, atmospheric stability and atmospheric humidity were almost the same between the periods 1 and 2'. Due to the large wind speed, the dry deposition velocities of Cs-134, Cs-136, and Cs-137 on the period 2 would become smaller.

The rainfall has a great effect to the deposition due to wet deposition. There was little rainfall in the period 1, and the wet deposition did not influence to the deposition during the period 1. On the other hand, some

rainfall occurred from 8 JST 21 March to 8 JST 22 March on the period 2. So, the wet deposition might influence to the deposition during the period 2.

3.3. Comparison of dry deposition velocity with other study

After the Chernobyl accident, which site was very far from the NCL compared to the 1F-NPP, the concentrations of I-131 in air and in the pasture were determined at NCL, and the dry deposition velocities were estimated by the same method as this paper [7]. The estimated dry deposition velocities of I-131 ranged from 1.3 to 17 mm/s with the mean of 6.9 mm/s, and was in the same order as those of I-131 in this paper.

The Iodine-129 (I-129) had discharged from the Tokai Reprocessing Plant, and was determined by the environmental monitoring around the plant [8]. The I-129 is the same radioactive Iodine as I-131, and the chemical properties of these radionuclides seem to be the same. The estimated dry deposition velocities of I-129 ranged from 5 to 7.3 mm/s, and were also in the same order as that of I-131 for the period 2 in this paper.

Sehmel reviewed the papers about the particle and gas deposition and summarized the dry deposition velocities of many radionuclides and other than radionuclides under various meteorological and surface conditions [9]. In Sehmel's paper, the dry deposition velocities for Cs-137 were 0.3 – 1.5, -0.1 – 0.9 and 2 – 5 mm/s for the deposition surface of water, soil and grass, respectively. The fallout sampler in this paper was prepared on the roof of the concrete building, and this building was surrounded by forest. If the deposition surface of this paper was assumed to be grass, the deposition velocities in this paper were the same as that in Sehmel's paper.

4. Conclusion

On the basis of the environmental radiation monitoring results at the NCL following the Fukushima Dai-ichi Nuclear Power Plant accident, the dry deposition velocities of radionuclides were estimated. As a result, the estimated dry deposition velocities for I-131, Cs-134, Cs-136 and Cs-137 were in the order of 1 mm/s, and were in the same order of those in other studies. The estimated dry deposition velocity varied according to the variations of the ratios of the particulate and gaseous forms of their radionuclides and

meteorological conditions such as wind speed.

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