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I-131 Dispersion from the Stack to Environment of Soil and Grass Aradioisotope Production Facility, Serpong, Indonesia

Gatot Suhariyono^{1,*}, Haryoto Kusnoputranto², Kardono³, Syahrir⁴

¹PhD Student, Postgraduate Program on Environmental Science, University of Indonesia, Jakarta
National Nuclear Energy Agency (BATAN), Jakarta, Indonesia

²Department of Environmental Health, Faculty of Public Health and Postgraduate Program on Environmental Science
University of Indonesia, Jakarta, Indonesia

³Air Pollution, Center for Environmental Technology, Agency for Assessment and Application of Technology (BPPT)
Jakarta, Indonesia

⁴Nuclear Energy Regulatory Agency (BAPETEN)
Jakarta, Indonesia

* Corresponding author's email: g_suhariyono [AT] batan.go.id

ABSTRACT--- *Radioisotope production installation in Serpong produces and processes I-131 that can disperse into the environment around the Serpong Nuclear Area (SNA). I-131 produced routinely for medical purposes in hospitals and pharmacies. I-131 can cause thyroid cancer. I-131 is the largest radionuclides released from a nuclear accident than other radionuclides. Grass is a plant that can be used to reduce heat and air pollution. The measurement method of the I-131 activity concentration at the soil and the grass was carried on simultaneously with the production of I-131 radioisotope production facility in real time. Concentration of I-131 activity in the environment (soil and grass) around the SNA is still below the I-131 quality standard in the environment (63 Bq/kg). Concentrations of I-131 activity average at the grass ($\pm 75\%$) greater than the concentrations of I-131 activity in the soil ($\pm 25\%$). Concentration of I-131 activity average in the grass and in the soil highest of the seven research sites are in Jaletreng (4,200 m at the North of stack (360°)) by 1.62 and 0.41 Bq/kg respectively. Concentration of I-131 activity is high during high humidity (more than 80%) at night or rain, but when the sun rises the opposite effect.*

Keywords--- I-131, stack, soil, grass, environment, rain, humidity, sun

1. INTRODUCTION

Radioisotope production facility in Serpong, Indonesia, produces and processes I-131 that may disperse into the environment around the Serpong Nuclear Area (SNA). The Facility is in SNA that are surrounded by dense residential population. Residents in the area of 5 km radius of the facility in 2012 approximately 213,837 inhabitants [1, 2]. I-131 produced routinely for medical purposes in hospitals and pharmacies. I-131 can cause thyroid cancer, for both domestic and export. Radionuclide of I-131 is a beta and gamma-emitting radioactive. The radiation exposure of I-131 to human may cause thyroid cancer [3]. Radionuclide of I-131 is the largest radionuclides released from a nuclear accident than other radionuclides. Activity of I-131 radionuclide was released from the nuclear accident in Chernobyl (Russia) in 1986 amounted to 1,850 PBq (1 PBq = 1×10^{15} Bq), and in Fukushima (Japan) in March 2011 amounted to 400 PBq [4, 5].

The grass family is one of the largest in world, and grasses as individual plants are probably the most abundant plants in the world [6]. Grass is a plant that can be used to reduce heat and air pollution [7]. The soil is where the growth of grass. The research of I-131 release at stack of radioisotope production facility was carried to the environment (grass and soil) in real time.

2. METHODOLOGY

2.1 The measurement method of the I-131 concentration in stack

Measurement method of activity concentration (Bq/m^3) of I-131 radioactive at stack of radioisotope production is carried on using air monitoring systems are portable and can be monitored directly continuously every time. I-131

measurement system is shown in Figure 1. The air measurement system was performed with scintillation detector of LaBr₃ (Lanthanum Bromide) is a portable, need no nitrogen gas and is directly connected to a laptop computer, so that the air radioactivity that is released into the environment can be monitored activity concentrations per unit time periodically. Measurement of the flow rate is designed using a digital flow rate, so that the flow rate of air that can be sampled is monitored on a laptop continuously. Measurement of the I-131 activity concentration at the stack was carried on 6 times from December 2013 to March 2014.

Air that contain of I-131 from the stack is absorbed by the vacuum pump through Marinelli and forwarded to charcoal and digital flowmeter. The sampling flow rate (F) of the measuring device of I-131 concentration is 20.65 lpm. Air that enter and contain of I-131 to marinelli is detected with a LaBr₃ detector. The count results of I-131 can be read directly every 10 minutes in the form of spectrum at energy of 364 keV.

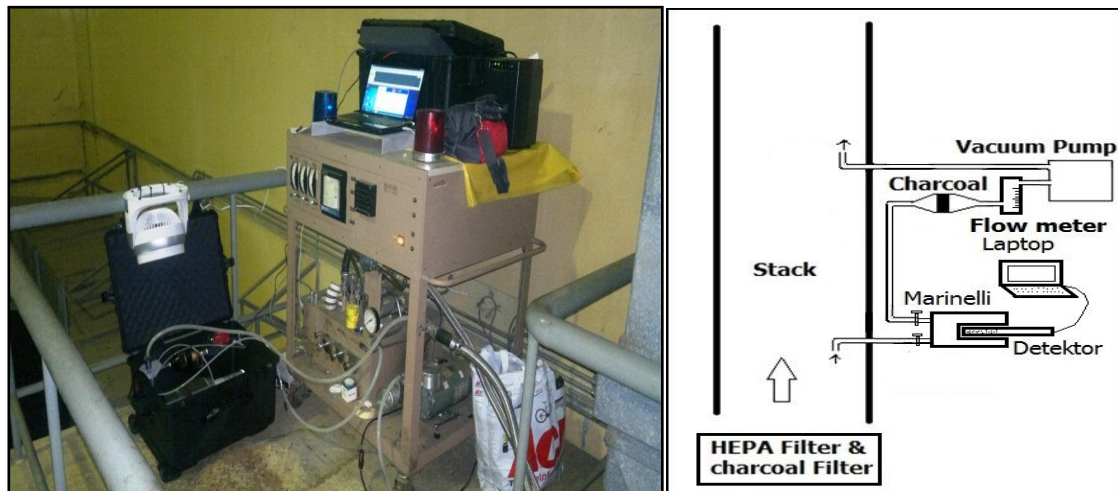


Fig.1 The air measurement of I-131 directly at the stack before being released into the environment

The concentration of I-131 that was obtained from the LaBr₃ detector and was calculated using the following equation:

$$C = \frac{(N_t - N_{Bg})}{Y \cdot \eta} \quad (1)$$

In this case C is concentration of I-131 (Bq/m³). η is counting efficiency of the LaBr₃ detector (cps/ (Bq/m³)). N_t is counts of I-131 from the LaBr₃ detector (counts). N_{Bg} is counts of background without I-131 (counts). Y is abundance of I-131 in nature (81.21%). Quality value of radioactivity standard levels in the air is 530 Bq/m³ based on regulation of PERKA BAPETEN No. 7/2013 [8].

2.2 The measurement method of I-131 concentration in soil and grass

The measurement method of I-131 activity concentration in soil was carried out together with the production of I-131 in the radioisotope production facility by taking a 1-2 kg of soil at a depth of 5 cm from the surface with a surface area of 0.36 m² in real time. Location of soil sampling should be free of trash dirt that there above the soil. Locations of soil sample is searched that flat and is open away from buildings and trees. Soil samples were weighed, and measured by gamma spectrometry using a NaI(Tl) detector. Based on UNSCEAR (2000), quality standard of I-131 in the soil is 63 Bq/kg [9]. The concentration of I-131 in soil samples is calculated by the following equation:

$$C = \frac{(N_t - N_{Bg})}{Y \cdot t \cdot \eta \cdot W} \quad (2)$$

In this case C is concentration of I-131 in soil sample (Bq/kg). η is counting efficiency of NaI(Tl) detector (cps/Bq). N_t is counts of I-131 in soil sample from NaI(Tl) detector (counts). t is counts time (s). W is weight of soil sample (kg).

The measurement method of the I-131 activity concentration in the grass is similar with the measurement method of the I-131 in the soil. This method is carried by cutting the grass at the I-131 measurement locations. The location of the grass cutting must be clean of leaves litter or other debris, and grass is located in an open location free from the influence of buildings or trees. The grass is cut approximately 1 cm above the grass hump or 2 cm above the soil surface with an area of 0.36 m² as much as the size of the 5 kg plastic and labeled the location name, date, time and name of the grass cutter. The grass samples is weighed and measured the content of I-131 by gamma spectrometry and NaI(Tl) detector in-situ at 364.48 keV energy. The concentration of I-131 in the grass is calculated by equation (2). Based on UNSCEAR (2000), quality standard of I-131 in the grass that same with quality standard of I-131 in the soil is 63 Bq/kg

[9]. There was 6 sampling locations of soil and grass (Fig. 2). The 6 locations are in Puri Serpong (2,200 m in the east of stack (90°)), in BATAN Indah (2,600 m in the North Northeast of stack (22,5°)), in Sengkol (825 m in the North of stack (360°)), in Pabuaran (1,900 m in the South of the stack (158°)), in Suradita (3,200 m in the West of the stack (292,5°)), and in Jaletreng (4,200 m in the North of stack (360°)).

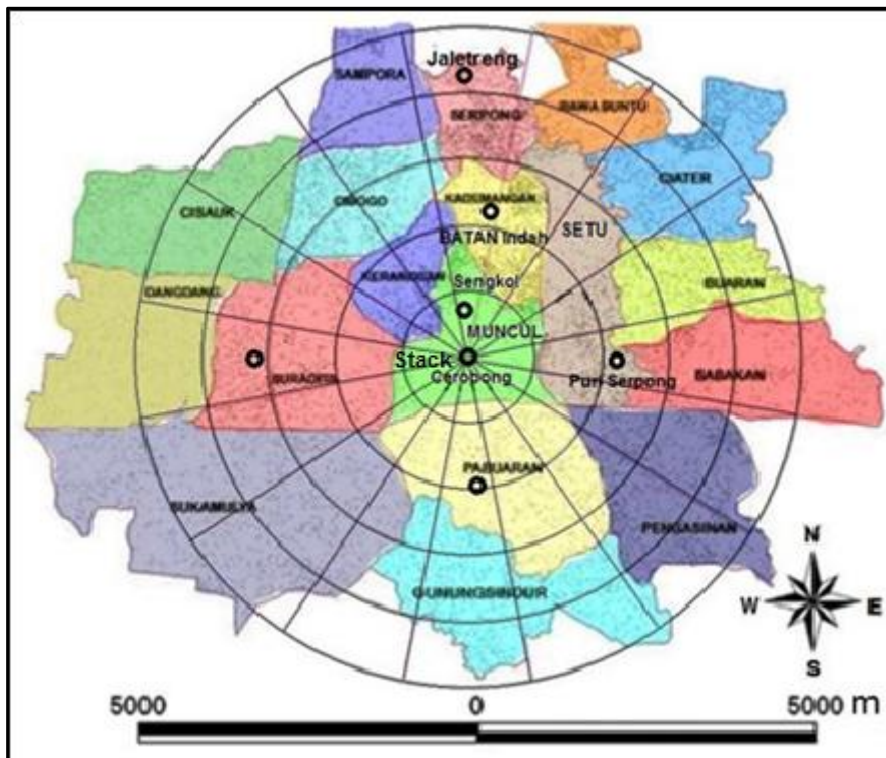


Fig.2.Locations of the research in the Serpong Nuclear Area

3. RESULTS AND DISCUSSION

3.1. Activity concentration of I-131 in the stack

Activity concentration of I-131 that the highest from the results of measurement directly in the stack obtainable is 498.35 Bq/m^3 on 19:37, 11 and 12 December 2013 (Figure 3). The high concentration of total activity on 19:37, because a process of phase change on hour 19:37 is from solution phase to gas phase during the dissolving process of the Mo-99 separation into the I-131 gas. There is I-131 gas that escape through the sidelines of the rubber connector into the stack during the gas phase, so that the concentration of I-131 activity was high momentarily on 19:37. Measurement of I-131 from the direct method is every 10 minutes. Overall of I-131 activity concentration average was 321.16 Bq/m^3 . I-131 activity concentration of direct measurement results the highest on 12:29 is 330.13 Bq/m^3 on 13 March 2014 (Figure 4). Activity concentration of I-131 average on 13 March 2014 was 162.46 Bq/m^3 .

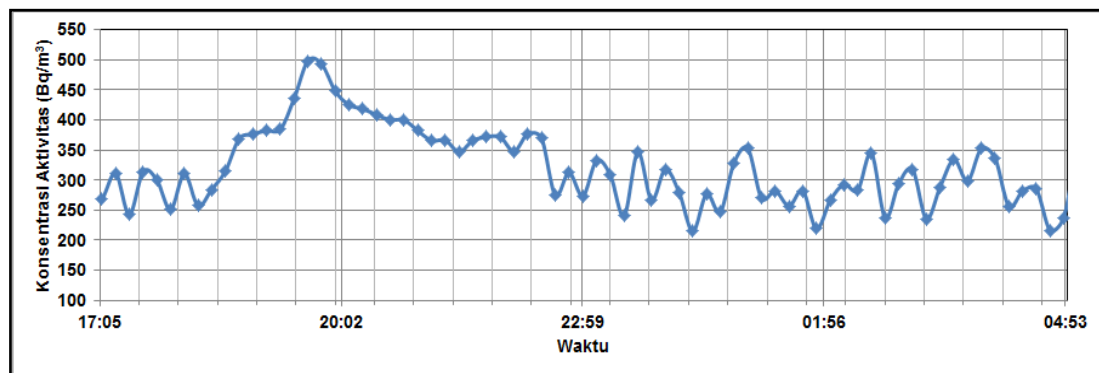


Fig.3.Measurement of I-131 activity concentrations in the stack on 11 and 12 December 2013

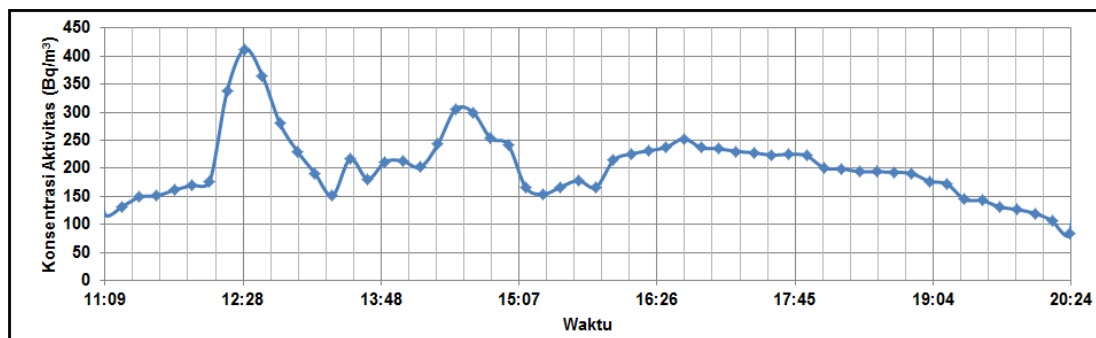


Fig.4.Measurement of I-131 activity concentrations in the stack on 13 March 2014

Overall measurement results of I-131 activity concentration in the stack is shown in Table 1. The concentration of I-131 activity the highest average on 11-12 December 2013 is 321.16 Bq/m³ and the lowest on 19 February 2014. Overall measurement results of the activity concentration of I-131 average in the stack is still below the quality standard of I-131 radioactivity levels in the air is 530 Bq/m³ based on regulation of PERKA BAPETEN No. 7/2013 [8].

3.2 Activity concentrations of I-131 in the soil and grass

Measurement results of activity concentration of I-131 in soil and grass in Puri Serpong, BATAN Indah, Sengkol, Pabuaran, Suradita and Jaletreng are shown from Figure 5 to Figure 10 respectively. Activity concentrations of I-131 adsorbed in the soil and the grass is getting high with rising humidity on the night. Humidity on the night is more than 75% starting on 18:00 to 06:00. The concentrations of I-131 activity decreased on certain hours. This case is most likely due to the fairly strong wind with a speed about above 1 m/s at these hours. A decrease in the activity concentration of I-131 was only briefly, after that the activity concentration of I-131 increased dramatically to reach the highest peak. By the time the sun began to rise starting around 06.00 am, activity concentration of I-131 in the soil and the grass began to decline. Possibly because of the I-131 nature of volatile and the solar nature of photolysis that break down the chemical reactions in the air.

Table 1. Summary of measurement results of the I-131 activity concentration in stack

No.	Date	Concentration (Bq/m ³) and Time in Stack		
	Production Time of I-131	Minimum	Maximum	Average
1	11-12 Dec 2013	214.97	498.39	321.16
	17.00-05.00	24.20	19.37	17.00-05.00
2	18 Dec 2013	160.12	400.84	257.73
	11.30-21.40	11.30	12.59	11.30-21.40
3	22-23 Jan 2014	51.30	1,113.75	192.29
	14.00-01.30	1.33	14.37	14.00-01.33
4	5 Feb 2014	100.46	247.82	155.29
	12.30-21.30	21.33	15.50	12.28-21.35
5	19 Feb 2014	37.88	228.10	132.38
	11.00-21.00	20.08	14.34	11.02-20.08
6	13 Mar 2014	67.26	330.13	162.46
	11.00-21.00	20.23	12.29	11.09-20.23

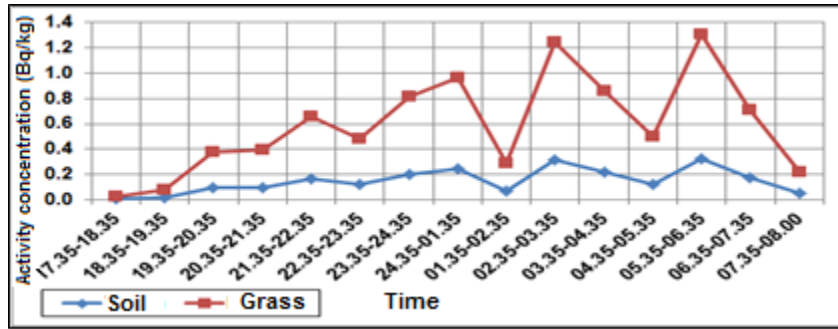


Fig.5. Activity concentrations of I-131 in the soil and grass, Puri Serpong, on 11 and 12 Dec 2013

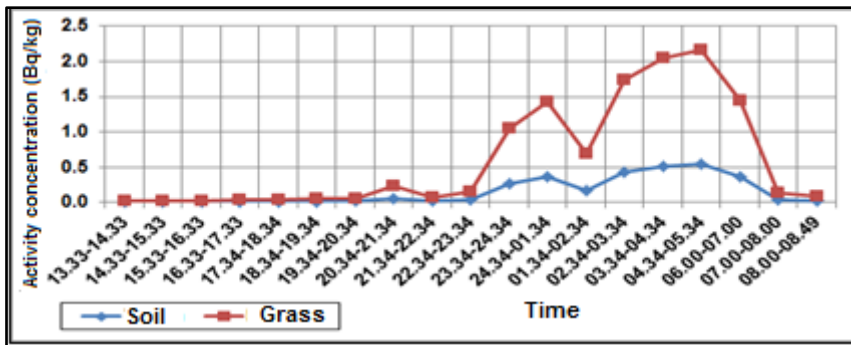


Fig.6. Activity concentrations of I-131 in the soil and grass, BATAN Indah, on 18 and 19 Dec 2013

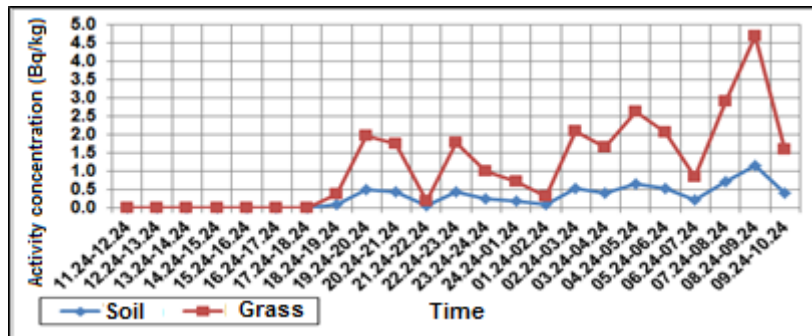


Fig.7. Activity concentrations of I-131 in the soil and grass, Sengkol, on 22 and 23 January 2014

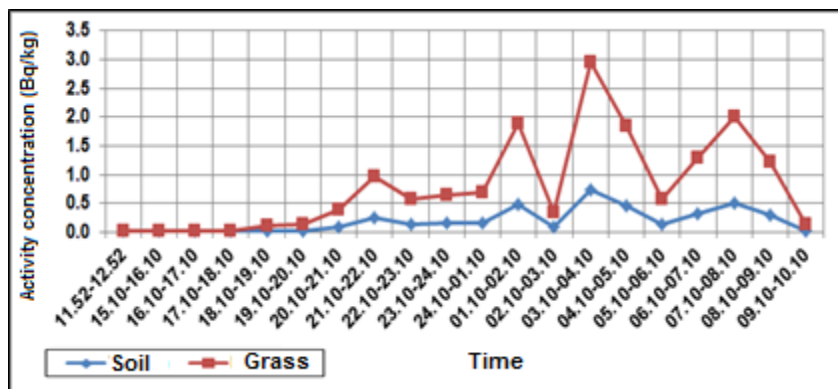


Fig.8. Activity concentrations of I-131 in the soil and grass, Pabuaran, on 5 and 6 February 2014

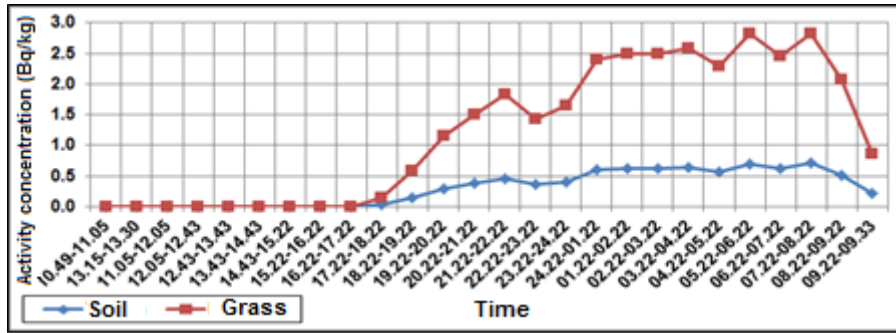


Fig.9. Activity concentrations of I-131 in the soil and grass, Suradita, on 19 and 20 February 2014

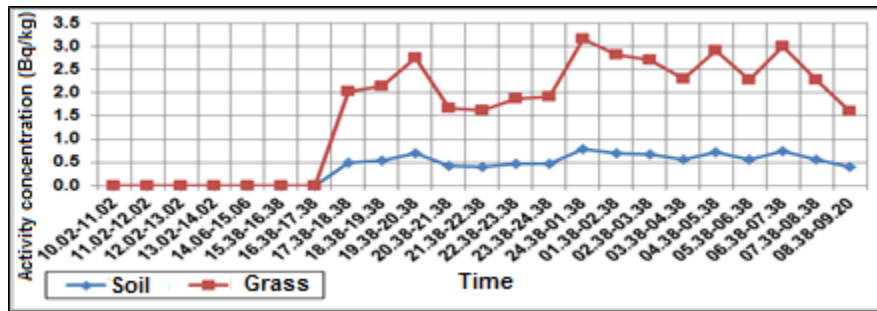


Fig.10. Activity concentrations of I-131 in the soil and grass, Jeletreng, on 13 and 14 March 2014

Summary of the activity concentrations of I-131 on average at all of research sites both on the ground and grass are shown in Table 2. The concentrations of I-131 activity on average in the grass ($\pm 75\%$) greater than the concentrations of I-131 in the soil ($\pm 25\%$). Concentrations of I-131 activity average in the grass and in soil the highest of the six research sites is in Jeletreng and the lowest is in Puri Serpong. This case is because the wind direction more to the north is to the Jeletreng direction than in to the other direction.

Interesting phenomenon occurs, the I-131 activity concentration is high, although when it rains. Rainy conditions at the time of the research are shown in Figure 11 to Figure 16. This case shows that the rain did not affect the dispersion of I-131 from the stack to the soil and the grass. So even if the rain persists deposition of I-131 radioactivity into the environment. Several researchers have carried on studies that the rain had no significant effect on the movement of gross beta radionuclide and Iodine radionuclide [10, 11, 12]. According to Baklanov, et al. (2001) and Widner, et al. (1999) that the dispersion of I-131 radionuclides as radionuclides dust fallout can be dispersed into the atmosphere by wind. Transport of I-131 concentrations in the air is influenced by the distance of dispersion location, dilution during dispersion in the atmosphere, processes of wet and dry deposition, the chemical form of the I-131 (elemental, particulate or gas) as well as the diameter of radionuclides. Transport processes of radionuclides in rainy conditions experiences wet deposition. Wet deposition includes rain, high humidity above 75%, and overcast no sunlight. Transport in wet deposition influenced washout coefficient, the radius of the particles and the rate of rain. According to Baklanov, et al. (2001), I-131 have a median aerodynamic diameter (AMAD) of 0.48 μm , dry deposition velocity of 0.6 cm/s and geometric deviation standard of 3-4 μm . Thus the radius (r) I-131 gas is 0.24 μm . Therefore AMAD from I-131 below 1 μm , then the factors of wet deposition such as the washout coefficient (Aw) and rain rate (q) has no effect, when seen from Figure 17. Moreover, I-131 emits gamma rays that can penetrate all the material in its path, including rain water, except a thick steel or concrete that can not be penetrated by gamma. Thus despite the rainy conditions, deposition of radioactive I-131 is still happening to the environment.

Table 2. Summary of measurement results of the I-131 activity concentration in soil and grass

No.	Location Direction- Distance of Stack	Date Time of I-131 production	Concentration (Bq/kg) and Time in grass			Concentration (Bq/kg) and Time in Soil		
			Minimum	Maximum	Average	Minimum	Maximum	Average
			1	Puri Serpong E, 2,2 km	11-12 Dec 2013 17.00-05.00	0.03 17.35-18.35	1.31 05.35-06.35	0.60 17.35-08.00
2	BATAN Indah N, 2,6 km	18-19 Dec 2013 11.30-21.40	0.01 13.33-14.33	2.16 04.34-05.34	0.60 13.33-08.49	0 13.33-14.33	0.54 04.34-05.34	0.15 13.33-08.49
3	Sengkol N, 0,8 km	22-23 Jan 2014 14.00-01.30	0 11.24-18.24	2.64 04.24-05.24	0.87 11.24-07.24	0 11.24-18.24	0.66 04.24-05.24	0.22 11.24-07.24
4	Pabuaran S, 1,9 km	5-6 Feb 2014 12.30-21.30	0.01 11.52-18.10	2.95 03.10-04.10	0.79 11.52-10.10	0.01 11.52-18.10	0.74 03.10-04.10	0.20 11.52-10.10
5	Suradita W, 3,2 km	19-20 Feb 2014 11.00-21.00	0 10.49-17.22	2.83 07.22-08.22	1.21 10.49-09.33	0 10.49-17.22	0.71 07.22-08.22	0.30 10.49-09.33
6	Jaletreng N, 4,2 km	13-14 Mar 2014 11.00-21.00	0.01 10.02-17.38	3.18 24.38-01.38	1.62 10.02-09.20	0.01 10.02-17.38	0.80 24.38-01.38	0.41 10.02-09.20

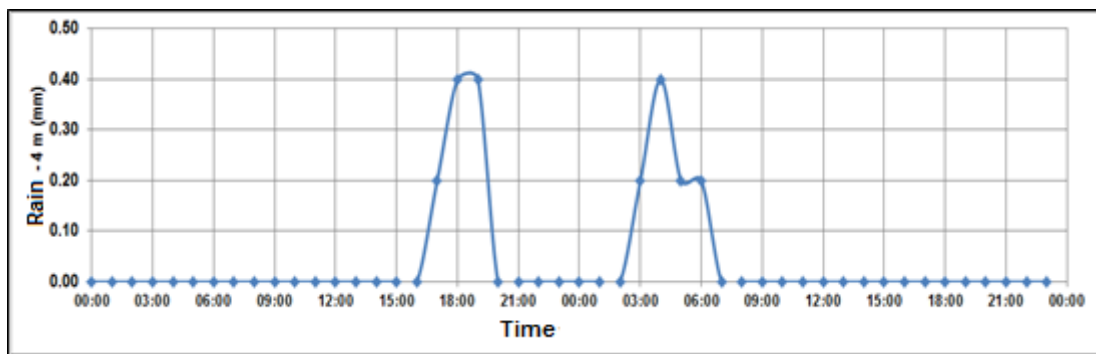


Fig. 11. RainatPuri Serpong on 11 and 12 December 2013

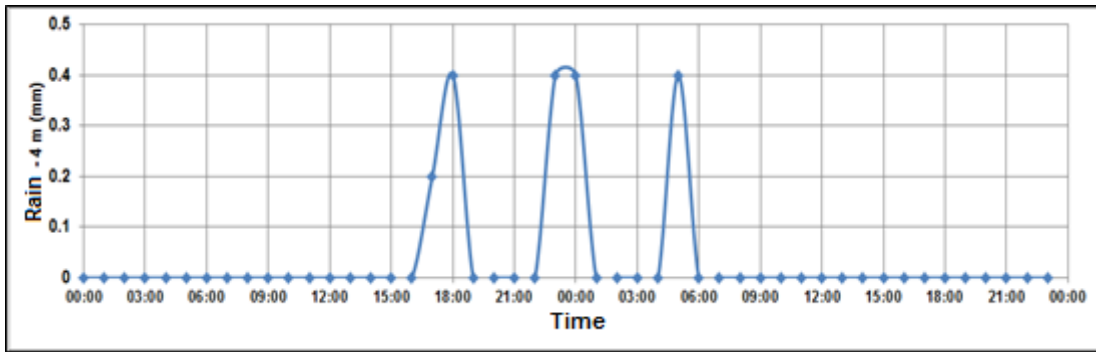


Fig.12. Rainat BATAN Indah on 18 and 19 December 2013

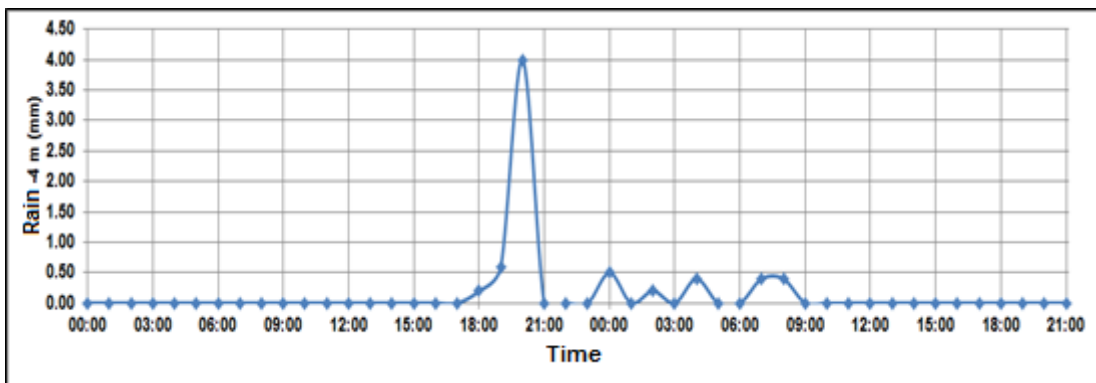


Fig.13. RainatSengkolon on 22 and 23 January 2014

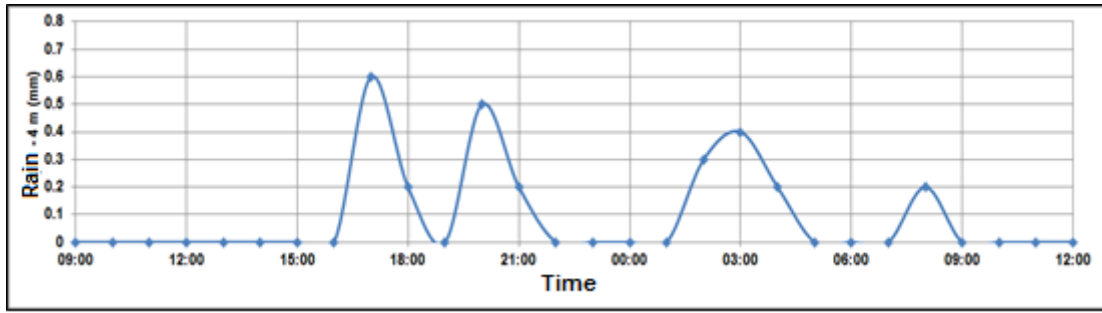


Fig.14. RainatPabuaranon5 and 6 February 2014

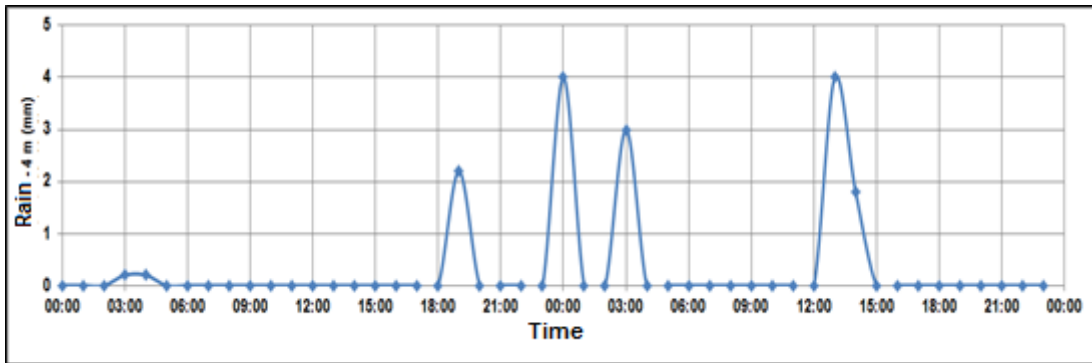


Fig.15. RainatSuraditaon19 and 20 February 2014

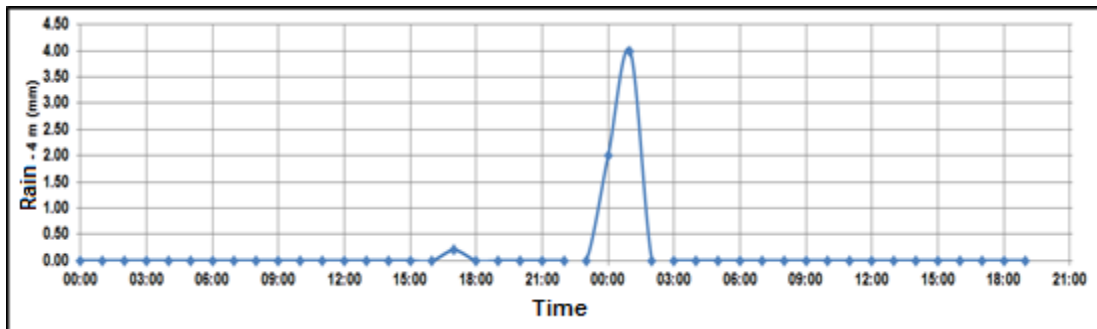


Fig. 16. Rain di Jaletrengon13 and 14 March 2014

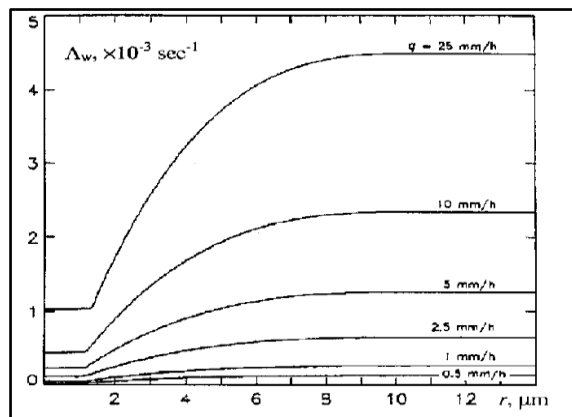
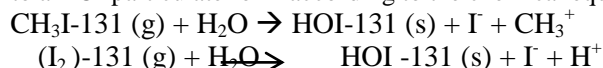


Fig.17. Wet depositionofrainis affected by A_w , r and q [7].

Natural phenomenon of the sun, moisture and rain to the activity concentration of I-131:

1. During this time there is a perception that every rain, clean air of radioactive pollutants. The results showed the rain did not affect the presence of radioactive I-131 in the air, so even if the rain persists deposition of radiation into the environment.
2. At night (high air humidity more than 80%) of I-131 concentration is higher than during the day.
3. By the time the sun began to rise (\pm between 6.00 and 7.00 hours), the concentration of I-131 tends to decrease. This case is due to the nature of the I-131 which is volatile, when exposed to sunlight. The effect of sunlight that is photolysis can break the concentration of Iodine-131 in the air. Thus the concentration of I-131 tends to decrease with the sun.

Activity concentration of I-131 is biggest in the form of CH_3I gas and I_2 gas. CH_3I gas when humidity is high (more than 80%) at night or rain changes into a HOI particulate form according to the chemical equation [13,14]:



I-131 occurs deposition into the environment easily in the form of HOI particulate at high humidity and rain, I-131 activity concentration remains high.

4. CONCLUSION

Concentration of I-131 activity in the environment (soil and grass) around the Serpong Nuclear Area(SNA) is still below the I-131 quality standard in the environment (63 Bq/kg). Concentrations of I-131 activity average at the grass (\pm 75%) greater than the concentrations of I-131 activity in the soil (\pm 25%). Concentration of I-131 activity average in the grass and in the soil highest of the seven research sites are in Jaletreng (4,200 m at the North of stack (360°)) by 1.62 and 0.41 Bq/kg respectively. Concentration of I-131 activity is high during high humidity (> 80%) at night or rain, but when the sun rises the opposite effect.

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