Preliminary Estimation of atmospheric Releases of $^{131}$I and $^{137}$Cs due to the Fukushima Daiichi nuclear power plant accident (II)

- Reanalysis of release rates from March 12 to 15 -

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1. Introduction

In the previous estimation report on May 12, the estimation of release rate from March 12 to 14 was neglected due to the lack of environmental monitoring data. However, the data during this period opened from the Nuclear and Industrial Safety Agency, the information of events occurred in the reactors reported from Japanese government and analysis of atmospheric dispersion considering short-lived radionuclides by JAEA made the reanalysis of release rates (Bq/h) of $^{131}$I and $^{137}$Cs during the objective period possible.

2. Estimation of release rates

For the estimation of release rates from environmental monitoring data, the three methods below are available and the priority to use is higher in the order of (1) to (3).

(1) Comparison of air concentrations of nuclides by dust sampling with predicted ones calculated by assuming unit release rate (1 Bq/h).

(2) Comparison of air dose rates measured in the environment with predicted ones based on unit release rate. The release rates due to grand-shine from measurements and calculations are compared to eliminate the effect of noble gases. In this method, the composition of Te, I, Cs must be assumed.

(3) Comparisons of air dose rates measured at the boundary of site with the Gaussian plume model. The demerit of this method is that small error of wind direction affects calculated air dose rate values because the plume is very narrow near the release point and it is difficult to exclude the effect of noble gases. The composition of Te, I, Cs must be also assumed.

In this report, method (2) was applied to the releases due to a hydrogen blast occurred at the Unit 1 on March 12 and after the explosion sound around the suppression chamber of Unit 2 on March 15. Method (3) was to one case and others were by method (1).

2.1 Calculation condition

Figure 1 shows the monitoring points which provided data for the estimation of release rates. WSPEEDI was used for calculating air concentrations and air dose rates under the assumption of unit release rates. Calculation conditions are as follows:

Meteorological data: Analysis data GPV (MSM) from Japan Meteorological Agency (every 3 hrs with horizontal resolution of 0.0625 × 0.05 deg) or GSM Japanese area data (0.25 deg. × 0.2 deg.).
Observation data (AMeDAS, measurement data at the boundary of the Fukushima Daiichi NPP and tower data from the Fukushima Daini NPP) were used for data assimilation.

Computational area and grid: Three domain nesting method was used. The calculation results in the inside domain including around Fukushima Pref., a part of Tochigi and Ibaraki Pref. (190 × 190 × 10 km, 1 km resolution) was used for the estimation of release rates.

Release height: The heights of 20 m, 120 m and 20 m were applied for the releases due to the damage of pressure container vessel (PCV), wet-well ventilation, and after the explosion sound around the suppression chamber of Unit 2. For the hydrogen blast at Units 1 and 3, volume source with 100 m × 100 m horizontally and 100 m and 300 m vertically were applied.

Release duration: continuous release for the damage of PCV, wet-well ventilation and 30 min. releases for hydrogen blast. The release duration after the explosion sound around the suppression chamber of Unit 2 was determined to represent the environmental monitoring data well.

Released nuclides: $^{131}$I, $^{132}$Te+$^{132}$I (assumed radioactive equilibrium), $^{137}$Cs, and $^{134}$Cs.

Major radionuclides detected in the environment were I, Te and Cs and their composition ratio varied widely. But the ratio of $^{132}$Te with $^{131}$I was higher than 1 in the early stage and almost 1 around March 15. The ratio of $^{137}$Cs with $^{131}$I also changed time to time, but averagely 0.1.

2.2 Estimated result

The temporal variation of release rates from March 12 to 15 is shown in Fig. 2. The figure indicates that relatively large amounts of release occurred, in time series, (1) around the hydrogen blast of Unit 1 at 15:36 March 12, (2) around the decrease of pressure in Unit 2 at 21:03 March 14 and (3) after the explosion sound around the suppression chamber of Unit 2. The release rates due to the hydrogen blast at Unit 3 could not be estimated because the radioactive plume flowed toward the Pacific Ocean. Thus, the release rate is assumed to be the same as the case of Unit 1 hydrogen blast. The release duration for each release rate was determined from the temporal variation of air dose rates at the boundary of the site, but 30 min. was assumed for hydrogen blasts for Units 1 and 3. Although it is not clear that the release was continuous, the same level of increases of air dose rates at the boundary sometimes occurred in the night of March 12 and early morning of March 14 when no events in the reactors related to the atmospheric releases were reported. It indicates that the atmospheric release continued from the first release in the morning of March 12.

To compare with the previous estimation of release rates, the previous values are shown by solid lines in Fig. 2. The compared results show that the release rates due to the PCV damage of Unit 1 and wet-well ventilations of Units 1 and 3 until March 14 were the same order as previous values and the increases of release rates due to hydrogen blasts were added. Concerning the release on March 15, the release rates of $^{131}$I and $^{137}$Cs decreased because present estimation considered the effects of short-lived nuclides.

(1) Release rate estimated from the air dose rates measured on March 13
The event which increased the air dose rates was investigated from the temporal variation of air dose rates at Minamisoma in high dose area of March 13 (Fig.1 (2)). The variation shows that the air dose rates rose to about 20 μSv/h at 20:00 March 12 and, then, the values of about 5 μSv/h continued due to probably radionuclides deposit on the ground. The nearest event before 20:00 is the hydrogen blast of Unit 1 (15:36) and it is reasonable that the plume released at the time of this event reached to Minamisoma with several hour delay, considering the wind fields. Thus, under the assumption that the air dose rates measured on March 13 was generated by radionuclides on the ground discharged by the hydrogen blast of Unit 1, the comparison between measurement and calculation to estimate release rates was carried out. The compared result is shown in Fig. 3. From the fact that the air dose rate at Iitate-mura was almost back ground level, about 5 μSv/h at Minamisoma and over 30 μSv/h between Iitate-mura and Minamisoma at that time, the radioactive plume discharged by the hydrogen blast of Unit 1 passed between Iitate-mura and Minamisoma. The calculation result represents this situation. Here, the contour line of $1 \times 10^{-14} \mu$Sv/h from WSPEEDI is assumed to be correspond with 30 μSv/h of measurements and the release rate of $^{131}$I was determine to be $3 \times 10^{15}$ Bq/h. However, as shown in Fig. 1 (2), some amounts of radionuclides in air were detected in the noon of March 13 and they might affect the air dose rate values and resulted in the small overestimation of release rates.

(2) Release rate estimated from the air dose rates measured on March 15

In Fukushima Pref. except near seacoast, the air dose rates rose after the explosion sound around the suppression chamber of Unit 2. In previous report, the release rates after this event was estimated about $10^{16}$ Bq/h of $^{131}$I during the period from 9:00 to 15:00. Here, the releases in first three hours in the morning were tentatively determined based on the time when the air dose rates at the boundary of site rose up. Present estimation improved the following points:

① Because the increase of air dose rates due to the release in the morning was found at Koriyama, the estimation of release rate in the morning was performed by extending computational area.

② $^{132}$Te and its progeny $^{132}$I were considered for the calculation of air dose rates.

③ Meteorological observation at Fushima Daiihi and Daini NPPs were considered to increase the accuracy of meteorological predictions.

④ Temporal variations of air dose rates measured several points in Fukushima Pref. were used for the estimation.


According to the estimation on March 15, the release rate of $2\times10^{15}$ Bq/h of $^{131}$I continued from about 7:00 to 10:00. The release decreased once a while and, then, the release of $4\times10^{15}$ Bq/h of $^{131}$I continued from about 13:00 to 17:00. The temporal variations of air dose rates at the main gate of Fukushima Daiichi NPP and the pressure level of Unit 2 also imply this release variation.

The reason why the release rates of $^{131}$I decreased to 1/2 – 1/5 compared with the previous
report is that the temporal value in the morning was re-estimated by acquiring new monitoring data and the air dose rate due to $^{131}$I, $^{137}$Cs and $^{134}$Cs decreased by deducing air dose rate due to short-lived nuclides from total one. However, considering the uncertainty of this release estimation method (at least factor 5), the discrepancy may be included in error bar.

Figure 4 shows the comparison of air dose rates at several monitoring points between measurements and calculations using estimated release rates. Because measurements and calculations generally agreed well, the estimated release rates are probably reasonable.
Fig. 1 (1) Environmental monitoring points used for the estimation of release rates on March 12 (Concentrations of $^{131}$I by dust sampling are only shown for reference.)

Fig. 1 (2) Environmental monitoring points used for the estimation of release rates on March 13 (Concentrations of $^{131}$I by dust sampling are only shown for reference. Values in the map indicate air dose rate.)
Fig. 1(3) Environmental monitoring points used for the estimation of release rates on March 14 (Concentration of $^{131}$I by dust sampling is only shown for reference.)

Tokai-mura (4:25-4:45 on March 15)
Concentration of $^{131}$I: 1260 Bq m$^{-3}$

Fig. 1(4) Environmental monitoring points on March 15 used for the estimation of release rates (continuous measurements of air dose rate were compared with calculations).
Fig. 2 Estimated temporal variation of release rates of $^{131}$I and $^{137}$Cs from March 12 to 15. Lateral bars on plots show estimated release durations. Lines indicate the preliminary estimation reported by NSC.
Fig. 3 Comparisons of air dose rate between measurements (plots) by car monitoring and calculations (contours) assumed with unit release rate (1Bq h\(^{-1}\)) at 12 JST on March 13.
Fig. 4 Comparisons of temporal variations in air dose rate between measurements and calculations at various monitoring posts from March 12 to 15 (plots: monitoring values, lines: calculations)

*The comparisons for Onagawa and Minamisoma were made using calculations at the locations to 10km north-northwest and 7 km west from actual monitoring points, respectively, because the calculated plume that corresponds to H₂ explosion at Unit 1 seems to have passed easterly and northerly courses than reality, respectively, based on other monitoring data.