Cesium 137 Pollution from Fukushima Nuclear Power Plant

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ABSTRACT

The great earthquake occurred and gigantic tsunami waves damaged Fukushima Nuclear Power Plants. Emergency core cooling systems did not work, three reactors in Dai-ichi nuclear power plant became meltdowns and hydrogen exploded to break the buildings. Finally a big amount of radioisotopes were emitted to the sky. On March 21, it rained in the Kanto Plain and high concentration of radioactive iodine 131 pollution was detected on March 22 and 23 at filtration plants in waterworks in the Tone River watershed. This paper estimated spatial distribution of radioisotopes from nuclear reactors including hydrogen explosions in the Tone River watershed. Some theoretical relationships of radioisotopes were presented for evaluating radioactivity at distant areas. At present, most effective radioisotope is cesium 137, which shows two different pollution aspects by land use. One is the secondary pollution from forest and soils, and started to distribute isotopes with leaves and soils through paths by wind and the public transportation. In many areas of Japan such contamination was observed. Another is accumulating isotopes in urban garbage processing plants and sewerage treatment plants, and approaches the upper limit for their capacity. This becomes a very serious problem.

Keywords*: ALOS, meteorology, runoff routing, stokes equation, THEOS*

I. Introduction

At 14:46 on March 11, 2011, the great earthquake with magnitude of 9.0 occurred from a focus of Sanriku-oki. From Hokkaido to Kyushu, all Japan was attacked with a big shake and Tsunami. In record, it was the maximum in Japan and the greatest of the world with 20335 of victims. At 15:50, the great tsunami attacked Fukushima Daiichi and Daini Nuclear Power Plants, Tokyo Electric Power Company. The emergent core cooling system did not work and lost cooling ability. Fukushima Daiichi had meltdown at Reactor 1, and a hydrogen explosion occurred, which made building structures flying to pieces and a big amount of isotopes leaking into the atmosphere. Reactors 2 and 3 also had meltdown in the same way. Reactor 4 was fired. At 13:10 on March 20, Reactor 3 had meltdown and isotopes leaked into the atmosphere again. On March 21, it rained in the Kanto plain, and high concentration of iodine 131 was detected from drinking water in waterworks in the Tone river watershed on March 22 and 23.

Here, time series of radioisotopes were estimated with hydrogen explosions at the nuclear power plants, and time series of pollutants falling in the atmosphere were analyzed for the spatial distribution of radioisotopes in the watershed. Next, from the meteorological data with rain on March 21, rainfall budget was analyzed for iodine concentration at the purification plant in the Tone river watershed.

At present, most effective radioisotope is cesium 137, which shows two different pollution aspects by land use: the secondary pollution from forest and soils and accumulating isotopes in urban garbage processing plants and sewerage treatment plants.

II. Method

A. Used data

Satellite data were THEOS/ Panchromatic/ Multispectral on March 18 and 28, 2011 and ALOS/PRISM/AVNIR-2 on March 28, 2011 and May 20, 2009. Meteorological data were 10 min rainfalls and winds from AMeDAS (Japan Meteorological Agency, 2011). Others were used from newspapers and internet references.

B. Rainfall analysis

From wind directions, flow directions of radioisotopes were determined each hydrogen explosion. The most wind directions were north and south. It was no rain in Kanto area till March 21, when the wind direction was north. Therefore, most of isotopes were emitted into the atmosphere during these days and the ones in the watershed ran off into the Tone River with rainfall.

C. Pollution model

From the Stokes equation, relationship between particle falling velocity and its size was obtained for

isotopes.

$$v = D^2\rho g/18\mu \quad (1)$$

where v: falling velocity, D: particle size, ρ: specific weight, g: gravity constant, and μ: viscosity coefficient.

The specific weight of isotopes varies widely from gas to solid state. As the falling time is reverse proportional to the specific weight, direct contaminants should be isotopes in solid state and ones adhering solid such as sand. This hypothesis leads to the next equation from Equation (1), as shown in Figure 3.

$$L = v_w t = 18\mu h v_w / D^2 \rho g \quad (2)$$

where L: approach distance, v_w: wind velocity, t: floating time, and h: the maximum height. If radioactivity is proportional to the mass of the particle, the next equations were derived from Equation (2).

$$I = 9*2^{1/2} N_a (1-2^{-1/T})(\mu h v_w)^{3/2} \alpha / (\rho^{1/2} g^{3/2} w_a L^{3/2}) \quad (3)$$

$$I \propto L^{-3/2} \quad (4)$$

where I: radioactivity, N_a: Avogadro number, T: a half-life, w_a: an atomic weight, α: the weight ratio of an isotope to the carrier (=1 if the carrier does not exist).

D. Hydrogen explosions

Hydrogen exploded in the reinforced steel concrete buildings, and radioisotopes such as iodine 131 were emitted into the atmosphere. If the initial velocity at explosion is 340 m/s, the next equation holds.

$$\rho_a \pi D^2 v_a^3 t/4 = \rho \pi D^3 v_0^2/6 \quad (5)$$

where ρ_a: air density (1.29 kg/m^3), v_a: velocity at the explosion (340 m/s), t: the mean flying time of particles in the building, v_0: velocity at the exit of the building.

The building volume is 75000 m^3, and the mean distance between the center and the wall in the building is 56.1 m (TEPCO, 2011). thus,

$$t = 56.1 / 340 = 0.165 \text{ s}$$

Therefore, the velocity at the exit of the building for particles is the next equation.

$$v_0 = (3\rho_a v_a^3 t / 2\rho D)^{1/2} = 68.8\, D^{-1/2} \quad (6)$$

With air resistance $\rho_a \pi D^2 v^2 c / 8$ and the gravity, the approach altitude will be the next equation as the first approximation by integration.

$$H \sim \beta v_0 \sim D^{-1/2} \quad (7)$$

where c: resistance coefficient (0.2), β: a coefficient(>1).

Figure1 Satellite images of Fukushima nuclear power plant before (Left: ALOS, May 20, 2009) and after (Right: THEOS, March 18, 2011) the earthquake (RESTEC). Numbers are reactors.

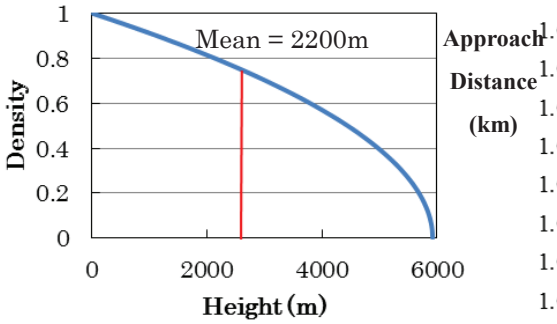

Figure 2 Particle vertical distribution by hydrogen explosion.

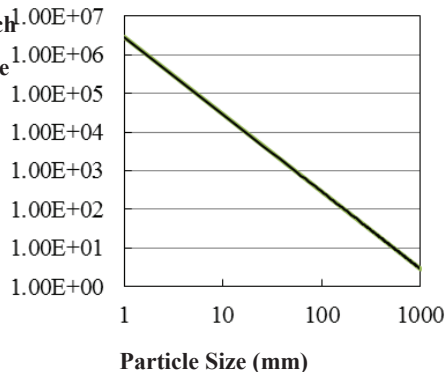

Figure 3 Relationship between particle size (mm) and approach distance (km). Both are logarithmic normal distributions.

III. Results

A. Particle size and its approach distance

Particles flying into the air by a hydrogen explosion distributed isotropically as shown in Figure 2, which was estimated by a parabolic distribution. On March 14, fine particles floating in the air approached in two hours to Mount Ryo in the north, and on March 15 in 2 to 6 hours to Kanto area. In sunny days, fine particles less than 0.1 mm were floating and coarse particles more than 0.1 mm had fallen on the earth. In rainy days, fine particles less than 0.1 mm had fallen with rain drops. However, during hydrogen explosions, it did not rain. The distribution of particle sizes can be estimated from the falling speed. From the hydrogen explosion the first contamination was detected after 3 hours at the front gate of the nuclear plant, which means that falling contaminants are mainly between 0.5 and 2.5 mm size particles. As shown in Figure 4, this distribution was a logarithmic normal distribution. This particle should be estimated as concrete debris and/or sand particles.

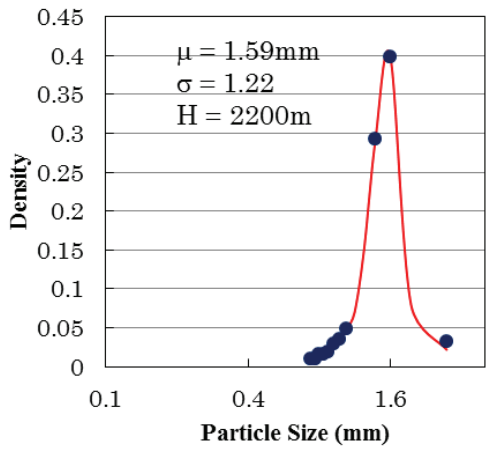

Figure 4 Particle size distribution estimated from dose observation and Equation (1).

It shows a logarithmic normal distribution with 1.6 mm of the mean size. Radioisotopes would move in the atmosphere by these particles, sand and concrete debris.

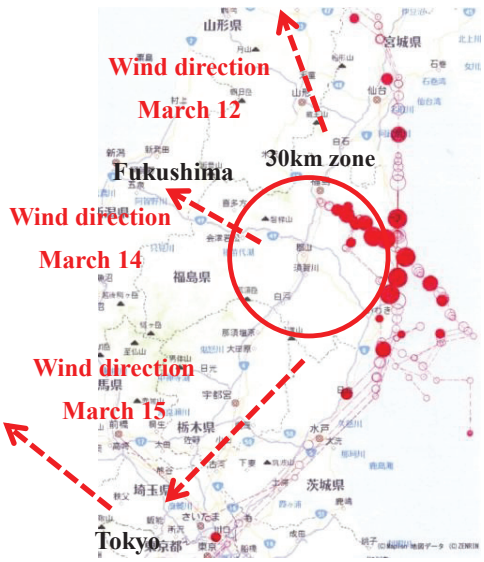

Figure 5 Radioisotope pollution shows a logarithmic normal distribution through the wind directions.

Isolated pollution areas were made through the downwind direction.

Table 1 Time series of hydrogen explosions at Fukushima Daiichi Nuclear Power Plant.

Date/Time	Reactor	Max Dose	Time Lag*	Dose Estimate**	Wind Direction
3/12; 15: 36	1	1204 μ Sv/h	16h57m	$4.3x10^4$TBq	South
3/14; 11: 00	3	3130 μ Sv/h	11h37m	$2.3x10^6$TBq	Southeast
3/15; 6: 14	2	11992 μ Sv/h	2h46m	$4.3x10^5$TBq	North
3/15; 20: 00	4	8124 μ Sv/h	?	$2.9x10^5$TBq	South
3/16; 5: 45	3	10850 μ Sv/h	6h45m	$3.9x10^5$TBq	East Northeast

*Time difference between explosions and observation of dose at MP4 station.

**The same estimate as Reactor 2.

B. Time series of explosions and isotope emission into the atmosphere

The first explosion occurred in Reactor 1 at 15:36 on March 12, when the south wind blew (Table 1). At 11:00 on March 14 a hydrogen explosion occurred in Reactor 3, when the south east wind blew. At 6:14 on March 15, Reactor 2 had a hydrogen explosion and next Reactor 4 fired, when the north and east wind blew, and radioisotopes flowed into the Kanto plain. At 5:45 on March 16, Reactor 3 had again a hydrogen explosion, when the east north east wind blew. Therefore, the radioisotopes from Reactors 1 and 3 by hydrogen explosions on March 12 and 14 flowed into Fukushima City and Sendai City, while the ones from Reactors 2, 3, and 4 by explosions and fire on March 15 and 16 flowed into the Pacific Ocean and the Kanto plain.

C. Estimate of radioisotope dose

From failure rate of fuel rods, about 725 tons of equivalent uranium might discharge outside of the reactors. Contaminated water became amount of 60000 tons with radioactivity of 1000 mSv at surface, which means that the concentration was estimated by 10 ppm to 0.1 %, and 0.6 to 60 tons of radioisotopes discharged outside as contaminated water.

From dose measurement at the front gate of the nuclear plant 12 mSv/h was observed on March 15, similarly four-times explosions and fire occurred. From iodine 131 equivalent conversion, this explosion emitted $4.3x10^5$ TBq of isotopes. Similarly total explosions emitted $3.4x10^6$ TBq of isotopes. On March 15, the radioisotopes emitted to the Kanto plain was estimated by $4.3x10^5$ TBq.

D. Rainfall Runoff Relationship

On March 20, it was rainy in the Kanto plain with 21.7 mm on average in the watershed. From 16840 km^2 of the watershed area, the total discharge became $3.7x10^8$ tons. If 430000 TBq of the total isotopes had fallen into the watershed, 58% of them, the concentration of iodine 131 would be $6.7x10^5$ Bq/kg. As 210 Bq/kg of iodine131 was detected at Kanamachi purification plant, which means 0.1% of the total with concerning its half-life.

IV. Discussion

A. Theoretical Approach

The major radioisotopes were 18 kinds and iodine 131 was on the middle in intensity (see Appendix). A hydrogen explosion emitted $4.3x10^5$ TBq of isotopes, which was most a radiation form. The radioisotopes flowing to Fukushima City were blocked by Abukuma highland, and were estimated to pollute this mountain at $2.28x10^6$ TBq. At that time, the government estimated the total amount of iodine 131 was 30000 to 110000 TBq, 6.5 to 24 g, which might be much underestimated. The hydrogen explosion emitted iodine 131 into the atmosphere, and most had fallen in the circle of 30 km and about 0.1% overflowed out of the circle.

By Equation (1), particle size distribution was estimated from actual radiation dose observation. As shown in Figure 3, this distribution was a logarithmic normal distribution. Radioactive particles for Fukushima distributed spatially in similar to particle size distribution.

Equations (2), (3) and (4) are available for evaluating radioactivity from wind velocity at the pollution source.

B. Radioisotope Budget

Radioisotopes from Fukushima Nuclear Power Plant were gas like materials emitted into the atmosphere by five-times hydrogen explosions, and solid and solution seepage into the ground by meltdown. By weight, the latter was dominant. By radioactivity, the former was dominant. The fuel bars were 725 tons in total. In Chernobyl, 30 % of fuel bars were emitted outside. Therefore, finally 218 tons of isotopes would be estimated as pollutant at least. Budget estimates are shown next.

Hydrogen explosion to the sky: $2.6x10^6$ TBq,
60000 tons of dirty water: 0.6 to 60 tons,
50000 tons of Ground water leakage: 0.5 to 50 tons.
In total, $2.6x10^6$ TBq of ^{131}I and 1 to 110 tons of RI were emitted outside.
In Chernobyl,
^{131}I: $1.3x10^6$ TBq (20%),
^{137}Cs: $2.9x10^5$ TBq (13%),
Total: $8.8x10^8$ TBq; 10 tons.
Therefore, the total radioisotopes at Fukushima Nuclear Power Plant are estimated 10% to 11 times as much as Chernobyl accident.

C. Chemical reaction of hydrogen explosion

Zirconium that was used for covers of fuel bars reacts with water and hydrogen occurred.

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$$

By this reaction, a big amount of hydrogen occurred in the reactors and leaked out of reactors and exploded inside the buildings. In this case, the amount of hydrogen was shown as next. Parenthesis values are total one including used fuels in the building.

Reactors 2 and 3: generated $3.1x10^5$ mols of H_2 (total: 3.3 and $2.9x10^5$)
Reactor 1: $2.3x10^5$ mols of H_2 ($1.7x10^5$)
Reactor 4: $5.5x10^5$ mols of H_2

Final partial pressure: reactor 1= 0.07 (total: 0.12), reactor 2 = 0.09 (0.19), reactor 3 = 0.09 (0.17), reactor 4 = 0.16 atm

Thus, the air pressure becomes 0.07 to 2.1 atm at 3000°C after the explosion.

Final partial pressure of hydrogen and generated water became at most 2 atm, while reinforced concrete buildings could not be broken with less than 10 to 20 atm. Therefore, the zirconium-water reaction cannot produce enough hydrogen to explode the buildings. Most hydrogen might be produced by radiation damage for water or high temperature contact with fuel bars.

D. Radioisotope carrier

SPEEDI calculates diffusion simulation from reactors to the atmosphere each isotope. However, actual isotope performances are categorized to two patterns. Most isotopes adhered sands and concrete debris, and diffused into the atmosphere, which explained the observation of isotope doses very well. By the observation of radioactive doses, after the hydrogen explosion in a few hours a single or some peaks were shown, which means isotopes fell down. Even most isotopes were 18 kinds (Appendix), maybe more than 200 kinds of isotopes leaked outside and diffused. As a carrier of sands or concrete debris, diffusion with a limited particle size distribution explains the results of observation. Sands in the atmosphere and concrete debris generated by hydrogen explosions diffused in the sky, and leaked isotopes caught them to fly into the sky and fell down from a certain elevation each particle size according to the wind velocity and directions. Therefore, the spatial distribution of the pollution was similar to the particle size distribution for east Japan. On the other hand, fine particles or gas like materials departed from a carrier and moved with the wind, and fell down to the earth by rainfall. Thus, these two ways were the main form of isotopes movement in the sky. All the isotopes with their carriers fell down to east Japan and the Pacific Ocean.

E. Present state: Cesium 137 performance

At present, most effective radioisotope is cesium 137, which shows two different pollution aspects by land use. One is the secondary pollution from forest and soils, and started to distribute isotopes with leaves and soils through paths by wind and public transportation. In many areas of Japan such contamination was observed. Another is accumulating isotopes in urban garbage processing plants and sewerage treatment plants, and approaches the upper limit for their capacity. This becomes a very serious problem.

V. Conclusions

(1) Iodine 131 contaminated into waterworks networks should be 0.1 % of the total radioisotopes emitted outside in the sky from the nuclear power plant, which might occur mainly by hydrogen explosions. 70 % of radioisotopes that occurred by five-times hydrogen explosions and fires distributed in Fukushima.

(2) A hydrogen explosion emitted $7x10^5$ TBq of radioisotopes on average, in total $3.4x10^6$ TBq. Half of them were iodine 131 with $2x10^6$ TBq. In Chernobyl it was estimated as $3x10^5$ TBq. The government stated it was 3 to $11x10^5$ TBq, which should be greatly underestimated. Probably it will be ten times as much as Chernobyl finally.

(3) The particle size distribution that contributed to directly contamination was a logarithmic normal distribution of concrete debris and sands with the mean particle size of 1.6 mm, which were emitted into the atmosphere over 2000 m, flied for several hours, dropped onto the ground and polluted. Theoretical equations explained these phenomena very well. Fine particles less than 0.1 mm have much air resistance and a different path from these

particles.

(4) Fuel rods that related with hydrogen explosions were estimated as 725 tons in total, 30 % of which might be emitted outside like Chernobyl; therefore, 218 tons should be the most probable amount.

(5) At present, most effective radioisotope is cesium 137, which shows two different pollution aspects by land use. One is the secondary pollution from forest and soils, and started to distribute isotopes with leaves and soils through paths by wind and the public transportation. Another is accumulating isotopes in urban garbage processing plants and sewerage treatment plants, and approaches the upper limit for their capacity.

ACKNOWLEDGEMENT

I greatly appreciate for cooperation of Dr. Yoyo Hinuma and Mr. Satoshi Aruga.

REFERENCES

Hinuma Y. (2011). *Radioactive pollution effect by Fukushima Nuclear Power Plant*.

Japan Meteorological Agency (2011). Weather forecasts and analysis: http://www.jma.go.jp/jma/en/menu.html

Koide H. (2011). *Beyond reality of radioactive pollution*, Kawade-shobo.

Nuclear and industry safety agency (2011): http://www.nisa.meti.go.jp/

TEPCO (2011). Fukushima Daiichi Nuclear Power Plant:http://www.tepco.co.jp/nu/f1-np/intro/outline/outline-j.html

Appendix: Main radioisotopes at Fukushima Daiichi Nuclear Power Plant.

Isotope	Half life	Radioactivity	Absorbed dose
Co56	77.27d	1116TBq/g	815390Gy/s
Co58	70.82d	1175	433757
Co60	1925.1d	4	18903
Mo99	65.94h	17760	3855090
Tc99m	6.01h	194800	4457166
Ru106	373.59d	121	761
Ag108m	418y	106	34696
Te129	69.6m	781300	187252378
Te129m	33.6d	1114	18898
Te132	3.204d	11430	901884
I131	8.0207d	4596	714108
I132	2.295h	38180	218493453
I134	52.5m	97910	653279065
Cs134	2.0648y	47	15755
Cs136	19s	2731	64647404071
Cs137	30.17y	3	601
Ba140	12.752d	2653	444532
La140	1.6781d	17250	10382169