Title	Formation mechanisms of insoluble Cs particles observed in Kanto district four days after Fukushima Daiichi NPP accident
Author(s)	Hidaka Akihide
Citation	Journal of Nuclear Science and Technology,56(9-10),p.831-841
Text Version	Accepted Manuscript
URL	https://jopss.jaea.go.jp/search/servlet/search?5063339
DOI	https://doi.org/10.1080/00223131.2019.1583611
Right	This is an Accepted Manuscript of an article published by Taylor & Francis in Journal of Nuclear Science and Technology on September 2019, available at http://www.tandfonline.com/10.1080/00223131.2019.1583611.

ARTICLE *Fukushima NPP Accident Related*

Formation Mechanisms of Insoluble Cs Particles Observed in Kanto District Four Days after Fukushima Daiichi NPP Accident

Akihide HIDAKA[†1,†2]

[1]Nuclear Human Resource Development Center, Japan Atomic Energy Agency,
2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan
[2]Department of Nuclear Engineering, Khalifa University, 127788, Abu Dhabi, UAE

(Received)

Insoluble Cs particles (so called Type A) were first observed in Tsukuba- city on the morning of March 15, 2011. The particles were considered to have been formed in the reactor pressure vessel of Unit 2 by evaporation/condensation based on the measured ^{134}Cs/^{137}Cs ratio and the accident progression of each Unit. However, the particles were covered by almost pure silicate glass and showed a trace of quenching. This can be explained by other mechanisms, that is, the particles were formed by the melting of glass fibers of the high efficiency particulate air filter in the Standby Gas Treatment System owing to the fire due to hydrogen detonation in Unit 3 at 11:01 on March 14 and atomization due to the explosion, followed by quenching of the molten materials. Although the particles formed in this way were mostly dispersed to the sea by wind at that time, some of them were deposited on the lower elevation of the reactor building of Unit 3, and they could have been re-suspended and released into the environment, by the flow owing to the generation of a large amount of steam as result of a restart of core

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[†1] Corresponding author, E-mail: hidaka.akihide@jaea.go.jp
Present address: 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan
[†2] Corresponding author, E-mail: akihide.hidaka@ku.ac.ae
Present address: PO Box 127788, Abu Dhabi, UAE

cooling water injection at 02:30 on March 15.

Keywords: Fukushima Daiichi NPP Accident, Source terms, Insoluble Cs particles, Hydrogen explosion at Unit 3, Detonation, Atomization, Resuspension, HEPA filter

1. Introduction

Cesium (Cs) released into the environment during severe accidents (SA) has been considered to have a soluble form based on the past experiences of various nuclear power plants (NPP) accidents and knowledge gained from SA studies [1]. However, the environmental measurements recorded after the Fukushima Daiichi NPP accident (called the Fukushima accident hereinafter) revealed that a part of the Cs released during the accident was in the form of insoluble Cs particles covered by silicate glass [2].

The insoluble Cs particles can be classified as Type A, commonly called “Cs ball” with spherical shape and small diameter (2 - 10 μm), and Type B with distorted shape and large diameter (70 - 400 μm). The major characteristics of Types A and B particles [3] are summarized in **Table 1**. Analyses of the ^{134}Cs/^{137}Cs ratio so far [4] have showed that Type A particles originated from Unit 2 or 3, and Type B particles originated from Unit 1. The Type A particles were found in Kanto district, including Tsukuba- city, Tokyo, and the region northwest of the Fukushima Daiichi Nuclear Power Plants (hereafter called as FDNPP), while the Type B particles were found only on north side near Unit 1. Location of Tsukuba- city, Tokyo, Tokai, and FDNPP on the Japanese map is shown in **Fig. 1**. The detailed measurements in Tsukuba-city and Tokyo showed that approximately 30% [6] and 90% [7] of the measured Cs being Type A particles, respectively.

A previous study [8] focused on the detailed analyses of the constituents of these particles and inferred their formation mechanisms based on the results. The previous study proposed that the Type A particles were formed by evaporation/condensation in the Reactor

Pressure Vessel (RPV) of Unit 2, quenching by water from the Suppression Pool (S/P), and subsequent leakage from the top flange of the containment vessel, and release into the environment at midnight on March 14 [5]. By contrast, the Type B particles were considered to have been formed during the hydrogen explosion in Unit 1 at 15:36 on March 12.

The process of formation of Type B particles is considered appropriate considering the phenomena that could occur during the hydrogen explosion and the physical properties of Type B particles. By contrast, for the Type A particles, it is difficult to explain the coverage of almost pure silicate glass owing to the evaporation/condensation process in the RPV because various types of core structures were evaporated and airborne in the RPV during SA, suggesting that the particles formed could be covered by multiple components, including the silicate glass. In addition, it is almost impossible to explain the reemission of particles into the gaseous phase after they were quenched by water from the S/P.

Because the particles covered by silicate glass could not be dissolved into water, they remained in the environment for a longer term in contrast to the soluble ones. When people breathe in these particles, the particles may remain in the human body for a longer term, but the influence of this phenomenon has not been understood completely yet.

By contrast, FDNPP decommissioning is making progress day by day. Because the possibility that some of the Type A particles remain somewhere in the FDNPP cannot be denied, it has become necessary to clarify their formation mechanisms to reduce radiation exposure of the workers to the extent possible during decommissioning work.

Therefore, in the present study, the formation mechanisms are reexamined from wider viewpoints, such as thermal hydraulics in the reactor building (R/B), environmental monitoring data at Tokai, which is on the way from FDNPP to Tsukuba- city, dose rate measurement at R/B after the accident: knowledge of fission products (FP) aerosol behavior during SA considering slightly bold hypotheses based on the author's experiences with SA studies [9].

2. Review of previous studies on the Type A particles

Japan Meteorological Agency, Meteorological Research Institute, which is located in Tsukuba- city in Kanto district, first observed that relatively large amounts of Type A particles were trapped by the dust filters used for environmental measurement between 21:10 on March 14 and 09:10 on March 15 [2]. Thereafter, many studies on Type A particles were conducted, and the following knowledge was obtained.

(1) The release timing of Type A particles reversely from the FDNPP, as estimated using wind direction and wind speed on March 14 and 15, was between 17:00 on March 14 and 02:00 on March 15 [2].
(2) The Type A particles were covered with almost pure silicate glass. The specific radioactivity of the Type A particles was higher by about two orders of magnitude than that of the Type B particles [3].
(3) Less than 0.5 wt. % of uranium [10] was included in the Type A particles.
(4) The Type A particles had an amorphous structure, and a quenched trace was observed [8].
(5) The Type A particles include an inorganic zinc coating ingredient [8] that was originally painted inside of the S/P wall.
(6) Particles almost identical to the Type A particles found in Tsukuba- city, were observed at the northwest region of the FDNPP as well, which is considered a contaminated area owing to Cs release from Unit 2 [11].

3. Inference of Formation Mechanisms

3.1 Formation of particles by evaporation/condensation or atomization

Representative formation mechanisms of radionuclide particles (aerosols) during SA of NPPs are evaporation/condensation or atomization of liquid [12].

In the evaporation/condensation process, particles are formed by nucleation under supersaturation of the vapor generated by heating of the target materials above the boiling point and subsequent cooling. Nucleation consists of homogeneous nucleation involving the vapor of a single ingredient and heterogeneous nucleation involving the vapor of multiple ingredients.

In the atomization process, particles are formed by separating of the molten (liquid) materials into fine pieces in a high-speed flow. Alternatively, particles can be formed by entrainment at the molten pool surface during molten core-concrete interaction (MCCI), but it can be ignored for the purposes of this study because the amounts of particles formed and released by this mechanism is very small.

In a RPV during SA, various types of vapors that originated from the core structures or radionuclides could become airborne because of high temperature. Therefore, during condensation at time of temperature decrease, the surface ingredients of the particles may not be uniform. Instead, heterogeneous nucleation could likely occur because various chemical species condenses onto the surface of the particles.

Based on the reasons mentioned above, it is concluded that the condition of Type A particles covered by mostly pure silicate glass cannot be explained by the evaporation/condensation process. By contrast, the condition of the said particles can be explained by the atomization process. However, the following additional conditions are necessary for the formation of Type A particle. Before melting, relatively large amounts of pure silicate glass must have existed at a certain location in the R/B, and the radionuclides including Cs must have been physically close to the silicate glass (See section **3.2.1**).

3.2 Study on plant phenomena and the formation process of Type A particles

To induce atomization, high temperature is necessary for melting the silicate glass, and a high-speed flow is necessary for dividing the molten materials into small pieces. In case of the Fukushima accident, such energetic phenomena were reportedly associated only with the hydrogen explosions in Units 1 and 3.

Hydrogen burn or deflagration occurs when hydrogen and oxygen concentrations exceed 4% and 5%, respectively, and hydrogen detonation occurs when hydrogen concentration exceeds 18.3% [13].

Hydrogen detonation with air blast speed greater than the speed of sound and accompanied by a shock wave, occurred in Unit 3, while hydrogen deflagration with air blast speed of several 100 m/s occurred in Unit 1 [14].

Therefore, the Type A particles could have been formed by the following process. Some silica sources might have been melted by the high-temperature fire (> 3000K) caused by hydrogen detonation, and the high-speed air blast might have atomized the molten materials and formed the Type A particles. An image of the atomization of silicate glass melt due to the hydrogen explosion is shown in **Fig. 2.**

3.2.1 Sources of silicate glass

The source of the silicate glass covering the Type B particles that originated from Unit 1, has not yet been identified clearly. However, because it has been reported that several heat insulators covering the isolation condensers (ICs) on the fourth floor of R/B were broken during the hydrogen explosion at Unit 1 [15], this could have been the source of silicate glass. That is, the heat insulators were heated and molten by the fire of the hydrogen explosion. Immediately after that, the molten insulators were divided into small pieces by the high-speed flow and drawn out from the fourth to fifth floor by the reverse flow which was caused by the formation of almost vacuum-like conditions at the center of the blast (somewhere on the fifth floor) immediately after the hydrogen explosion. During these processes, Type B particles could have been formed.

By contrast, because there were no ICs and the associated heat insulators in Unit 3, the source of silicate glass covering the Type A particles must be different from that of the Type B particles. Therefore, the author focused on the measured dose rates at High Efficiency

Particulate Air (HEPA) filters made of glass fiber in the Standby Gas Treatment System (SGTS), of which the measured dose rates in Unit 3 after the accident were extremely lower relative to those of Units 1 and 2 although the three units caused similar SA.

The dose rates [16], [17], [18], [19] at the HEPA filters on the SGTS line in Units 1 to 4 were measured in Aug. 2011 – Nov. 2014 are listed in **Table 2**, together with the schematic of SGTS with HEPA filters in Unit 3 (see **Fig. 3**). Because the SGTS line was designed as "Fail open" during a total station blackout, it was expected that the atmosphere of R/B, including various types of radionuclides, flowed continuously into this line during the accident. As a result, large amounts of Cs could have been deposited on the HEPA filter.

Therefore, the low dose rates measured in Unit 3 suggest that only the HEPA filters in Unit 3 were not located in their original place owing to several reasons. That is, the glass fibers could have melted (the boiling point of glass fiber = about 1120K) by the fire of hydrogen detonation and been blown out by the high-speed air blast. Otherwise, vacuum-like conditions were formed at the detonation center just after the explosion, and a countercurrent flow was formed in the SGTS line up to the gravity dumper (GD), which prevents countercurrent flow. As a result, atomization of the molten glass fiber occurred at the inlet or the failure points of SGTS line, and this might have been the source of the silica glass covering the Type A particle.

If this hypothesis is assumed correct, the silicate glass covering the Type A particles is the material of the HEPA filter that traps radionuclides, including Cs. Therefore, this explains without contradiction that the specific radioactivity of the Type A particles was more than two orders of magnitude higher than that of the Type B particles, which were covered with silica glass that originated from the heat insulator.

3.2.2 Study of particle diameter

Because most of the Type A were spherical, the surface tension of the molten materials was thought to be dominant in the process of particle formation by atomization. In that case, the following dimensionless Weber (We) number in Eq.(1), that is, the ratio of inertia to surface tension, could constant regardless of the particle size.

$$We = \frac{\rho L V^2}{\sigma} \qquad (1)$$

where, ρ: density, L: diameter, V: velocity, and σ: surface tension.

Hydrogen deflagration occurred in Unit 1, while hydrogen detonation occurred in Unit 3 [14]. Therefore, the velocity of the air blast associated with the hydrogen detonation in Unit 3 was higher than that of the hydrogen deflagration in Unit 1. If it is assumed for simplification that the density and surface tension of the silica sources are the same between Units 1 and 3, the diameter of the Cs particles that originated from Unit 3 would be smaller than that of the particles that originated from Unit 1. This corresponds to the trend seen in the measured values.

3.3 Release mechanism of Type A particles into the environment

The hydrogen explosion that occurred in Unit 3 on the fifth floor of the R/B at 11:01 on March 14 [14] is considered the detonation. Because the wind direction in the FDNPP neighborhood at that time was west-southwest, it has been inferred that most of the Type A particles formed during the hydrogen detonation were transported along the east-northeast direction (the Pacific Ocean).

By contrast, lateral damage to the west wall of R/B in Unit 3 (see **Fig. 4**) suggests that the air blast associated with the explosion spread in all directions, that is, not only to the ceiling, walls, and floor of the fifth floor but also the third and fourth floors were partly damaged by the blast. Therefore, the Type A particles formed during the hydrogen detonation were considered to have been transported partly to elevations lower than the fifth floor or the deep portion of the SGTS line, and deposited by inertia, thermophoresis, gravitational settling [20], and so on.

Thereafter, a few flows could have been generated several times inside R/B in Unit 3 because of the containment venting or steam generation inside the drywell (D/W) or RPV owing to the restart of core cooling water injection after an interruption lasting several hours. Then, the once-deposited Type A particles could have been released by resuspension when the drag force of the flow exceeded the adhesive force of the deposited particles. The expected mechanisms of particle deposition and resuspension in R/B at Unit 3 are shown in **Fig. 5**.

4. Verification of proposed formation mechanisms

4.1 Phenomena within NPPs and discussion of ties with environmental monitoring data

The attainability of the Type A particles to Tsukuba- city or to the region northwest of the FDNPP was examined by combining the plant phenomena responsible for the release of the Type A particles with the atmospheric diffusion factors, such as wind direction and velocity. The phenomena that caused resuspension of the particles in the R/B in Unit 3, wind direction, and particle attainability to Tsukuba- city or to the region northwest of the FDNPP are listed in **Table 3.**

4.1.1 Hydrogen explosion at 11:01 on March 14

When the hydrogen explosion in Unit 3 occurred at 11:01 on March 14, the wind direction was west-southwest. Therefore, most of the formed particles were transported to the Pacific Ocean. However, the wind direction changed toward the north at 20:00, north-northeast at 22:00, and northeast thereafter. Therefore, the particles that were transported to the Pacific Ocean mostly reversed and approached the Kanto district [21], but they could not reach Tsukuba-city before 09:00 on March 15.

In addition, most of the Type A particles may have been removed by the sea during the 24 hours travel because the gravitational settling was more effective than that of soluble particles due to larger Type A particle size than that of soluble one, and the Type A particles could not have been re-suspended from the sea surface, once they touched the sea (see section 5.2).

4.1.2 Restart of core cooling water injection at 15:30 on March 14

The core cooling water injection was restarted at 15:30 on March 14 although it was stopped owing to a break in the line just after the hydrogen explosion in Unit 3[14]. Then, it is expected that the Type A particles deposited at the lower elevations of R/B were re-suspended owing to the steam flow generated by the injection. However, the wind direction at that time was west-southwest, and most of the particles were transported to the Pacific Ocean for about 5 h. Although the particles approached the land early in the morning of March 15 because of the reversal of wind direction, these particles were not measured at Tokai, which is on the way from the FDNPP to Tsukuba- city. Therefore, the Type A particles released at this time were less likely to be observed in Tsukuba- city.

4.1.3 Restart of core cooling water injection at 02:30 on March 15

After that, the core cooling water injection was stopped again at 20:30 on March 14 and restarted at 02:30 on March 15 after an interruption of about 6 h (See pages 256 - 257 in Reference [14]). Notably, this interruption was not well considered or treated in most of the precedent studies such as BSAF [22].

Just after this restart of core cooling water injection, a flow was thought to have been formed in R/B owing to the generation of a large amount of steam in the high-temperature RPV or D/W, followed by a leakage from the top flange of D/W or D/W failure locations. Notably, measured D/W pressure did not increase considerably at 15:30 on March 14 and at 02:30 on March 15 [14]. This is possibly because the pressure increase was suppressed owing to the balance of the flow between the leakage to R/B and the steam generation inside D/W. In addition, the measured water level in the RPV did not increase at these times owing to the possibilities of failed indications [14].

Accordingly, it can be concluded that the R/B flow just after 02:30 on March 15 could have led to resuspension of the Type A particles formed as a result of the hydrogen detonation in Unit 3, and their deposition at lower elevations of R/B or deep locations in the SGTS line. At that time, the wind direction was north-northwest and most of the re-suspended Type A particles were transported to Kanto district.

4.1.4 Consistency with environmental monitoring data at Tokai

The ^{137}Cs environmental monitoring data recorded early in the early morning on March 15 at Tokai, which is on the way from the FDNPP to Tsukuba- city is shown in **Fig. 6**. At 06:30 on March 15, a high concentration of Cs particles was measured at Tokai as the third peak in the monitoring data recorded in the morning [23].

Actually, if it is assumed that the Type A particles were released at 02:30 on March 15, they could have passed through Tokai (114 km away from the FDNPP) between 06:30 and 7:00 and arrived in Tsukuba- city between 08:30 and 09:00 (172 km away from FDNPP). Therefore, 02:30 on March 15 is considered promising as the release time of the Type A particles.

In addition, if the release of Type A particles occurred because of resuspension, it should have lasted for a relatively short time immediately after the core cooling water injection was restarted because the large amounts of steam required for the process can be generated only for short periods. Therefore, the first half of the third peak was appropriately occupied mostly by Type A particles.

The first and second peaks in **Fig. 6** can be ascribed to steam generation due to restart of the core cooling water injection in Unit 1 at 21:30 on March 14 after an interruption of 19 h, and leakage from the top flange of the D/W in Unit 2 at 00:00 on March 15 based on the measured increase in D/W pressure and subsequent stabilization at 0.8 MPa, respectively.

4.1.5 Hydrogen explosion in Unit 4 at 06:10 on March 15

The hydrogen explosion occurred on the fourth floor of the R/B in Unit 4 at 06:10 on

March 15 because a part of the hydrogen produced in Unit 3 was delivered to the R/B in Unit 4 through the SGTS line of Unit 3, the downward piping of which is connected to that of Unit 4. Just after the hydrogen explosion at Unit 4, the air blast of the explosion possibly flew backward in the SGTS line and caused resuspension of the once-deposited Type A particles in locations within the SGTS line or at lower elevations of the R/B in Unit 3.

The released Type A particles were transported to Kanto district by the north-northeast wind, but the particles could not arrive in Tsukuba- city before 09:10 on March 15 based on the wind velocity at the time. The particles might have arrived in Tsukuba- city in the daytime or might not have arrived on the same day because the wind direction varied from the northeast to south just before noon on March 15. This corresponds to the other study that ^{137}Cs observed at Naraha at 7:00 on March 15 was not transported to the Kanto area but to southern Nakadori by a northeasterly or easterly wind [24].

4.1.6 Venting of S/P at Unit 3 at 16:05 on March 15

The possibility that the Type A particles were re-suspended by a flow in the R/B that was generated just after S/P venting in Unit 3 at 16:05 on March 15 exists. The released Type A particles could have been transported along the northwest direction of the FDNPP by the east-southeast wind, together with the radionuclides released from Unit 2 because of an increase in the core temperature of Unit 2 at the time.

4.2 Consistency of previous studies with proposed formation mechanisms

This section discusses the consistency between the results of previous studies described in **section 2** and the hypothesis of present study about the formation mechanisms of the Type A particles.

(1) Considering the wind direction and velocity from the evening of March 14 to the early

morning of March 15, the release time of the Type A particles was estimated reversely to be between 17:00 on March 14 and 02:00 on March 15 [2]. By contrast, the proposed time of release that corresponds to the timing of core cooling water injection identified in **section 4.1.3** is only 30 min later than the predicted time. However, the wind velocity tended to increase toward the south in the morning of March 15; therefore, the proposed timing is thought to be within the error range of atmospheric dispersion calculations.

(2) The specific radioactivity of the Type A particles was higher by two orders of magnitude than that of the Type B particles, and the Type A particles were covered with mostly pure silicate glass [7]. As described in **section 3.3**, if it is assumed that the Type A particles were produced by the hydrogen detonation that induced atomization of the liquefied HEPA filters installed in the SGTS line of Unit 3, the mostly pure silicate glass that covered the Type A particles and their high specific radioactivity can be explained without any contradiction.

(3) Less than 0.5 wt. % of uranium [10] was included in the Type A particles. Because the core temperature was thought to have exceeded the melting point of UO_2 during the Fukushima accident, the release fraction of uranium could be higher than 0.1% [25]. Considering the core inventory of uranium is larger at least by 3 orders of magnitude than that of radionuclides such as Cs, the uranium released with a core temperature rise could be carried with other radionuclides to R/B and is more likely to be included in the Type A particles.

(4) The Type A particles have an amorphous structure, and they exhibit a quenched trace [7]. If energetic phenomena, such as steam explosion followed by atomization, occur, the resulting particles can have an amorphous structure because of quenching due to a rapid increase in the contact area between the melt and atmosphere [26]. It is thought that a similar quenching phenomenon could have occurred in the process of atomization due to the hydrogen detonation.

(5) The Type A particles contained the inorganic zinc coating ingredient [8] that was originally painted inside of the S/P. If the coating ingredient was partly dissolved in the S/P water, a

hygroscopic aerosol such as cesium hydroxide (CsOH) takes this water in the aerosol, and the aerosol is released into the R/B through the vacuum breaker and the failure points of D/W, and the particles formed in R/B due to the hydrogen detonation could contain the inorganic zinc coating ingredient.

(6) Particles similar to the Type A particles found in Tsukuba- city were found in the region northwest of the FDNPP as well, which is considered as the area contaminated by the radionuclides released from Unit 2 [11]. This contamination occurred by the radionuclides released from Units 2 and 3 concurrently with the rain on the afternoon of March 15. However, the contaminated area varied delicately based on the wind direction at time of releases, and the major releases occurred when the core temperature increased. The major release from Unit 3 in the northwest direction is considered to have occurred after 21:00 on March 15. By contrast, the release of the Type A particles may have occurred at 16:05 at time of venting of Unit 3 when the major release occurred from Unit 2. In that case, it can be explained how the Type A particles originated from Unit 3 origin were found in the area contaminated by Unit 2.

5. Discussions

5.1 Fraction of Type A particles to the total amount of Cs release

If the proposed hypothesis is assumed correct, the Cs that contributed towards formation of the Type A particles was equal to the amount that pre-existed on fifth floor of the R/B or had been trapped by the HEPA filters in Unit 3 before hydrogen detonation. Since the previous studies have not quantified yet the fraction of Type A particles to the total Cs amount released into the environment, the fraction was preliminarily estimated based on the proposed formation mechanisms.

The ^{137}Cs inventories of Units 1 - 3[27] and their cumulative releases into the environment predicted by the MELCOR code[28] are shown in **Tables 4 and 5**, respectively. For simplification, the following assumptions were made. 1) All the molten fuels and Cs moved from the RPV to the containment at time of the RPV melt-through in the morning on March 13, 2) Time from the RPV melt-through to the hydrogen detonation was about one day, 3) Since the containment may have been kept intact before the hydrogen detonation, leak rate of the containment was equal to the design value, 0.5%/day[29], and 4) Cs release into the environment before the hydrogen detonation could be ignored.

The calculated Cs amount in the R/B in Unit 3 is $2.4 \times 10^{17} \times 0.005 = 1.2 \times 10^{15}$Bq, which is equal to about 8% of the total amount of Cs released into the environment (1.5×10^{16} Bq). Approximately 92% of the Cs, except for these insoluble Cs particles, is considered to have soluble forms such as CsOH, CsI, and $CsBO_2$, as predicted in the previous studies. Notably, the present calculation was performed using an expedient and the result needs to be validated by detailed calculations or the studies based on the environmental monitoring data.

5.2 Removal of radionuclides in air during an atmospheric dispersion on the sea

The other study [21] suggested that the hydrogen detonation in Unit 3 may have not released Iodine and Cs in large amount because these radionuclides were not measured in the plume that traveled about 24 hours on the sea and the deposited mass at Kanto district on March 15. In addition, from our experience, the depletion of radionuclides in air due to the dry deposition for about 24 hours travel is not so significant in case of an atmospheric dispersion on the land.

By contrast, the prediction reversely by the WSPEEDI code [5] with the measured sea surface concentration showed a huge amount of Cs and Iodine release at the time of hydrogen detonation in Unit 3. Another study [30] also showed a large amount of Tellurium (Te) release at the time of hydrogen detonation in Unit 3. The Te release was reversely predicted by using an atmospheric dispersion code with the measured increase in Te deposited mass at Kanto district on March 15. That is, certain amount of Te actually reached Kanto district after about 24 advection and diffusion on the sea. It is difficult to consider that only the release of Te without Cs or Iodine occurred at the time of hydrogen detonation in Unit 3 taking into account their volatilities. Judging from this consideration together with the WSPEEDI reverse prediction, a large amount of Cs and Iodine could have been released together with Te, but most of Cs and Iodine were removed by the sea during their travel on the sea due to the following reasons.

The Cs released at the time of hydrogen detonation in Unit 3 could have included soluble particles such as CsOH and insoluble Cs (Type A) particles. In case of soluble particles, overall concentration in the plume could have decreased continuously for about 24 hours travel because the soluble particles near the sea surface dissolved into the sea even if there is no precipitation and the dry deposition is less significant. Otherwise, since CsOH makes hygroscopic particles, the deposition velocity of the particles could have increased because the particles absorbed humidity in air and increased the diameter by hygroscopicity.

In case of the insoluble Cs particles, they could have been also removed by the sea because the gravitational settling was more effective than that of soluble particles due to their larger particle size, and the insoluble particles could not have been re-suspended from the sea surface, once they touched the sea.

In contrast to the soluble Cs and Iodine, Te is insoluble. Therefore, it is considered that Te concentration in air was not depleted significantly during an atmospheric dispersion on the sea. Accordingly, comparison of the two studies [21], [30] suggests that there could be a difference in depletion of radionuclides in air during an atmospheric dispersion on the sea between soluble and insoluble particles. Further investigation on this issue is necessary in future.

6. Conclusions and Future Challenges

In previous studies, it was proposed that Type A particles were formed by evaporation/condensation in the RPV of Unit 2 and subsequent quenching by water from the S/P. By contrast, in the present study, a different formation mechanism was proposed, according to which the particles were formed due to melting of the glass fiber of the HEPA filters in the SGTS line as a result of the fire of hydrogen detonation in Unit 3 at 11:01 on March 14, and atomization of the liquid by the air blast of the explosion, followed by quenching of the molten materials.

Most of the formed particles were dispersed to the Pacific Ocean because of the wind direction at that time. However, some of them were deposited at lower elevations of the R/B in Unit 3 or at deep locations in the SGTS line. These particles could have been subsequently re-suspended and dispersed toward Kanto district or the region northwest of the FDNPP by the steam flow in the R/B owing to the large amounts of steam generated just after the core cooling water injection was restarted at 02:30 on March 15 or owing to venting of the S/P at 16:05 on March 15, respectively.

The author strongly expects that this information will be utilized to reduce radiation exposure to the workers at time of, for example, SGTS demolition work in Unit 3. In addition, because this study was conducted based on the limited measured data that the author focused on intentionally, and calculated guess based on circumstantial evidence, as well as considering the slightly bold hypothesis of the author, the acquisition of real data that support this hypothesis is an urgent matter. Moreover, the adequacy of the present hypothesis should be confirmed through separate effect tests such as reproduction of the insoluble particles by atomization of melted glass fibers or through further investigation of consistency between the resuspension timings or the spatial distribution of the insoluble particles based on the hypothesis and detailed environmental information, i.e. by analyzing filter tapes from suspended particulate matter (SPM) monitors.

Acknowledgments

The author would like to express his deep gratitude to Dr. Yukihiko Satou of Japan Atomic Energy Agency and Dr. Shinya Mizokami of Tokyo Electric Power Company Holdings, Inc. for their helpful comments to prepare this paper. Special thanks are also due to Dr. Mizuo Kajino and Dr. Kouji Adachi of Meteorological Research Institute for their giving a lot of technical suggestions. Professor Philip Beeley of Khalifa University kindly checked English expressions and grammar of this article.

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Table lists

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Fig. 6 Environmental monitoring data of ^{137}Cs measured at Tokai in the early morning on March 15 [23]

Table 1 Outlines of measured insoluble Cs particles [3]

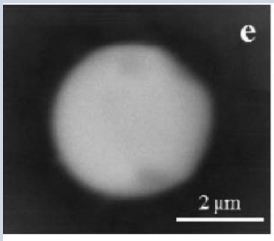
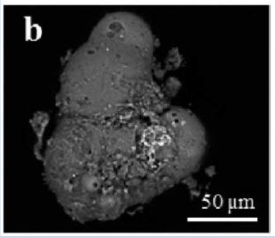
	Type A	Type B
Reference pictures		
Diameter（μm）	1～10	Several tens ～ Hundreds
Radioactivity（Bq/1 particle）	1～ Several tens	Several tens ～ Thousands
Shape	Spherical	Not spherical
Measured location	Tsukuba-city, Tokyo, North West direction of Fukushima Daiichi	North side neighborhood of Unit 1
$^{134}Cs/^{137}Cs$	～1.04 Origin of Unit 2 or 3 ?	～0.94 Origin of Unit 1
Condition	Fission Products covered by silicate glass	

Table 2 Measured dose rates at HEPA filters of SGTS line [16], [17], [18], [19]

Unit mSv/h

Measure location in Fig. 3	Measured date	①	②	③	④	⑤	⑥
Unit 1	Sep. 2015	10*)					
Unit 2	Nov. 2014	220	460	1000	44	160	850
Unit 3	Dec. 2011	2.0	3.5	1.3	1.6	3.2	3.1
Unit 4	Aug. 2011	0.1	0.5	6.7	0.1	0.5	5.5

*) Dose rates near HEPA filters in Unit 1were so high that they were measured from outside of the shielding.

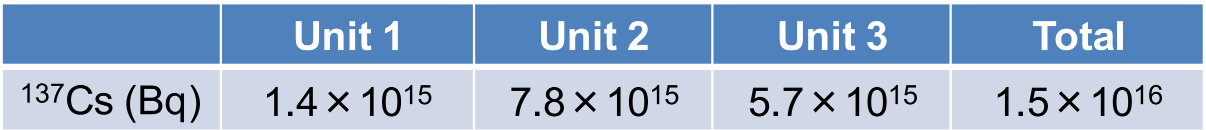
Table 3 Phenomena that cause resuspension of particles in the R/B of Unit 3, wind direction and attainability of re-suspended particles to Tsukuba- city or to the northwest region of FDNPS

Events of Unit 3	Particle condition	Wind direction	Particle attainability	Judge
Mar. 14 11:01 H_2 explosion	Generation of type A Cs particle by atomization	Just after H_2 explosion: WSW. 10 hr later: NNE	The particle can not reach Tsukuba before 09:10, March 15 but can reach Tokyo before evening of March 15.	×
Mar. 14 15:30 Restart of water injection after 4.5hr interruption	Resuspension of once deposited particles	Just after release: W. 6 hr later: NNE.	The particle can reach Tsukuba before 09:10 In removal of most of particles by sea. Not measured in Tokai.	×
Mar. 15 02:30 Restart of water injection after 6 hr interruption	Ditto	NNE	The particle can reach Tsukuba before 09:10. Measured in Tokai.	○
Mar. 15 06:10 H_2 explosion at Unit 4	Resuspension of particles by blast of the explosion	NNE	The particle can not reach Tsukuba before 09:10 but may reach after that.	△
Mar. 15 16:05 Ventilation of S/C	Resuspension of particles by flow of S/C ventilation.	SE	Dispersion of particles to NW direction of Fukushima Daiichi	○

Table 4 ^{137}Cs inventories of Units 1 - 3 [27]

	Unit 1	Unit 2	Unit 3
^{137}Cs (Bq)	2.02×10^{17}	2.55×10^{17}	2.41×10^{17}

Table 5 Cumulative ^{137}Cs release into the environment predicted by the MELCOR code [28]



	Unit 1	Unit 2	Unit 3	Total
^{137}Cs (Bq)	1.4×10^{15}	7.8×10^{15}	5.7×10^{15}	1.5×10^{16}

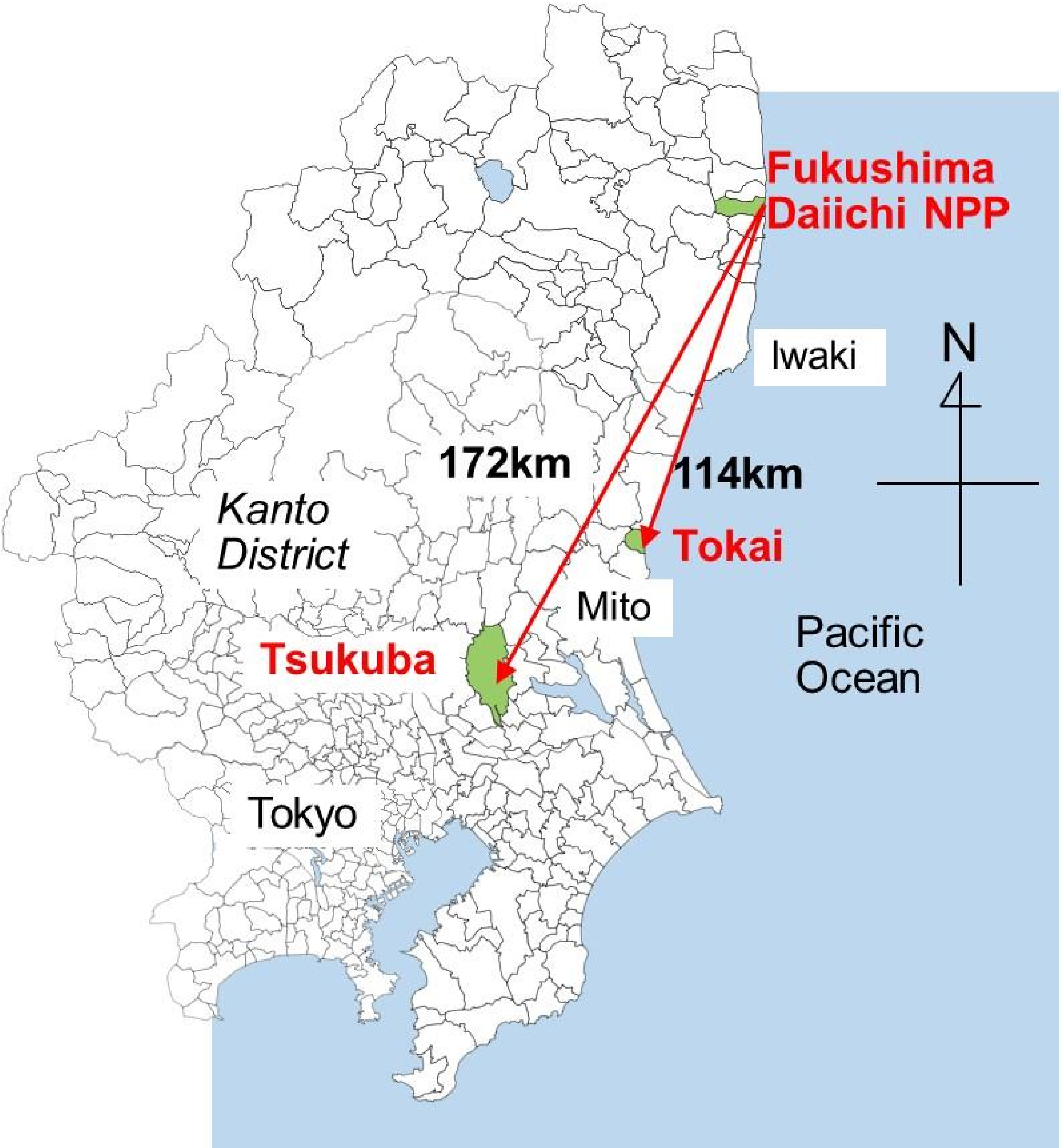


Fig. 1 Location of Tsukuba- city, Tokyo, Tokai, and Fukushima Daiichi NPS on Japanese map

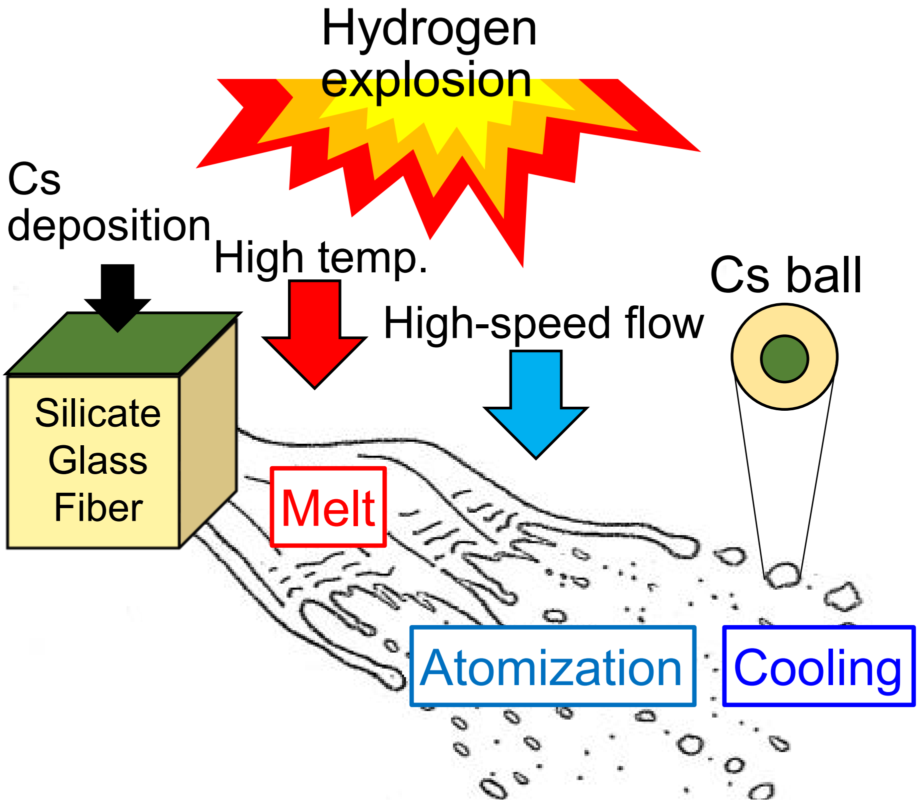


Fig. 2 Image of atomization of silicate glass melt due to the hydrogen explosion

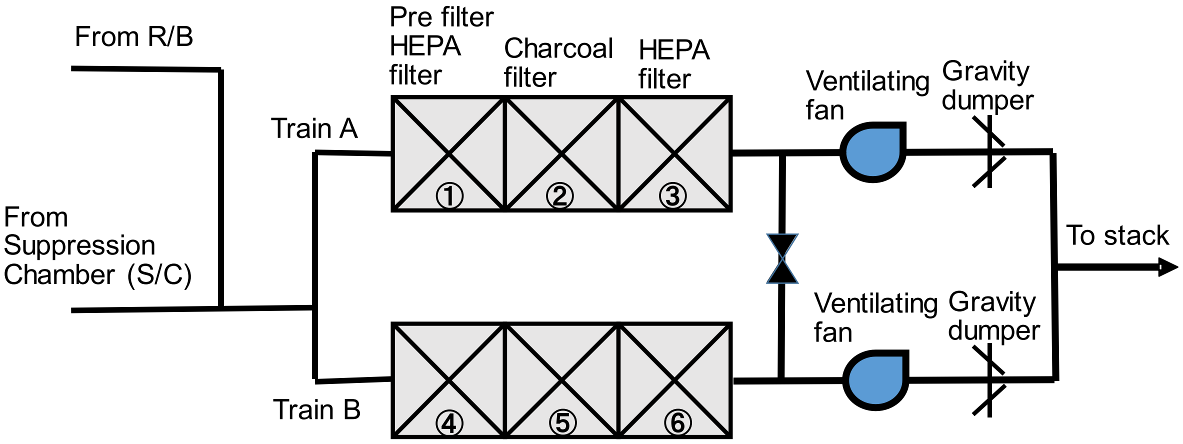


Fig. 3 Schematic of SGTS line with HEPA filters in Unit 3

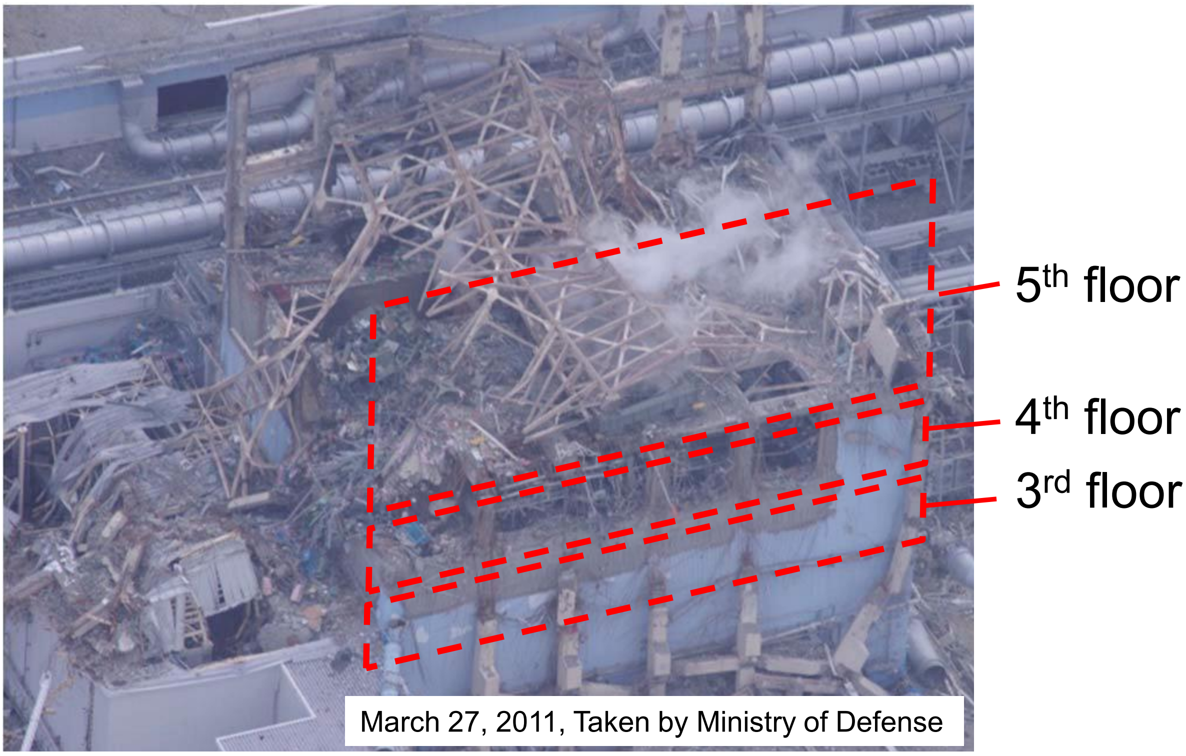


Fig. 4 Lateral damage to the west wall of R/B in Unit 3

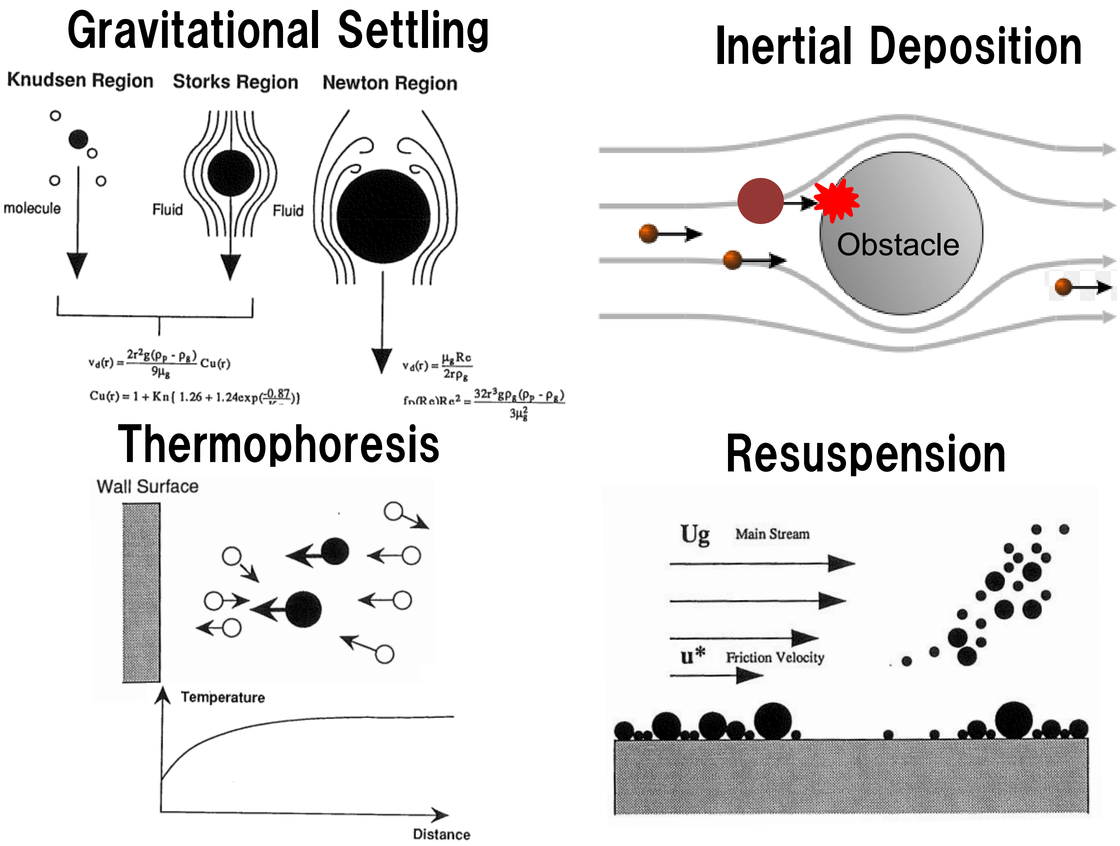


Fig. 5 Mechanisms of particle deposition and resuspension expected in Unit 3

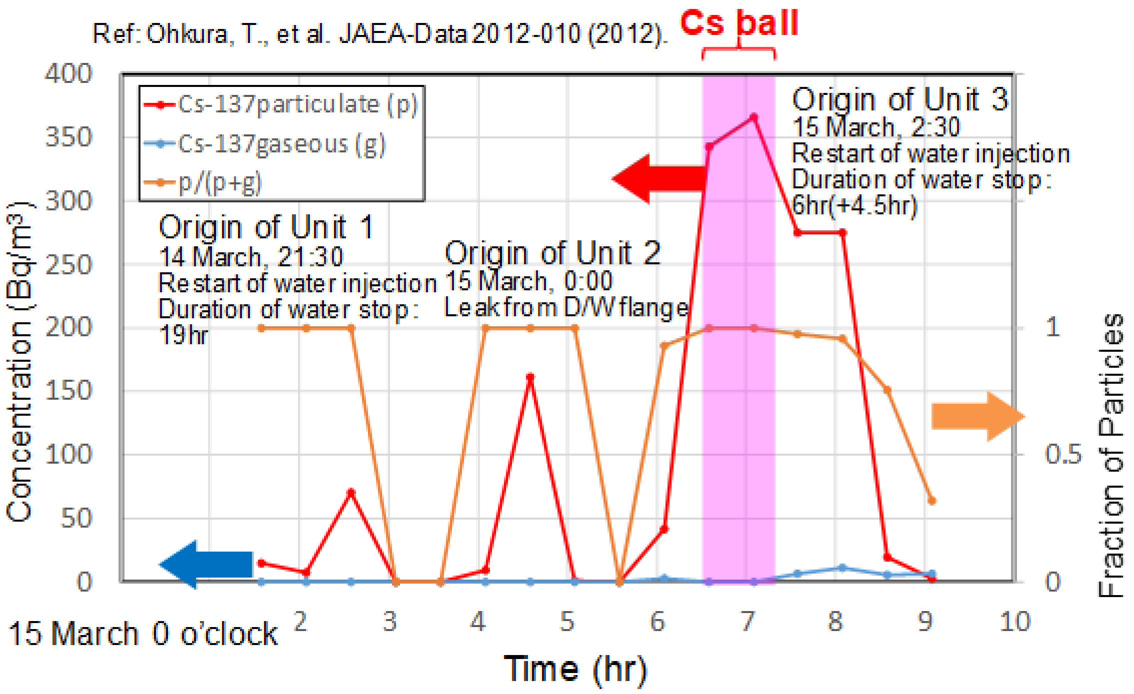


Fig. 6 Environmental monitoring data of ^{137}Cs measured at Tokai in the early morning on March 15 [23]