Settlement process of radioactive dust to the ground inferred from the atmospheric electric field measurement

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Abstract. Radioactive materials from the accident at Fukushima Dai-ichi nuclear power plant (FNPP) in March 2011 spread over a large area, increasing the atmospheric electric conductivity by their ionizing effect, and reducing the vertical (downward) component of the DC electric field near the ground, or potential gradient (PG). PG data at Kakioka, 150 km away from the FNPP, showed independent changes compared to the radiation dose rate, and a comparison of these data revealed the local dynamics of the radioactive dust.

(1) The initial drop of the PG to almost zero during 14–15 March is most likely due to radioactive dust suspended in the air near the ground during cloudy weather. (2) An episode of PG increase to more than 50 V m\(^{-1}\) on 16 March is most likely due to the re-suspension of the radioactive dust from the surface and subsequent removal from Kakioka by the strong wind from the non-contaminated area. (3) Low but finite values of the PG during 16–20 March most likely reflect a reduced amount of radioactive material near the ground after the above wind transported away the majority of the suspended radioactive dust. (4) Very low values of the PG after substantial rain on 20–22 March most likely reflect settlement of the radioactive material by rain-induced fallout. (5) Temporal recovery of daily variations from the end of March to the middle of April with low nighttime fair-weather baseline PG most likely reflects re-suspension of the radioactive dust into the air from the ground and trees, and subsequent transport to the other region or fallout to the ground until late April. (6) Weakening of the daily variation and gradual recovery of the nighttime fair-weather baseline after mid-April suggests a complete settlement of the radioactive material to the ground with partial migration to the subsurface.

Keywords. Atmospheric composition and structure (Aerosols and particles; Pollution – urban and regional) – Meteorology and atmospheric dynamics (Atmospheric electricity)

1 Introduction

The accident at the Fukushima Dai-ichi nuclear power plant (FNPP) that was triggered by the Tohoku Earthquake on 11 March 2011 caused a release of a massive amount of radioactive material. Although the total amount of the radioactive material released by the Fukushima Accident (\(10^{17}\) Bq for Iodine \(^{131}\)I and \(10^{16}\) Bq for Cesium \(^{137}\)Cs according to NISA, 2011) is less than that from the Chernobyl disaster on 26 April 1986, it still caused a substantial contamination of an area of several hundreds kilometer in diameter. A map of the contamination level has been obtained after a large number of soil sampling was analyzed using manpower (MEXT, 2011). However, dynamics of the radioactive dust and temporal change of the inter-regional contamination have not been well understood for both primary and secondary contamination. For the future planning for health protection and agricultural/fishery activities, it is important to understand the settlement process of the radioactive materials to the soil as well as the secondary transport, including daily dynamics by wind blowing or rain. Therefore, any methods to diagnose the transport and dynamics of the radioactive materials are useful.

We use here the vertical (downward) component of the atmospheric electric field, or potential gradient (PG), and combine it with the radiation dose rate data. Large-scale DC
electric current is flowing in the air between the ground and the ionosphere at a global scale (e.g., reviews by Rycroft et al., 2000, 2008; Williams, 2009). Since both the ionosphere and the ground are highly electrically conductive compared to the atmosphere, the current generates a relatively vertical electric field of about 100–150 V m\(^{-1}\) at ground level except near electrified clouds. The PG is affected by the atmospheric electric conductivity that is partly controlled by ionization rate and loss rate, obeying the ion balance equation (Rosen and Hofmann, 1981; Makino and Ogawa, 1985; Rycroft et al., 2008). Therefore, the PG decreases when local ionizing radiation increases (Pierce, 1959; Hamilton and Paren, 1967; Harrison, 2003). This effect is not significant for already-conductive ground or seawater, but it is significant for the near-ground atmosphere.

This mechanism actually caused quick PG drops by one order of magnitude within one to a few hours after rain-induced radioactive fallout (wet contamination) related to nuclear tests or to the Chernobyl disaster at PG stations at distances up to more than 1000 km from the test site or Chernobyl (Harris, 1955; Israelsson and Knudsen, 1986; Tuomi, 1988, 1989). The quick drop of PG was also observed at Kakioka, which is located 150 km southwest of the FNPP, after the first massive southbound release of radioactive material from the FNPP on 14 March 2011 (Takeda et al., 2011, hereafter referred to as Paper 1). The FNPP-related PG changes took place as a result of wind-driven low-altitude transport of the radioactive dust without rainfall (dry contamination). Therefore, the contamination forms of the radioactive materials near the ground during this drop is most likely different from those during the previous events of quick PG drops; i.e., radioactive fallout in the FNPP case is most likely suspended in the air near the ground surface or just attaching on the surface without strong binding to the soil matrix (IAEA, 2006, Sect. 3.1).

Ideally, the degree of the changes in the electric conductivity or local ion density is expected to reflect the level, form, and transport of the contaminated radioactive materials that have different spatial distribution compared to the ions. Therefore, the PG is expected to respond differently at the different phases of the settlement and dynamics of the FNPP-origin radioactive materials for the same amount of the contamination. In fact, the time profile of the PG at Kakioka is not well correlated to those of radiation dose rates at the nearest stations to Kakioka, as one can see in Fig. 1 (see next section for explanation). By comparing the simultaneous observations of the radiation dose rates and weather records, it is possible to interpret minor PG changes that reflect physical processes or dynamics of the local radioactive materials and local ions. We use 1-min resolution PG data at Kakioka and 1-h resolution data of the ground-level radiation dose rate and meteorological records.

2 Observation

Figure 1 shows 1-h averaged PG at Kakioka, radiation dose rate at Ibaraki-cho (25 km east of Kakioka) and rainfall from Kakioka during March–May 2011. The sensor uses the Water-Dropper Collector, and is placed at 2.55 m high with 1.17 m separation from the wall inside a house (Shigeno et al., 2001). The sampling rate is 1 Hz. The earthquake caused power failure for about three days. The geographical locations of Kakioka, Ibaraki-cho, and the FNPP are shown in Fig. 2. The numbers (1) to (6) and the stepped blue lines refer to segments in the time series that are discussed below. Local noon is near 03 universal time (UT). The initial drop of PG to near-zero values at around 21 UT, 14 March together with the increase of the radiation dose rate have been reported in Paper 1, and interpreted as the signature of arrival of a massive amount of radioactive materials at Kakioka by low-altitude wind. We examine here the variation after 15 March.

Although the PG is affected by the ionizing radiation, the changes in the PG are not well correlated with the changes in the radiation dose rate. To show this, data in March are enlarged in Fig. 3. After the arrival of the radioactive materials at Kakioka on 14 March, radiation dose rate at both Ibaraki-cho and Mito (30 km northeast of Kakioka) showed spike-like increases lasting 1–3 h at around 03 UT and 20 UT on 15 March, and also at around 02 UT and 20 UT on 20 March. Apart from the first spike, the radiation dose rate stepped to a higher level after the spike than the value before the spike. Among these four events, only the last and largest increase occurred together with a substantial change in the PG. The PG increase on 16 March is related neither to the radiation dose rate nor to rain. Another example of non-correlation is the PG value: it is completely different between the first period and the third period for the same level of the radiation dose rate at Ibaraki-cho.

The spikes of radiation dose rate shown in Figs. 1b, 3b, and 3f between 13 and 20 March are observed when, and only when, the wind steadily blows from northeast, i.e., from the FNPP for a few hours, and are not necessarily related to rain. Even for the spikes observed together with the rain (20 UT on 15 March and 20 UT on 20 March), the timing of the rain and that of the spike of the radiation level are not exactly the same. The time profile of each spike is not exponential decay, and the exponential decay started after the spike was over. Since exponential decay generally corresponds to physical radioactive decay of radionuclides that contaminate the surface, the spike part should be caused by radiation sources that do not fallout to the ground, i.e., remain suspended in the air, without contributing significantly to the atmospheric electric conductivity near the ground. All these data indicate that wind carried a dense radioactive dust plume from the FNPP during the spikes, leaving only a part of the dust to fallout to the ground, and this fallout process is simply enhanced by rain.
The spike of the radiation dose rate on 20 March is the last one, as one can see in Fig. 1. The termination of the spike-like increase of the radiation level on 20 March is also confirmed by the radiation dose rate data in the other directions from the FNPP (not shown here). The other stations’ data also show the wind-driven spread (spike-like peak) on all days during 12–20 March, i.e. the FNPP area was the continuous source of the radioactive dust until 20 March. After the last major spread of the radioactive dust on 20 March, the radiation dose rate continuously decreased with nearly exponential decay. We note that the rain on 20 March was the first major rain in the entire area after the accident, and that this rain most likely caused the majority of suspended radioactive dust in the air near the FNPP to settle on the ground. This explains the termination of the spread of the radioactive dust because no significant change in the status of the release of the radioactive materials from the FNPP facility was reported on 20 March. This means that a substantial amount of radioactive dust stayed in the air before this rain, i.e. after the earlier rains.

Next, we examine the PG behavior that provides additional information on the dynamics of the radioactive materials. We divide the PG data after the initial drop on 14 March into six different periods as marked by stepped lines that are numbered (1)–(6) in Figs. 1 and 3:

1. Near-zero PG over 28 h around 15 March (Fig. 3a) while the radiation level at Ibaraki-cho at 25 km east of Kakioka (Fig. 3b) showed two spike-like increases at around 03 UT and 21 UT, both of which are related to the wind direction change. The weather was cloudy with nearly a constant humidity except a short period of minor rain (total 1 mm) at around 20 UT, 15 March.

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2. Increase of the PG during 01–07 UT, 16 March to more than 50 V m$^{-1}$, and subsequent settlement to about 20 V m$^{-1}$ (Fig. 3a) while the radiation level at Ibaraki-cho stayed constant (Fig. 3b). Standard deviation of PG (not shown here) has many spikes during this period, indicating the existence of the electrified cloud during the elevated PG. In fact, a weather system passed at the beginning of this episode and mostly cloudy conditions continued. More importantly, this weather system brought first strong wind (>5 m s$^{-1}$ for 1 h average and >10 m s$^{-1}$ for instantaneous peaks) together with the first sunshine (sunshine more than 70 % of the time) after the accident on 14 March, during 02–07 UT at Kasama, 01–08 UT at Shimodate, and 02–08 UT at Tsuchiura. The observed strong wind was from west, i.e. from a large angle to the direction of the FNPP.

3. Low but finite (about 20 V m$^{-1}$) PG with some fluctuations during 16–20 March (Fig. 3c). The radiation level (Fig. 3d) also stayed constant except for a slight increase together with a spike at around 02 UT, 20 March, although no outstanding signature is recognized in the PG data at this time. This spike corresponds to the first
4. Intermittent negative PG during the first strong rain from 22 UT, 20 March to 05 UT, 23 March (Fig. 3e). After the rain, the PG settled to near-zero level without daily variation. The radiation dose rate (Fig. 3f) increased spike-like at around 20 UT on 20 March, two hours prior to the rain, and it settled to a new value at the peak of the rain. This value fluctuated until the end of the rain on 23 March.

5. Recovery of daily variation of PG from 26 March to the middle of April. An example period is shown in Fig. 3g and h. The nighttime fair-weather baseline PG stayed low while minor recoveries of the baseline PG started, reaching to about 10 V m$^{-1}$ by the end of this period. The radiation dose rate continuously decayed with a half-decay time of about 10 days that corresponds to $^{131}$I.
6. Weakening of the daily variation and gradual recovery of nighttime fair-weather baseline after late April, except for few-days episodes of finite PG values (about 20 V m$^{-1}$) during 22–25 April and 11–14 May. The decay rate of the radiation dose rate is much slower than that of the previous period, suggesting that the contamination by $^{131}$I (half-decay time is 8 days) became less significant than those by $^{137}$Cs (half-decay time is 30 year) or $^{134}$Cs (half-decay time is 2 year).

3 Interpretation

Before any interpretation of the data for this single special event, we must consider possible contamination of the measurement system by the radioactive fallout. The entire system is inside a house, a backup system (rotation type) shows very similar data, and leak-tests are made monthly. All indicate that the sensor cannot be contaminated by the radioactive materials. Furthermore, negative spikes during rainy days are as large as a few thousand V m$^{-1}$ even on 22, 26, and 30 March, leading us to believe that the insulation system worked as normal.

During the first period marked by (1) in Fig. 1, the near-zero level PG at Kakioka means that the atmospheric electric conductivity increased by at least one order of magnitude due to the radioactive contamination near the ground (Paper 1). No daily variation was recognized. The stable near-zero PG did not change after the first rain (20 UT, 15 March) of 1 mm or during the spikes of the high radiation level at around 03 UT and 21 UT at both Ibaraki-cho and Mito as shown in Fig. 3a and b. This is somewhat controversial because the value of about 0.2 µGy h$^{-1}$ at both Ibaraki-cho and Mito is only five times larger than that before the accident (peak value is another five times more), and such an increase does not enhance the electric conductivity by five times or more if the increase of the radiation level is the same at Kakioka. In other words, the radioactive contamination at Kakioka was most likely higher than that at Ibaraki-cho during this period. One obvious candidate is extra stagnation of the wind by trees or interception by trees during the dry contamination period. One obvious candidate is extra stagnation of the wind by trees or interception by trees during the dry contamination period. At the first time after the arrival of the radioactive dust. Although the electrified cloud causes large positive PGs for the normal condition, the enhanced electric conductivity (by an order of magnitude during the first period) predicts much smaller PG than the observed value unless the contamination condition did not change. In other words, the contamination level must have decreased during the second period.

One possible explanation is that the sudden increase of the wind speed lifted the majority of the radioactive dust that was stagnant (suspended in the air or sitting on trees) from near the surface (cf. Fig. 4c) and transported it away from Kakioka. This scenario agrees with the observed increase of PG during both the second period (affected by the electrified cloud) and the third period (not affected by electrified cloud on 16 March). It is possible that strong wind lifts a majority of the radioactive material if it is not settled to the ground.

The settlement of the radioactive dust to the ground was observed between the third period and the fourth period (Fig. 3e and f), i.e. during the second rain at Kakioka (22 UT, 20 March to 02 UT, 22 March) right after the arrival of the last radioactive dust plume. The PG settled to near-zero after this rain while the radiation dose rate at Ibaraki-cho and

![Fig. 4. Illustration of the possible contamination forms of the radioactive materials near the ground: (a) migrating subsurface soil, (b) attaching at the top layer of the soil, (c) suspended in the air near the ground.](image-url)
The nighttime near-zero baseline of the PG at Kakioka also slowly recovered during sunny days, but recovery is somewhat interrupted to set back to near zero on 8 and 18 April. One may wonder if these resets are real resets of the PG or simply due to weather. Since 8–11 April and 18–24 April were continuously cloudy or rainy, and since the radiation dose rate constantly decayed without any setback during the entire period, these resets of the baseline to the near-zero level could simply be associated with the cloudy or rainy weather. However, the baseline PG values during clear sky also indicate the reset of the baseline PG: the baseline PG values on 12–14 April (clear sky) are similar to those on 4–5 April (clear sky), while that of 25–28 April (clear sky on 25 April and partially cloudy on 26 and 28 April) is less than these values. Considering that the decay time of free charges in the air-earth system is less than 1 h, the reset of the baseline PG on 8 and 18 April can be real ones due to secondary transport of the radioactive materials.

According to the Chernobyl experience, trees intercept the radioactive fallout quite effectively, causing delayed contamination of the ground (IAEA, 2006, Sect. 3.4). If this process happened at Kakioka during April, one can explain the resets of the baseline PG on 8 and 18 April. Such a new contamination of the ground is not necessarily only from the trees but could originate from the upwind direction or from high altitude. In these scenarios, the minor recovery of the baseline PG can be due to gradual migration to the subsurface or removal by wind during dry periods (cf. the same mechanism as the recovery of the daily variation) or both.

To examine whether there is any substantial transport of the radioactive materials, we compare the radiation dose rate between different stations that are located closely to each other, as shown in Fig. 5. Figure 5 shows 80-day profiles of ratios of radiation dose rate from three stations near Ibaraki-cho compared to the value at Ibaraki-cho. The relative value continuously changed toward unity until around 20 April, indicating a systematic secondary transport of radioactive materials either from high contamination areas to low contamination areas within 15 km or from thick altitude distribution to thin distribution near the ground. After 20 April, the ratio in Fig. 5 stayed rather constant, and this transition date is the same as the transition from the fifth period to the sixth period that is judged from the PG behavior in Fig. 1.

The same type of analyses at 12 stations within 100 km from the FNPP (Yamauchi, 2012) also indicate inter-regional secondary transport of radioactive materials from high contamination areas to low contamination areas until the end of April. Figure 5 also shows a clear change at Hiroura and Onuki on 8 April, on the rainy day. Thus, the offsetting of the Kakioka’s PG can be related to the change in the radiation dose rate, which was not obvious for Ibaraki-cho or Mito.

We next consider the sixth period that is characterized by the disappearance of the daily variation. The PG recovery was no longer interrupted by the rain if we compare sunny days. Both characteristics indicate a settlement of the
radioactive materials to the ground such that the settled radioactive materials are no longer lifted up from the surface by the daily wind even during dry periods, as illustrated in Fig. 4a. The settlement to the ground includes partial migration of radioactive materials to the subsurface (Fig. 4a) because repeated rains help fix the radioactive materials to the soil at both surface and subsurface (IAEA, 2006, Sects. 3.1, 3.3, and 3.5). Once the radioactive materials migrate to subsurface, radiation from there is absorbed by the soil particles, making the radiation dose rate small while ionizing the soil particles (IAEA, 2006, Sects. 4.2, 4.3). The majority of the ionized soil particles most likely stayed on the ground, so that the daily variations during the sixth period are expected to be less than those during the fifth period.

4 Summary and conclusion

The PG data from Kakioka, together with the radiation dose rate from the nearest station (located about 25 km away), show the detailed processes of the transport, migration, and dynamics of the ionizing radioactive materials. We have shown six different periods that represent different forms of the contamination.

(1) The initial drop of the PG to almost zero during 14–15 March is most likely due to radioactive dust suspended in the air near the ground during cloudy weather. (2) An episode of PG increase to more than 50 V m$^{-1}$ on 16 March is most likely due to the re-suspension of the radioactive dust from the surface (ground and trees) to the air and subsequent removal from Kakioka by a first strong wind (>5 m s$^{-1}$ in hourly average) from the non-contaminated area. (3) Low but finite values of the PG during 16–20 March most likely reflect a reduced amount of radioactive material near the ground after the above wind transported away the majority of the suspended radioactive dust. (4) Very low values of the PG after the substantial rain on 20–22 March most likely reflect settlement of the radioactive material by the rain-induced fallout. (5) Temporal recovery of daily variations from the end of March to the middle of April with low nighttime fair-weather baseline PG most likely reflects re-suspension of the radioactive dust into the air from the ground and trees, and subsequent transport to the other region or fallout to the ground until late April. (6) Weakening of the daily variation and gradual recovery of the nighttime fair-weather baseline after the mid-April suggests a complete settlement of the radioactive material to the ground with partial migration to the subsurface.

Thus, the PG observation gives extra information on the dynamics and redistribution of the radioactive materials, which cannot be monitored only by the radiation dose rate. The information on the suspension of the radioactive dust obtained here is particularly important in understanding the internal dose that the local residents have received as the result of the Fukushima nuclear accident. In this report, we did not examine all changes in the weather/wind on the hourly scale, human effects that produce other types of dust, spread of the radioactive materials into the water, nor the possible effect of the ionizing radioactive dust in changing the weather through the electrical effects on cloud microphysics.

Acknowledgements. The atmospheric electricity (PG) data is available to the public through http://www.kakioka-jma.go.jp/cgi-bin/plot/plotSetNN.pl?lang=en. We thank T. Toya for his advice on the PG data. The radiation dose rates are published on the web sites of Ibaraki-cho prefecture (http://www.pref.ibaraki.jp/20110311eq/index2.html). The meteorological data was provided by Japan Meteorological Agency through http://www.jma.go.jp/jma/indexe.html. The relation between the radiation dose rate and the wind...
direction near Ibaraki-cho that was discussed in relation to Fig. 5 can be seen in 10-min resolution movie of 40 stations at http://dl.dropbox.com/u/8076743/eq2011/Radio20110315-31b.mov). The lead author (MY) wishes to thank Sweden programs of providing help for physically disabled individuals which have made it possible for him to conduct this work.

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References


