INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

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Updating of the knowledge relating to the dispersion and deposition of atmospheric releases from the Fukushima Daiichi nuclear accident in March 2011

Since 2011, IRSN has been working on improving the understanding of the Fukushima accident and of its environmental consequences, by coupling the analysis of the measurements in the environment with the modelling of the atmospheric dispersion. This work has been largely carried out as part of international collaborations. For example, the SAKURA[1] project, a bilateral collaboration between IRSN and the MRI[2], gave IRSN access to numerous data (radiological and meteorological measurements, meteorological models) that proved to be decisive. IRSN also participated in the UNSCEAR expert group for the assessment of population exposure levels due to the Fukushima accident [UNSCEAR (2013)][3]. Finally, the Institute participates in international comparison exercises of atmospheric dispersion model regarding the Fukushima accident, coordinated by the Science Council of Japan [SCJ (2014)][4].

Five years after the accident at the Fukushima Daiichi nuclear site, IRSN evaluated the understanding of the behaviour of atmospheric releases and of the main contamination sequences of the terrestrial environment. Many radiological measurements in the environment have made it possible to reconstruct to a great extent the various contamination sequences, to identify the likely trajectories of radioactive plumes and to link them to rain events to explain deposition zones[5]. However, many uncertainties remain, and measurements alone do not allow satisfactorily reconstructing of all of the contamination sequences.

The remainder of this paper outlines the various measurements in the environment. A second part is devoted to the current understanding of contamination events, provided by the cross analysis of radiological and meteorological observation data. In the third part, additional insight provided by modelling is presented. Two additional notes discuss these last two topics in detail:

- Note on the contamination events;
- Note on the modelling of the atmospheric transport and fallout of the releases.

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[1] This Hubert Curien partnership (PHC) is funded by the Foreign Ministry.

[2] The MRI is the research unit of the Japanese meteorological forecasting centre (JMA).

[3] The UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) posted the report online.

[4] The first exercise resulted in a report in September 2014. The second was launched in early 2016.

[5] The term "deposition" means the fallout that has been deposited on the ground, buildings and plants during the passage of the radioactive plume through the atmosphere. When the deposition is made without rain, it is called a "dry deposition". When radionuclides have been dragged down to the ground by rain, it is called a "wet deposition". This process is generally considered to be the most effective.

Measurements in the environment

Many radiological measurements were made public, some as early as March 2011 and others very recently. These are of three types:

1. **Dose rate measurements**[6] are those that were available more quickly during the accident. Fixed measurement stations are well distributed throughout Japan (Figure 1) and give the evolution over time of the contamination. However, the signal is difficult to interpret because it corresponds to radiation emitted by all radionuclides, whether on the ground or airborne, without it being possible to distinguish them. Thus, in locations highly contaminated by deposits, the signal remains very high after the passage of the plume, possibly masking the detection of subsequent events. The dose rate measurements give no information on the isotopic composition of the contamination.
2. **Measurements of the deposits on the ground enable precise mapping of the contamination. They come from airborne measurement campaigns**[7] [Sanada *et al* (2014)] and soil samples [Endo *et al* (2012; Saito *et al* (2015)] taken after the main atmospheric releases. These measurements therefore concern mainly caesium isotopes, ^{134}Cs and ^{137}Cs, with long half-life. Some studies evaluate, from samples, the deposition of radioactive iodine including ^{131}I [Kinoshita *et al* (2011)], whose half-life is short, but these assessments are more uncertain. In addition, the mapping carried out *a posteriori* does not allow distinguishing the periods during which the various deposition zones were formed.
3. **Activity concentration measurements** give direct concentration in the air of an isotope over a given period (in Bq/m^3). These measurements, invaluable for the reconstitution of the plume trajectory in the atmosphere, were scarce until recently, and integrated over long periods of time, making them of little use. In 2015, activity concentration measurements of ^{137}Cs estimated from particulate filters of air quality monitoring stations in Japan were made public [Tsuruta *et al* (2014)]. These measurements, numerous and well distributed throughout Japan, have hourly time increments. Access to these data enabled real progress in the understanding of the sequence of events. However, since the air samples were taken at ground level, they only indicate the presence of plumes that touch the ground; however, it can be located at a high level. In the event of rain, this can lead to soil contamination by wet deposition (leaching of the plume at high altitude), while the activity concentration measurements are very low. Furthermore, these measurements may be underestimated if there is fog. Finally, given that the filter measurements are relatively recent, there is no available information regarding short-lived radionuclides. The location of the activity concentration stations is shown in Figure 1.

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[6] The dose rate is the gamma radiation emitted by all radioactive isotopes at a given point per unit time. Its unit is Gy/h.

[7] The results of the campaigns are listed on the NRA (Nuclear Regulation Agency) website.

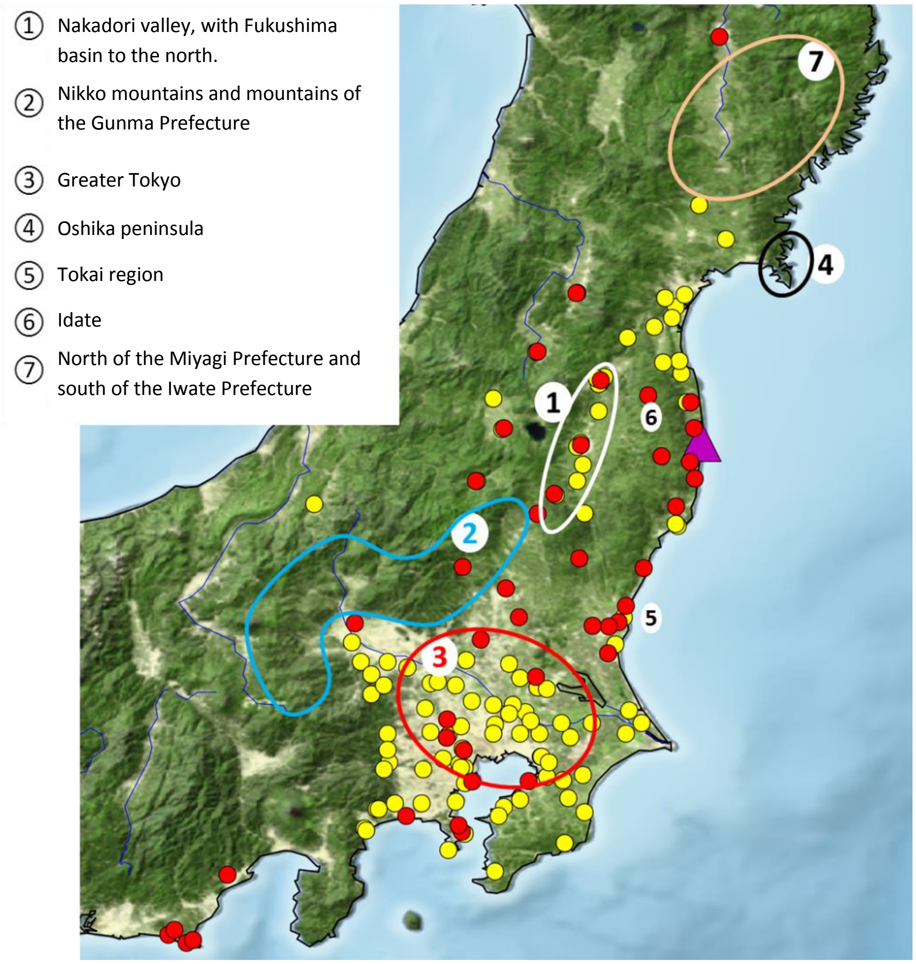

Figure 1: locations of the dose rate (red) and activity concentration (yellow) beacons are represented on a relief map of Japan. Regions whose importance is mentioned in the text are also located on the map.

With regard to meteorological observations, IRSN had access, as part of the SAKURA project, to the JMA AMEDAS measurement network data[8].These include:

- Wind observations (speed and direction) at 10 meters (around 1,200 stations)
- Rain observations given by pluviometers. These give the amount of rainfall in mm/h in discrete 0.5 mm/h increments. Light rain (less than 0.5 mm/h) was thus not measured. However, some more sophisticated pluviometers indicate the occurrence of light rain without giving the amount;
- Temperature, humidity, visibility and snow depth observations, which were used for example to determine the possible presence of snow or fog. This information is important for estimating depositions.

In addition to the AMEDAS data, rainfall observations made by radar[9] (corrected by the pluviometers) were used. These give spatial information at a very fine resolution (1km), but do not allow light rain to be identified either.

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[8] Automated Meteorological Data Acquisition System (http://www.jma.go.jp/en/amedas/)

[9] http://www.jma.go.jp/en/radnowc/

Contamination of Honshu Island

The major releases into the atmosphere took place over three weeks between 11 March and 31 March 2011. During this period, part of the Japanese territory was contaminated. Several studies based on modelling estimate that around 80% of the releases into the atmosphere were deposited over the Pacific Ocean and approximately 15-20% on Honshu, the main island of Japan [Morino *et al* (2011), Morino *et al* (2013), Yasunari *et al* (2011)]. Assessments by IRSN corroborate these estimates [Korsakissok *et al* (2013) ; Groëll *et al* (2014)]. The observations indicate that the surface area of Japan contaminated with more than 185 kBq/m² of ^{137} Cs [10] covers about 1700 km² [Steinhauser *et al* (2014)] and the surface area contaminated with over 10 kBq/m² covers about 24,000 km [Champion *et al* (2013)]

In 2012, IRSN presented a description of the main atmospheric release events and their future [IRSN (2012)]. In light of the new measurements available, particularly as part of the SAKURA project, the Institute has re-analysed the various sequences.

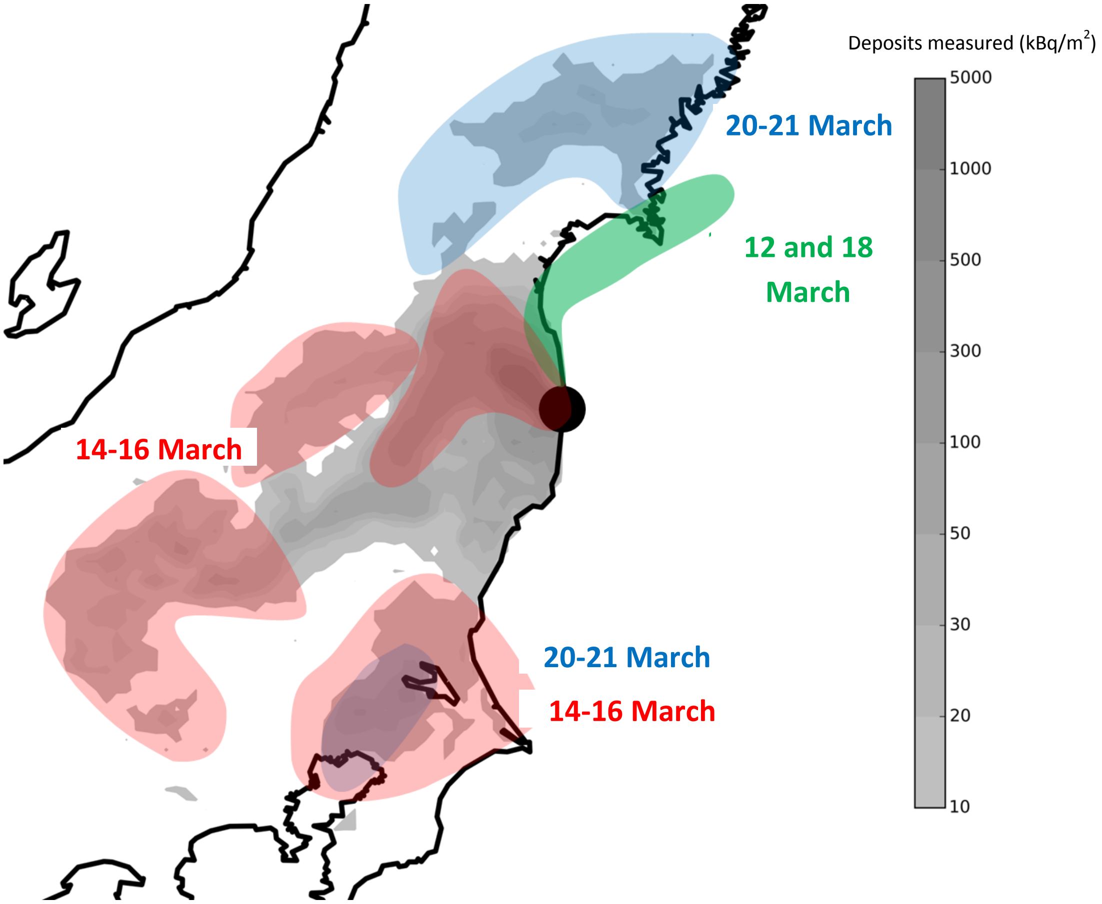

Figure 2: Regions affected by the fallout of the main contamination episodes are indicated in colour and superimposed on the map of measured deposits of 137 Cs. The episodes of 12 and 18 March are in green, the episode of 14-16 March is in pink and that of 20-21 March is in blue.

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[10] The value of 185 kBq/m² corresponds to 5 Ci/km², which was the threshold value defining the evacuated areas following the Chernobyl accident. After Chernobyl, the surface area contaminated with more than 185 kBq/m² of ^{137} Cs was approximately 29,400 km².

The ground contamination of the Japanese territory primarily occurred in four time slots: that of 12 March, that of 14-16 March, that of 18 March and that of 20-21 March (Figure 2).

The 12 and 18 March episodes did not lead to significant depositions (Figure 2), but the activity concentration measurements at stations H, I and J show that they were significant from the point of view of the population exposure to radioactive plumes in the area located to the north of the power plant, along the coast of the Fukushima Prefecture (Figure 3). Farther north, the Oshika Peninsula (Zone 4 Figure 1) shows ^{137} Cs deposits that formed during these episodes. These are dry depositions.

The 14 to 16 March episode is the most studied event of the accident sequence because it was responsible for the main depositions on the island of Honshu. During this event, the winds shifted gradually pushing the contamination to the Southwest, West, Northwest and then the South along the coast. As from 6:00 am on 15 March[11], the plumes present in the Nakadori valley (Zone 1 Figure 1, Stations A to G) and between the power plant and the Fukushima basin (Station A), were leached, causing most of the deposits observed in these areas. The analysis of the observations tends to reject the hypothesis, long raised by several Japanese teams, of depositions by the fog in the valley Nakadori. The most significant deposits occurred with the early rainfall, some of which were too weak to be measured in some locations. The analysis of the observations seems to suggest that a significant part of the activity in the air was located at a high altitude, probably because of the relief of the regions crossed. Figure 3 shows that the portion of the activity measured in the air from 14th to the 16th March in the Fukushima Basin (Station A) is lower than for the 20-21 March episode. However, deposits were mainly formed on the 15th of March. Further south, the deposits on the side of the Nikko Mountains and on the mountains of Gunma Prefecture (Zone 2 Figure 1) probably occurred during this episode. The measurements are too incomplete to study the sequence of contamination, but the assumption of a deposition caused by a locally created fog or by clouds caught in the mountains is very likely [Kaneyasu *et al* (2012) ; Hososhima et Kaneyasu (2015)].

The 20 to 21 March episode is the main contamination episode of the Kanto plain, the Tokai region and the area between the north of the Miyagi Prefecture and south of the Iwate prefecture.

Between the 20th and the 21st of March, several plumes were detected to the North West of the power plant (the area of Stations J, I, H and beyond), in the Nakadori Valley (Stations A to G) and in Greater Tokyo. Light winds favoured their stagnation, which led, overall, to a high ^{137}Cs activity concentration, over the period between the 20th and the 21st of March. Based on soil contamination and dose rates, the event of the period between the 14th and the 16th of March has been up until now considered to be the episode that led to one of the greatest exposures of the population of the Fukushima basin, the Nakadori valley and Greater Tokyo. Measurements of the activity concentration in ^{137} Cs challenge this idea by indicating a non-negligible exposure to the plume during the episode of the 20-21 March (Figure 3). However, measurements by radionuclide indicate that the proportion of iodine, tellurium and of rare gases contained in the plume was lower on those dates than during previous episodes. Yet it is these elements (especially iodine and tellurium) that are the most critical regarding health impact during the passage of the plume.

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[11] All times are given in Universal Time (UTC). 9 hours must be added to obtain the local Japanese time.

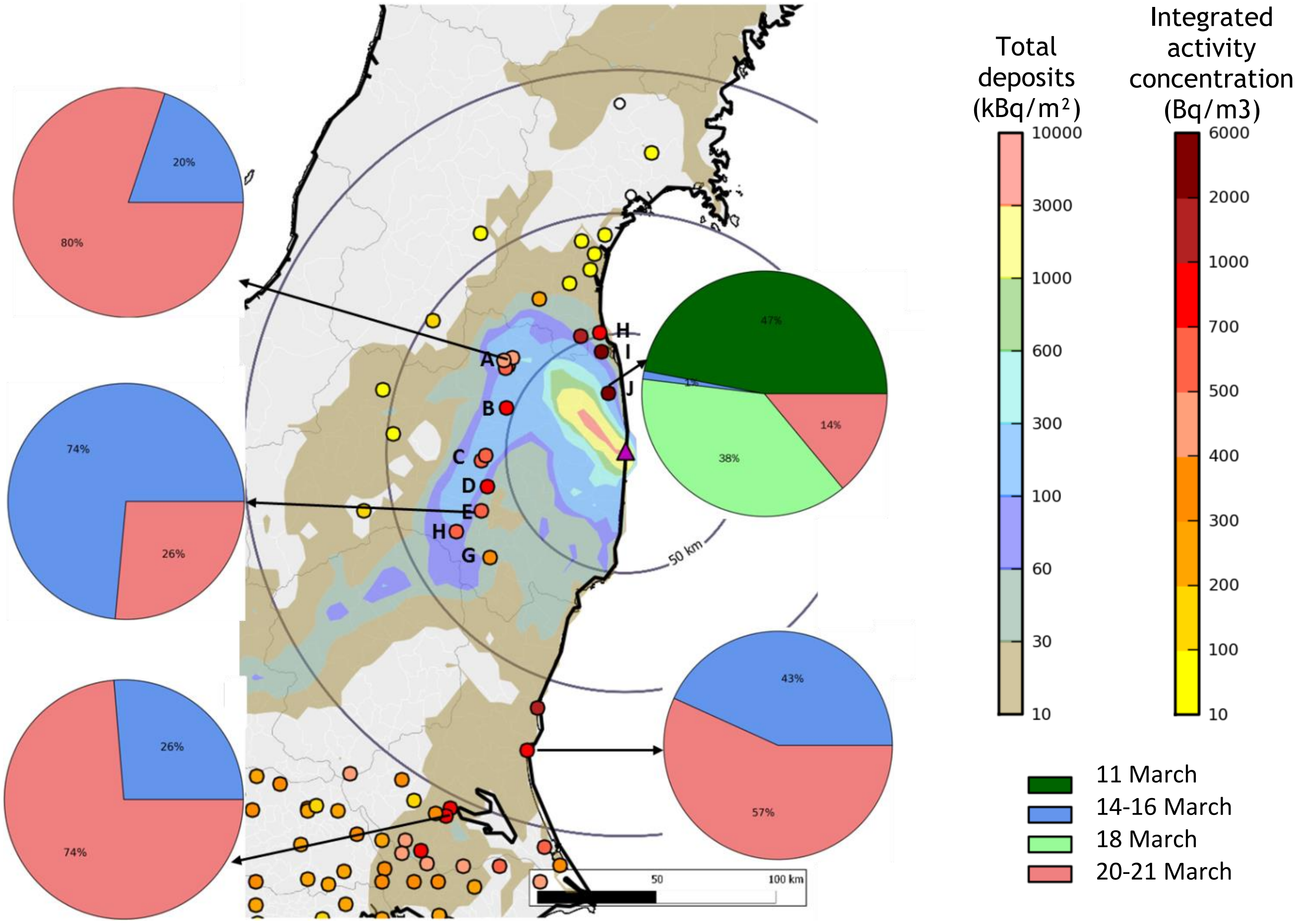

Figure 3: The coloured dots represent the values of the 137 Cs activity concentrations at the stations integrated between 11 March and 24 March. They are superimposed on the map of total 137 Cs deposits. The pie charts show the distribution of the activity concentrations observed for each of the main contamination periods.

To summarize, the joint analysis of the radiological measurements in the environment and the meteorological observations have made it possible to determine the path of the plume through the atmosphere and the periods of leaching by rain, leading to the main soil contamination episodes. The analysis of the activity concentrations revealed areas that were subjected to a significant atmospheric contamination by ^{137}Cs, while the deposits measured are low and the dose rates show no significant increase (Figure 3).

The analysis of the measurements highlighted the main challenges limiting the understanding of the events.

- The complex orography (influence of the coast and reliefs) played a crucial role in the path taken by the plume, but also in its vertical rising. It also affected the distribution of the depositions. When the plume was at a high altitude, no measurement could detect it (the stations are on the ground). However, several plumes were probably carried by high altitude winds, to be subsequently leached by rain and form contamination patches that cannot be explained otherwise.

- The deposition cannot be satisfactorily explained by taking into account only rain observations (pluviometers or radars). Indeed, light rain (less than 0.5 mm/h) is not measured. Analysis of the measurements shows that the main ground contamination, on 15-16 March, is linked to plume leaching having started earlier than the first rain observations, thus consequent to light rain.
- No measurement provides access to the isotopic composition of the plumes and deposits. Measurements of ^{129}I have started to be published and make it possible to deduce the ^{131}I content [Muramatsu *et al* (2015)].

Transport and fallout modelling

Since 2011, the international community of atmospheric dispersion modelling has been working on simulating as closely as possible the environmental consequences of the Fukushima accident. The motivation is twofold. The first objective is, of course, to achieve a better understanding of the accident. The second is to improve dispersion models and the representation of physical processes. IRSN extends this second objective to the operational tools that it develops for the assessment of radiological consequences. This objective responds to the need to improve the relevance of the Institute's expertise, in the event of a nuclear accident.

First objective: to precisely simulate and better understand the accident

Fairly quickly after the accident, the main release events were identified and their consequences were roughly assessed. Since then, the understanding of the various episodes has been greatly improved, the processes responsible for the depositions have been determined, and simulations have become more realistic. The most significant progress has been the result of efforts focused on improving the main input data of dispersion models. These are the weather conditions and the quantification of the releases to the atmosphere.

The weather conditions are provided by forecasting models. They determine the transport of the radioactive plume through the atmosphere. Rainfall is responsible for soil contamination, due to leaching of the plume. Spatial or temporal shifts in the forecast, such as a delay of a few hours in the change of wind direction, or the onset of precipitation, can strongly penalise the reproduction of some contamination episodes. After the accident, simulations were made using weather forecasts with too low resolution as shown, in 2012, by IRSN [Mathieu *et al* (2012), Korsakissok *et al* (2013)]. Forecasts with a finer resolution [WMO (2011), Sekiyama *et al* (2015)] were thus produced, to significantly improve the Fukushima accident simulations. IRSN had access to many forecast sources, especially as part of its participation in the UNSCEAR and SAKURA project work. Radar rainfall observations were also used in the simulations, replacing the precipitations forecast by the models.

The source term, that is to say, the temporal evolution of the rate of each radionuclide released into the atmosphere, is the other essential input data of atmospheric dispersion models. Currently, there is no detailed source term estimated only through modelling the changes in the state of the reactors. The existing ones were all assessed based on methods using environmental measurements and atmospheric dispersion simulations. Several source terms have been published. The first were relatively rough. They have since been extensively refined. However, there is no consensus on a source term considered to be more realistic than the other. The contribution of IRSN to the detailed assessment of the accident releases is significant and original [Winiarek *et al* (2012) ; Saunier *et al* (2013) ; Winiarek *et al* (2014)].

IRSN was the first team to develop an automatic method for reconstructing the source term from measurements of the dose rates in the environment.

The detailed source terms and fine scale weather forecasts have made it possible to realistically simulate the transport and fallout of the plumes. The comparison with observations was significantly improved. However, some episodes remain difficult to simulate. For example, simulations of the sequence between the 14th and 16th of March are still unable to reproduce the various deposition events. This fact reflects the remaining uncertainties and the complexity of the situation encountered. These difficulties have oriented the progress areas that should be favoured to improve dispersion models.

Second objective: to improve the dispersion models and crisis management tools

The improvements of the atmospheric dispersion models favoured by the community working on the Fukushima accident relate to the representation of the deposition process. Indeed, regarding population exposure, modelling the deposition is a major challenge, both due to the doses induced by deposits and to the depletion of the plume that it generates. The studies carried out, in particular by IRSN [Quérel *et al* (2016)], show that the remaining uncertainties with regard to the input data in the case of Fukushima are still too large to study the deposition process. Their precise modelling therefore does not provide, at this stage, any real added value to improve the realism of the simulations. The subjects of the vertical distribution of plumes and the deposition during light rain episodes are considered to be a prior necessity before a better simulation of soil contamination can be envisaged. Further analysis of the accident from the perspective of environmental measurements and inter-comparisons of models such as those organised by the SCJ are two means available to the community to continue to make progress on these issues and improve the simulation of the accident.

Beyond working on dispersion models, modelling the Fukushima accident has demonstrated the value of having operational tools to estimate detailed source terms from environmental measurements. IRSN has been quite innovative in this regard. Now, all operating-oriented teams are seeking to develop such a tool (European Project PREPARE, PHE and Met Office[12] teams for England, ZAMG[13] for Austria).

The great lesson learned from the Fukushima accident concerns the uncertainties related to the weather data and releases. These are inevitable and the major challenge for the consequence modelling community is to successfully take them into account in the management of an accident situation. Assessing their impact on the simulations and modelling them for more relevant decision making in view of the risk of exposure constitutes an important challenge. Since 2011, IRSN initiated a project on this theme [Girard *et al* (2014) ; Girard *et al* (2015)]. At the European level, other teams are also moving in this direction [Sørensen *et al* (2015)].

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[12] PHE is Public Health England and MetOffice is the English weather centre.

[13] The ZAMG is the Central Institute for Meteorology and Geodynamics.

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