Part I

Transport of Radioactive Materials in the Environment

Part I presents scientific knowledge related to environmental contamination by radioactive materials emitted from the accident at the Fukushima Daiichi Nuclear Power Station of the Tokyo Electric Power Company. Chapter 1 summarises basic concepts for understanding the event. Chapter 2 estimates the amount of emitted radioactive materials. Chapters 3 through 6 describe the transport of radioactive materials in the regional atmosphere, global atmosphere, ocean and land areas.

1

Introduction

Basic Concepts Regarding the Fukushima Accident and Radiation and Radioactivity

This chapter presents an overview of the accident and basic scientific concepts that are important for understanding the migration processes of radioactive materials in the environment.

1.1 Overview of the Fukushima Accident

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The accident of the Fukushima Daiichi (First) Nuclear Power Station (FDNPS) of the Tokyo Electric Power Company (hereafter, Fukushima accident) transpired after the Tohoku Region Pacific Coast Earthquake occurred in March 2011. Table 1.1 summarises the main events of the accident. After the earthquake occurred at 14:46 on 11 March 2011, tsunami waves of 13 m in height arrived at the FDNPS (TEPCO, 2011); the diesel power engine stopped at 15:41. Due to this electricity loss, the nuclear reaction became uncontrollable. The Fukushima Daini (Second) Power Station was able to make a controlled stop for cooling even after the intrusion of seawater from a tsunami wave with a height of 9 m. The estimated maximum height in the design of the Daiichi and Daini Power Stations was 5.1 m. In contrast, the estimated maximum tsunami height in the design of the Onagawa Nuclear Power Station of the Tohoku Electric Power Company, which avoided serious damage, was 14.8 m (Matsumoto, 2007).

A hydrogen explosion occurred at the first power unit of the FDNPS at 15:36 on 12 March and in the third power unit at 11:01 on 14 March. Radioactive materials were emitted in large quantities into the atmosphere due to these events. It is suggested from monitoring data that there were several other emission sources, such as water vapour release by vent operations in the reactor pressure vessel and direct discharge of leaked high-level contaminated cooling water from the reactor into the ocean.

Table 1.1 *Main events associated with the Fukushima accident in 2011, based on government reports (NERH, 2011a; 2011b; NSC, 2012; CSIC, 2012)*

11 March
14:46: Tohoku Region Pacific Coast Earthquake
~15:30: Tsunami wave arrives at the FDNPS
15:42: Electrical power lost at the FDNPS; special measures concerning nuclear emergency preparedness enacted; Article 10 issued by TEPCO and related organisations
16:45: Water injection into the reactor is not possible; Nuclear Disaster Special Measures Law Article 15 is issued by TEPCO and related organisations
19:03: Emergency declaration of nuclear disaster countermeasures; headquarters for the FDNPS implemented by the prime minister
20:50: Fukushima prefecture governor requests the evacuation of Okuma Town and Futaba Town within 2 km distance of the FDNPS
21:23: Evacuation of residents within 3 km of the FDNPS instructed by the Nuclear Disaster Headquarters, with a sheltering distance of 10 km
12 March
5:44: Instruction of evacuation to residents within 10 km of the FDNPS by the Nuclear Disaster Headquarters
15:36: Hydrogen explosion in the FDNPS first power unit
18:35: Instruction of evacuation to residents within 20 km of the FDNPS by the Nuclear Disaster Headquarters
20:20: Seawater injection into the FDNPS first power unit
14 March
11:01: Hydrogen explosion of the FDNPS third power unit
15 March
00:00: Dry vent operation of the FDNPS second power unit
11:00: Shelter instructions to residents within 20–30 km of the FDNPS by the Nuclear Disaster Headquarters; the Japan Coast Guard sets the navigation danger zone off the coast of the FDNPS; Fukushima Prefecture Fishermen's Union Federation consults with the Fukushima Prefecture Agriculture, Forestry and Fisheries Department, resulting in self-restraint operations
17 March
The Ministry of Health, Labour and Welfare notifies local governments to carry out regulations pertaining to the Food Sanitation Law Article 6 No. 2 so as not to use food that exceeds the provisional regulation value of 'the food intake restriction index value' indicated in the 'Disaster prevention measures of nuclear facilities' of the Nuclear Safety Commission
17–19 March
A radiation measurement survey is carried out by US military aircraft of the US Department of Energy (DOE)
21 March
Instructions issued on shipping restrictions of certain foods by the Nuclear Emergency Response Headquarters
22 March
Reports are made of detection of radioactive iodine and caesium exceeding regulation levels from seawater near the nuclear power plant's southern outlets outside the peripheral monitoring area defined by TEPCO

Table 1.1 (*cont.*)

23 March The marine radioactivity survey based on the sea monitoring plan begins The results of the first SPEEDI calculations by the Nuclear Safety Commission are published
25 March Promotion of the voluntary evacuation of residents within 20–30 km of the FDNPS by the Nuclear Emergency Response Headquarters
5 April Radioactive iodine exceeding the provisional regulation values of radioactive substances in food from sand lance (Kounago) obtained off the coast of Ibaraki Prefecture are detected
6–29 April Airborne monitoring is first carried out by the US Department of Energy
13 April There is a joint planning meeting for soil investigation by the Safety Confirmation Project and Earth Science Project
14 April Marine observations are carried out by the oceanographic research vessel *Mirai* on cruise MR11-03
25 April Daily publication of the SPEEDI diffusion simulation results by the Nuclear Safety Commission begins
18–26 May The second airborne monitoring campaign by MEXT and the US Department of Energy takes place
31 May to 2 July The third airborne monitoring campaign by MEXT takes place
6–14 June The first investigation campaign of the radioactive material distribution by MEXT begins, conducting soil measurements and research into the atmosphere and rivers within 2 km of FDNPS
13 June Start of operation of the circulating seawater purification system for radioactive material
27 June to 8 July The second investigation campaign by MEXT investigating radioactive material distribution begins
19 July The Ministry of Agriculture, Forestry and Fisheries begins the national survey for the radioactive substance contamination of rice straw
10 August Operation of the circulation cooling system for all fuel pools of the FDNPS begins
25 October to 5 November The fourth airborne monitoring campaign after a typhoon by MEXT takes place
16 December Completion of step two of ‘Path to convergence of the accident’ (cold shutdown state, controlled radioactive material release and others), as announced by the Nuclear Disaster Headquarters

The total amount of atmospheric emissions of radioactive materials were estimated to be 9–37 (mean and RMSD: 17 ± 8) PBq (PBq = 10^{15} Bq) for radioactive caesium ^{137}Cs (Stohl *et al.*, 2012; Terada *et al.*, 2012; SCJ, 2014; Katata *et al.*, 2015). The large uncertainty in the estimated value is related to various causes, such as data loss from monitoring posts and weather stations due to the tsunamis and power outages, and a lack of observations over the transported area of the Pacific Ocean. The amount of direct discharge into the ocean was estimated to be 3–6 PBq (Kawamura *et al.*, 2011; Estournel *et al.*, 2012; JAEA, 2012; Tsumune *et al.*, 2012, 2013; Miyazawa *et al.*, 2013; Aoyama *et al.*, 2016).

The spring wind regime at the time of the accident was that of a winter-like regime with stronger-than-normal winds that transported large amounts of the emitted radioactive materials in the atmosphere to the ocean region (Takemura *et al.*, 2011), with only 12–37% being deposited onto the Japanese land area (SCJ, 2014). These radioactive materials were transported by winds associated with various pressure systems and deposited on the land and ocean surface in a complex distribution (e.g. see Figure 3.1, which shows that an area of elevated ^{137}Cs contamination exceeding 1000 kBq/m^2 was distributed beyond 30 km around the FDNPS in the northwest direction). This distribution pattern closely overlaps the total accumulation distribution in March to April as simulated by SPEEDI system (see Chapter 8 for details). The SPEEDI simulation results were made available to the public every day after 25 April 2011, which was more than one month after the accident.

The government set the Evacuation Order Zone (20 km) and the Indoor Evacuation Zone (20–30 km) until 15 March, and defined the Planned Evacuation Zone in the northwestern area, which included Kawamata Town, Iitate Village and Katsurao Village, until 22 April. During this period, people evacuated in these areas were considered to have received radioactive exposure. There were also shipping restrictions for agricultural and livestock products, and occurrences of rice straw and gravel contamination. The number of displaced persons in the areas inside and outside Fukushima Prefecture reached 160,000 in 2012 (Fukushima Prefectural Government, 2018). Large land use changes in Fukushima areas after 2011 have been reported, detected by high-resolution satellite observations as shown in Figure 1.1 (Ishihara and Tadono, 2017); it is highly possible that this can be attributed to the disaster comprising the large earthquake, tsunami, and Fukushima accident.

Furthermore, 98% of the deposited radioactive materials on soil were adsorbed by the 5 cm surface layer (Kato *et al.*, 2012a; Takahashi *et al.*, 2015), while the portion deposited on the forest remained in the canopy for a long time. The mean lifetime of the radioactive material in the forest canopy reached 200 days following the Chernobyl event, while there were reports of the material remaining for more

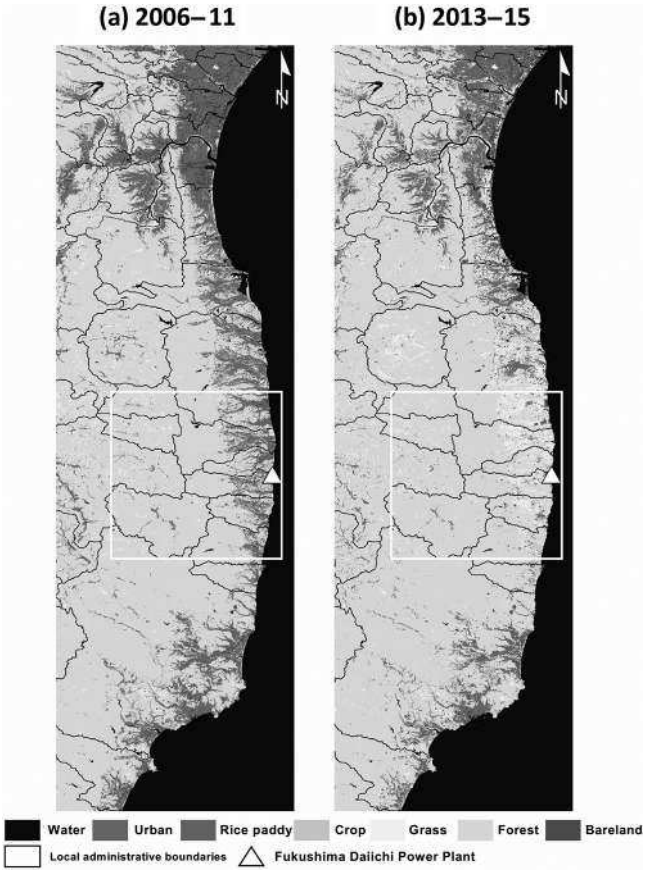

Figure 1.1 Land use changes before and after 2011 by (a) ALOS AVNIR-2 observation in the years 2006–11 and (b) Landsat-8 OLI in 2013–15 as found by Ishihara and Tadono (2017).
Provided by JAXA. A black and white version of this figure will appear in some formats. For the colour version, refer to the plate section.

than 300 days following the Fukushima event (Kato *et al.*, 2012b), with differences depending on the type of forests (Imamura *et al.*, 2017; Kato *et al.*, 2017). These radioactive materials migrate in the environment and flow into lakes and rivers and finally are carried to the sea unless they undergo decontamination. The third and fourth MEXT airborne monitoring campaigns of the radiation dose rate (May to July 2011 and October to November 2011) indicated that the concentration decreased in the 80 km inland area of the Abukuma Mountains, whereas that in rivers and coasts increased, suggesting that migration and river inflow of the radioactive materials had occurred over a long period (MEXT, 2011; NRA, 2013). A MEXT river measurement of solid suspended particles indicated a particle concentration of 55 kBq/kg ^{137}Cs in the main stream of the Abukuma

River. A riverbed ^{137}Cs concentration of up to 16 kBq/kg was also observed, with a good linear correlation with the suspended particle concentration, indicating that large amounts of radioactive materials remained in the riverbed. Moreover, a good linear correlation between suspended ^{137}Cs and the riverbed concentration was observed. Continued outflow of radioactive materials is expected from paddy fields (Yoshimura *et al.*, 2015) and organic materials from forests.

There were two main pathways to the ocean: deposition of the atmospheric radioactive materials ejected by the explosions and direct discharge of polluted water from the FDNPS. Shipboard measurements from research and volunteer vessels after early April 2011 observed radioactive caesium (^{134}Cs and ^{137}Cs) in the surface water over a wide area of the North Pacific Ocean. The ^{137}Cs concentration reached 196 Bq/m^3 in some areas, which is two orders of magnitude higher than that in the surrounding ocean areas (Aoyama *et al.*, 2012, 2016). Radioactive caesium was detected from the suspended materials and zooplankton sampled in a North Pacific area (47°N and 167°E) 2300 km from the FDNPS (Honda *et al.*, 2012). A shipping restriction on seafood was issued after a concentration exceeding the regulation level (100 Bq/kg) was detected in June and August 2012 from Pacific cod at the Hachinohe port (NERH, 2012). In August 2012, a radioactive caesium level of 25 kBq/kg was detected from greenlings caught 1 km offshore of the Ohta River mouth in Fukushima Prefecture (TEPCO, 2012).

A full analysis of the Fukushima accident requires investigation in many fields, such as reactor phenomena, health impacts and societal impacts (SCJ, 2012); here, we focus on environmental aspects during the first year following the event.

1.2 Radioactive Elements, Radioactive Nuclides and Radioactive Substances

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An element is the minimum unit of tangible components comprising a substance. As of 2019, there were 118 elements listed in the Periodic Table, and all elements were given formal nomenclature by the IUPAC (International Union of Pure and Applied Chemistry). Two-thirds of the elements are stable, and the remainder are so-called radioactive elements. Some elements have several isotopes that have different mass numbers, with each isotope being called a nuclide. Over 90% of nuclides known today are radioactive. Only a few of these radioactive elements and radioactive nuclides exist in the natural environment.

One of the major elements comprising organisms such as humans is carbon. In living organisms, there are three types of carbon nuclides (^{12}C, ^{13}C and ^{14}C) that