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Review

Radioiodine in the atmosphere after the Fukushima Dai-ichi nuclear accident

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ABSTRACT

About 160 PBq of ^{131}I was released into the atmosphere during the accident at the Fukushima Dai-ichi Nuclear Power Plant. The chemistry of radioiodine is complicated, and it can be released in several different forms. In addition, the different physical forms, like molecular iodine, aerosol-form iodine, or organic iodine, would have all behaved differently once in the atmosphere, and would have been removed at different rates. These releases were detected by monitoring stations throughout Japan, and from these measurements, key insights can be made about the different chemical forms that were released, as well as the persistence of each in the environment.

Contents

1. Introduction

During the accident at the Fukushima Dai-ichi Nuclear Power Plant, approximately 160 PBq (1.6×10^{17} Bq) of ^{131}I, in addition to 18 PBq and 15 PBq (1.8×10^{16} Bq and 1.5×10^{16} Bq), respectively, of ^{134}Cs and ^{137}Cs, as well as other radionuclides, were emitted into the atmosphere (MEXT, 2011). This prompted large evacuations near the vicinity of the plant, compounding an already devastating earthquake and tsunami.

In terms of the short term public health risk, the radioiodine emissions are of most concern during a nuclear accident. Radioiodine emissions, however, are particularly complex due to the variety of chemical forms that radioiodine can take. Radioiodine in the atmosphere can exist in either gaseous or aerosol form, and can

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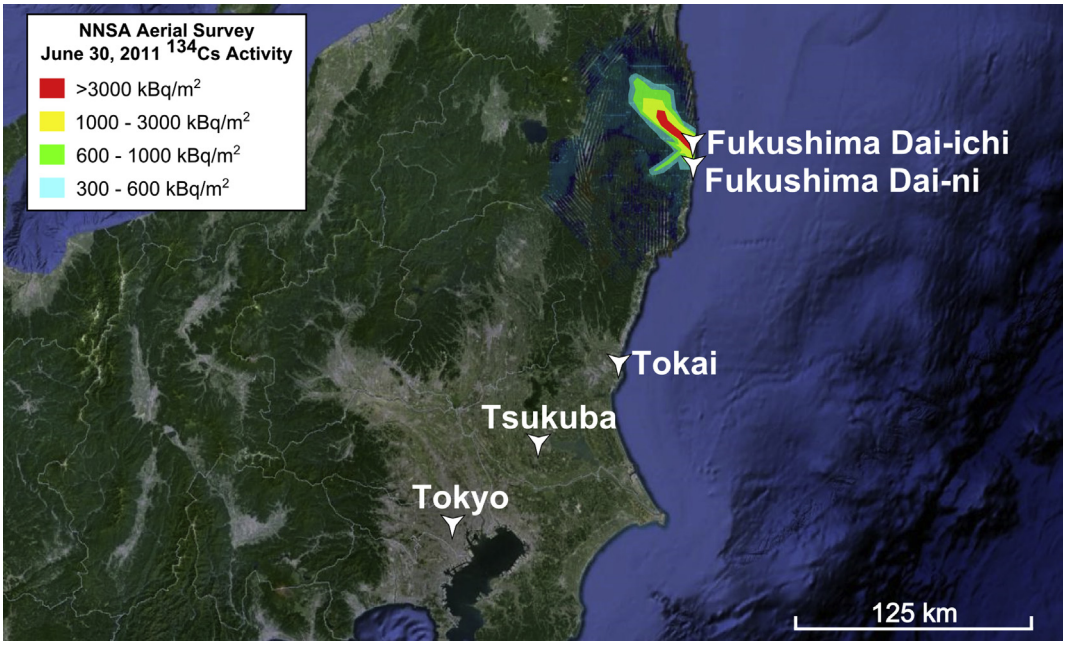

Fig. 1. Contamination map around the damaged Fukushima Dai-ichi reactors as of June 30, 2011 (data from NNSA aerial survey (NNSA, 2011)).

bind to different types of soil and vegetation, or deposit in lakes, rivers, and oceans. Prior even to being emitted to the atmosphere, the composition of different radioiodine species depend strongly on conditions in the core of the damaged reactor, the heat transport system, and the surrounding containment structure.

The conditions in each of the different power plant systems would have strongly affected the chemistry of radioiodine, and therefore the nature of the releases to the atmosphere. This paper reviews a number of the different radionuclide monitoring measurements that were made in the aftermath of the Fukushima accident, and uses this information, first to make some conclusions about the environmental behaviour of radioiodine, and with those, make some inferences about the chemical nature of the releases.

After the earthquake struck the Fukushima Dai-ichi plant at 14:46 on March 11, 2011, the three operating reactors, units 1, 2, and 3, immediately shut down (units 4, 5, and 6 were offline and not operating at the time). The earthquake damaged the electrical transmission lines leading to the plant, and off-site power was cut off as a result. In addition to this, the diesel backup generators at the plant were disabled when the tsunami struck at 15:39. With only about 8 h of DC electrical power available from battery banks, the station quickly lost the ability to cool the reactors.

The remainder of the accident at the TEPCO-owned plant has been well documented (Ohnishi, 2012; Butler, 2011; Chino et al., 2011; Kobayashi et al., 2013; Schwantes et al., 2012). Hydrogen explosions occurred in the reactor buildings of unit 1 (March 12 at 15:36), unit 3 (March 14 at 11:01), and unit 4 (March 16 at 05:45), destroying large parts of the outer superstructures. Large quantities of radioactivity were emitted from the plant, which ended up contaminating hundreds of square kilometers around the plant, as shown in Fig. 1.

Table 1
Largest emissions among individual radionuclides.

Radionuclide	^{133}Xe	^{131}I	^{132}Te	^{133}I	^{134}Cs	^{137}Cs
Released activity	11 EBq	160 PBq	88 PBq	42 PBq	18 PBq	15 PBq
Half life	5.2 d	8.0 d	78 h	21 h	2.1 y	30 y

Although estimates for radioactivity releases vary, it is generally accepted that ^{133}Xe, ^{131}I, ^{132}Te, ^{133}I, ^{134}Cs, and ^{137}Cs were the six radionuclides that were released in the largest quantities. Official estimates from the Japanese Government (MEXT, 2011) are shown in Table 1. Most of the releases occurred in three main phases, with the first being between March 14 and 16, the second being about a week long, between March 19 and 26, and the third, smallest release being between March 29 and 31 (Chino et al., 2011).

Adachi et al. (2013) collected aerosol samples over the course of the accident from their facility in Tsukuba (about 170 km south of Fukushima Dai-ichi), and found that radiocesium-bearing particles from the first plume were spherical and around 2 μm in size. They were also insoluble in water, and coexisted with fairly significant amounts of iron and zinc, suggesting that they were either metallic or composed of metal oxides. By contrast, the radiocesium-bearing particles collected from subsequent times were found to be hydrophilic and to coexist with aluminosilicate and sulphate minerals, suggesting that they were emitted in a chemical form that can more easily interact with water vapour and naturally occurring aerosols in the atmosphere.

Doi et al. (2013) also sampled airborne radionuclides in the wake of the Fukushima Dai-ichi accident, again from Tsukuba, and in two different ways. In measurements employing a quartz fibre filter in series with an activated carbon filter, the gas phase iodine collected on the charcoal filter made up about 40%–80% of the total ^{131}I observed in the three months following the accident, and generally made up a larger fraction of the total as time went on. A second set of measurements employed a cascade impactor, and were taken between April 4 and April 21. These showed that, although a significant amount of radioiodine still existed in the gas phase, its particulate form had activity median diameters of around 0.7 μm. Interestingly, the particle size distribution of the ^{131}I bearing particles were not precisely correlated with ^{134}Cs and ^{137}Cs particles; the latter were slightly larger on average with activity median diameters measured between 1.0 and 1.8 μm.

A third group from Tsukuba, Kaneyasu et al. (2012), carried out airborne radionuclide measurements at a later time, between April 28 and May 26, with a cascade impactor, and the detailed chemical analyses they performed on aerosols in different size ranges