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Depositional fluxes and residence time of atmospheric radioiodine (^{131}I) from the Fukushima accident

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ABSTRACT

Activities of radioiodine (^{131}I) along with ^{210}Pb and ^{210}Po in time series precipitation samples were measured to determine the depositional fluxes of ^{131}I in the Southern United States and its removal rate and residence time in the atmosphere during the Fukushima nuclear accident. Radioiodine released from the Fukushima accident reached the Southern United States within 11 days, giving rise to a concurrent ^{131}I peak and anomalous ^{210}Po/^{210}Pb ratios in the precipitation samples. The cumulative ^{131}I depositional flux was 4.6 ± 0.2 Bq m^{-2} during the maximum fallout. The removal rate of ^{131}I out of the atmosphere, derived from a definite ^{131}I integral model, ranged from 0.03 to 0.14 d^{-1} with an average of 0.08 ± 0.02 d^{-1}, which corresponds to a residence time of ^{131}I in the atmosphere of 12 ± 3 days, consistent with the resident timescale constrained by the ^{210}Po/^{210}Pb disequilibrium technique. These results support our hypothesis that radioiodine was removed from the atmosphere by precipitation within two weeks. It seemed that regions reachable by ^{131}I transport within two weeks from Fukushima Japan would receive much more fallout, whereas places outside that distance would be relatively less polluted with radionuclides.

1. Introduction

The Fukushima Dai-ichi nuclear power plant explosions during March 2011 was rated at International Nuclear and Radiological Event Level 7 (IAEA, 2011), and had emitted vast quantities of radioactive materials into the environment (Brumfiel and Cyranoski, 2011). Among the radionuclides derived from the nuclear accident, radioiodine (^{131}I) and cesium (Cs) including ^{134}Cs and ^{137}Cs are of special concern to people because they could trigger dangerous health effects as the case in the Chernobyl accident (Balter, 1996; Bleuer et al., 1997). About 150 peta-Bq (10^{15} Bq) of ^{131}I was emitted into the atmosphere during the Fukushima disaster (Nuclear and Industrial Safety Agency (NISA) Report, 2011), which dispersed across the globe. Indeed, elevated ^{131}I has been measured in Europe, Asia, and the US (e.g., Diaz Leon et al., 2011; Lozano et al., 2011; Masson et al., 2011; Morino et al., 2011). Observations on ^{131}I activities in the atmosphere have provided spatial distributions of ^{131}I after the Fukushima accident (Masson et al., 2011) and constituted the foundation of assessing its health and environmental effects. However, data on time series depositional fluxes are still lacking, preventing the assessment of the removal rate of ^{131}I and the evaluation of its actual dosage deposited on the ground, as well as the prediction of the scope of dispersion especially in North America. In addition, the resident and transport timescales of ^{131}I in the atmosphere remain poorly quantified owing to its short half-life (8.0 days) and rare occurrence except for nuclear tests/accidents. Nevertheless, the removal rate and residence time of ^{131}I in the atmosphere are crucial to predicting spatially its affected scope and quantitatively its depositional levels along its dispersion dimensions.

The unfortunate accident of Fukushima has provided an opportunity to evaluate the transport and resident timescale of ^{131}I in the atmosphere. In this study, time series depositional fluxes of ^{131}I from Fukushima were measured along with naturally occurring radionuclides (^{210}Po and ^{210}Pb) to quantify the ^{131}I dosage deposited on the southern Mississippi. Furthermore, a definite integral model based on time series ^{131}I fluxes has been developed to estimate the removal rate and resident timescale of ^{131}I in the atmosphere, providing crucial parameters for the atmospheric transport model of radionuclides.

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2. Methods

The sampling site is located at the NASA-Stennis Space Center (SSC) in South Mississippi (30°22′31″N, 89°36′51″W). The deposition collectors were deployed on the roof of Building 1020 at the University of Southern Mississippi. Both wet and dry depositions were collected using cleaned polyethylene containers. The bulk deposition was transferred into a cleaned polyethylene bottle and processed using established methods for the analyses of iodine in Chernobyl accident, nuclear tests and natural seawater samples (Palmer, 1969; Thomas and Martin, 1986; Suzuki et al., 2008). Predetermined amounts of ^{209}Po spike were then added into the samples. Radioiodine and other nuclides (^{209}Po, ^{210}Po and ^{210}Pb) were co-precipitated with iron hydroxides/oxides at pH of 9 (Couture and Seitz, 1983; Yang et al., 2011). The precipitate was collected through centrifugation. The river water sample was collected from the east channel of the Pearl River, Mississippi, at the Stennis Space Center (30°20′56″N, 89°38′29″W) on April 6, 2011. About 40 L of river water were collected using pre-cleaned polyethylene buckets. Sample processing for the river water was the same as those described for the rainwater.

The precipitate was dissolved with HCl and transferred to a 10 mL counting vial for gamma spectroscopy analysis. Activities of radionuclides including ^{131}I and ^{210}Pb were determined by non-destructive gamma counting using a Canberra ultra-high purity germanium well detector interfaced with a multi-channel analyzer (Guo et al., 2002; Su and Huh, 2002), at 364.5 and 46.5 KeV for ^{131}I and ^{210}Pb, respectively (Pringle et al., 1986). The net counts for these nuclides were all higher than 400, resulting in a counting error of <5%. All activities of ^{131}I and ^{210}Pb were corrected to the mid-time of sample collection.

Procedures for ^{210}Po separation were based on Flynn (1968) and modified by Yang et al. (2011). Briefly, after non-destructive gamma measurements, samples were digested by HNO_3 and HF until the solution became clear. The residue was dissolved with 1 mol/L HCl solution. After the addition of ascorbic acid, hydroxylamine hydrochloride and sodium citrate, the pH value was adjusted to 1.5 with NH_4OH, and Po isotopes were deposited onto a silver disc at 90 °C for 4 h (Yang et al., 2011). Polonium isotopes including ^{209}Po and ^{210}Po were measured by a Canberra alpha analyst system. The activities of ^{210}Po were corrected for its decay and contribution from its parent ^{210}Pb after sampling. The reported data were corrected to the mid-time of sample collection.

3. Results and discussion

3.1. ^{131}I deposition and activities

Following the Fukushima accident, a high level of ^{131}I was first detected on March 23 in a dry deposition sample at the SSC, Mississippi with a depositional flux of 23 ± 2 mBq m^{-2} d^{-1}. After this initial fallout, the ^{131}I depositional flux increased up to ten- to twenty-fold during the following two weeks, varying from 218 ± 18 mBq m^{-2} d^{-1} to 463 ± 16 mBq m^{-2} d^{-1} (Fig. 1). However, the ^{131}I abundance became nearly undetectable after April 19, 2011. During this fallout period, the cumulative ^{131}I depositional flux at the SSC was 4.6 ± 0.2 Bq m^{-2}. The elevated ^{131}I fallout coincides with the two atmospheric ^{131}I peaks observed in Europe between March 28 and April 5 (Masson et al., 2011). Clearly, almost simultaneous occurrence of the highest ^{131}I in the two different continents implies that the radioactive materials were transported to the southern United States and Europe through different pathways. Modeling results also suggested different diffusion passages of radionuclides from Fukushima to the contiguous United States and Europe (e.g., Diaz Leon et al., 2011).

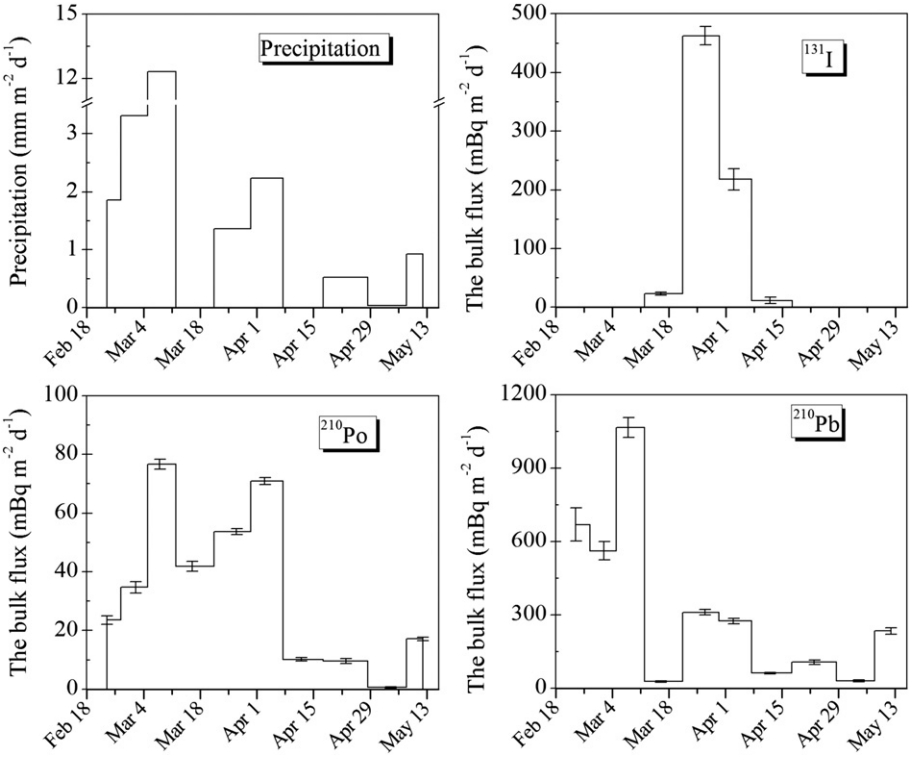

Fig. 1. Precipitation and time series depositional fluxes of ^{131}I, ^{210}Po and ^{210}Pb at the Stennis Space Center on the Mississippi Gulf coast before, during and after Fukushima nuclear plant explosions in March 2011.