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Fukushima-derived fission nuclides monitored around Taiwan: Free tropospheric versus boundary layer transport

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ABSTRACT

The 2011 Fukushima nuclear accident in Japan was the worst nuclear disaster following the 1986 Chernobyl accident. Fission products (nuclides) released from the Fukushima plant site since March 12, 2011 had been detected around the northern hemisphere in about two weeks and also in the southern hemisphere about one month later. We report here detailed time series of radioiodine and radiocesium isotopes monitored in a regional network around Taiwan, including one high-mountain and three ground-level sites. Our results show several pulses of emission from a sequence of accidents in the Fukushima facility, with the more volatile ^{131}I released preferentially over ^{134}Cs and ^{137}Cs at the beginning. In the middle of the time series, there was a pronounced peak of radiocesium observed in northern Taiwan, with activity concentrations of ^{134}Cs and ^{137}Cs far exceeding that of ^{131}I during that episode. From the first arrival time of these fission nuclides and their spatial and temporal variations at our sampling sites and elsewhere, we suggest that Fukushima-derived radioactive nuclides were transported to Taiwan and its vicinity via two pathways at different altitudes. One was transported in the free troposphere by the prevailing westerly winds around the globe; the other was transported in the planetary boundary layer by the northeast monsoon wind directly toward Taiwan.

1. Introduction

The Fukushima nuclear accident occurred in the wake of the double disaster of the 9.0 magnitude Tohoku earthquake and tsunami on March 11, 2011. The earthquake triggered the shutdown of the three active reactors at the Fukushima Daiichi nuclear power station, and the tsunami stopped the station's backup diesel generators, causing a station blackout. The subsequent lack of electric cooling led to a series of explosions and complete meltdowns of three active reactors' cores at the Fukushima facility, with problems at all six reactor units and the central spent fuel pool (Wikipedia website). At the first several days of the accidents, air transport in the mid-latitudes was dominated by prevailing westerly winds, which could circle around the globe in 2–3 weeks (Uno et al., 2009; Warneck, 2000). Therefore, if Fukushima-derived radionuclides were introduced into and above the planetary boundary layer, their dispersal on both regional and global scales would be inevitable. Indeed, fission products released from the accidents had since been spread around the globe (information available from the website of CTBTO Preparatory Commission). For instance, it took only 4 days for the gaseous ^{133}Xe ($T_{1/2}=5.2$ d) to be transported >7000 km eastward, from the emission point in Fukushima Japan (37°25′17″N, 141°1′57″ E) across the Pacific Ocean, and be detected on March 16 at the Pacific Northwest Laboratory in Richland, Washington, U.S.A. (46°16′47″N, 119°16′53″W) (Bowyer et al., 2011). In the ensuing days, ^{131}I ($T_{1/2}=8.02$ d), ^{134}Cs ($T_{1/2}=2.06$ yr) and ^{137}Cs ($T_{1/2}=30.1$ yr) originated from Fukushima were detected across the contiguous United States, progressively from the western seaboard to the eastern seaboard (US Environmental Protection Agency website). By March 24, Fukushima-derived radioactive materials had traveled across the Atlantic and been detected at many CTBTO (Comprehensive Nuclear-Test-Ban Treaty Organization) monitoring stations and elsewhere in Europe (Masson et al., 2011), from the Iberian Peninsula (Lozano et al., 2011) to Thessaloniki, Greece (Manolopoulou et al., 2011). As of April 13, about one month after the nuclear accident, Fukushima-derived radioactivity had spread to the southern hemisphere and had been detected in the Asia-Pacific region at stations located for example in Fiji, Malaysia, Papua New Guinea and even Australia (CTBTO Preparatory Commission website).

Compared with the information about the dispersion of Fukushima-derived fission nuclides in the western hemisphere, much less data is available in eastern Asian countries and we are not aware of any data outside CTBTO's signatory nations. Based in Taiwan, we have been continuously collecting aerosol samples on a daily basis from a ground-level network and high-mountain sites. The samples were routinely analyzed by nondestructive gamma spectrometry for two radionuclides commonly used as aerosol tracers (*i.e.*, ^{7}Be and ^{210}Pb) followed by

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destructive analyses for various chemical constituents (Hsu et al., unpublished results). Besides the naturally occurring nuclides, anthropogenically produced fission products including ^{131}I, ^{134}Cs and ^{137}Cs were detected coincidentally on the air filters collected from late March to late April subsequent to the Fukushima nuclear accident. We feel it warranted to make a timely report of the time series and discuss its implications for radiation safety and atmospheric transport.

2. Observation sites and methods of sampling and analysis

Fig. 1 shows the location of our sampling sites with respect to that of Fukushima in eastern Japan. Of the four sampling sites, two are located in Taiwan (NK and MLL) and the other two are on offshore islets (PCY and DS). The choice of these sites for this particular study is dictated by their altitude and orientation in the regional and global wind field. MLL (Mt. Lulin; 23°28′07″N, 120°52′25″E, 2862 m) is a high-mountain site in Taiwan's Central Range, which is under the influence of the westerlies, especially from March to May. The other three ground-level sites are confined in the boundary layer and are influenced primarily by East Asian monsoon which blows from the northeast toward the southwest in this season. NK (Nankang; 25°02′26″N, 121°36′50″E,) is in the Taipei Basin in northern Taiwan; PCY (Pengchiayu Islet; 25°37′12″N, 122°4′12″E) is upwind in the southern East China Sea; and DS (Dongsa Islet; 20°41′54″N, 116°43′43″E,) is downwind in the northern South China Sea.

The aerosol samples were collected by 24-hour pumping of ~1400–2000 m^3 air through cellulose and/or glass fiber filters (8 in.×10 in. in size). A total of five HPGe detectors with 100–150% relative efficiency (with respect to 3×3 NaI) were employed to provide the necessary throughput for counting daily samples returned from all sites in a timely manner. Absolute efficiencies of the detectors for counting nuclides on the filter samples were calibrated using blank filters spiked with a mixed reference materials containing known activity concentrations of ^{210}Pb, ^{214}Pb (in equilibrium with ^{226}Ra), ^{134}Cs and ^{137}Cs, among other nuclides. The efficiencies for counting ^{134}Cs and ^{137}Cs were calculated directly and that for ^{131}I was derived from the efficiency curve obtained by least-squares fitting. Activity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in each sample were calculated based on net counts registered under the photon peaks centered at 364.48 keV, 604.66 keV and 661.62 keV, respectively. A sample spectrum is given in the Supplementary Material (Fig. S1). The minimum detection limits (MDL) of the nuclides of interest can be approximately evaluated using the equation:

$$MDL = A \times 3\sqrt{C_{bkgd}}/C_{sample}$$

where A represents the activity concentration of the nuclide of interest in the sample, C_{sample} is the accumulated count in the photon peak of interest in the sample spectrum, and C_{bkgd} is the corresponding count from the spectrum obtained by counting blank filters for the same length of time. Since the accumulated counts (C_{sample} and C_{bkgd}) are proportional to the counting time (T), it can be shown that $MDL \propto 1/\sqrt{T}$. Thus, MDL is actually a dependent variable whose value can be reduced by increasing the sample size (i.e., air volume pumped through the filters) and/or the counting time. To maintain the sample throughput and achieve the specified temporal resolution, the counting time of our samples varied between 12 and 48 h, resulting in detection limits generally on the order of μBq/m^3, significantly below the A values reported here for all three nuclides.

It should be noted here that, using the filtration method, the measured radioactivities represent those in the particulate form. Although radioactivity in the gaseous form may be minor or negligible for the particle-reactive ^{137}Cs, it could constitute a major fraction of the total ^{131}I (Masson et al., 2011; Morino et al., 2011), especially near the source of emission. However, for the purpose of this study, it is unnecessary and actually provides no advantage to measure absolute and total (*i.e.*, gaseous plus particulate) activities, which requires the

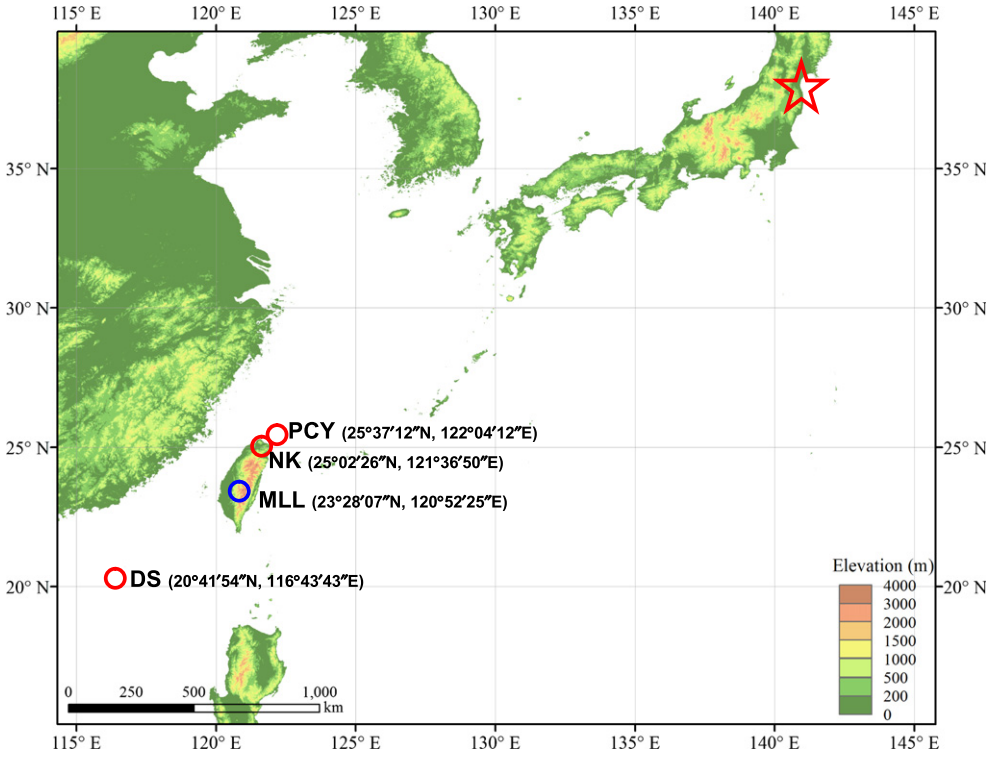

Fig. 1. Map showing the location of Fukushima, Japan (red star) and sampling sites in and around Taiwan. Mt. Lulin (MLL) is a high-mountain site (the blue circle) while Pengchiayu (PCY), Nankang (NK) and Dongsa (DS) are ground level sites (indicated by red circles).