Atmospheric radionuclides from Fukushima Dai-ichi nuclear accident detected in Lanzhou, China (Postprint)

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Abstract

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Full Text

Preamble

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Atmospheric radionuclides from Fukushima Dai-ichi nuclear accident detected in Lanzhou, China*

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After the Fukushima Dai-ichi nuclear power plant accident on March 11, 2011, the radioactivity released from the accident was transported around the globe by atmospheric processes. The radioactivity monitoring program on atmospheric particulate in Lanzhou, China was activated by GSCDC to detect the input radionuclides through atmospheric transport. Several artificial radionuclides were detected and measured in aerosol samples from March 26 to May 2, 2011. The peaked activity concentrations (in mBq/m3) were: 1.194 (131I), 0.231 (137Cs), 0.173 (134Cs) and 0.008 (136Cs), detected on April 6, 2011. The average activity ratio of 131I/137Cs and 134Cs/137Cs in air were 13.5 and 0.78. The significant increase of 137Cs activity concentration, one order of magnitude higher than pre-Fukushima accident levels, in ground level aerosol was observed in 2013, as its resuspension from soil. The back-trajectory analysis simulated by NOAA-ARL HYSPLIT shows a direct transfer of the air masses released from Fukushima to Lanzhou across the Pacific Ocean, North America and Europe at the height close to 9000 m AGL. The value of effective dose for inhalation is close to one millionth of the annual limit for the general public.

Keywords: Fukushima, Nuclear accident, Radioactivity, Lanzhou, China
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INTRODUCTION

On March 11, 2011, a powerful earthquake of magnitude 9.0 on the Richter scale hit the eastern part of mainland Japan, triggering a gigantic tsunami with waves over 10 m high that damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). When the backup electricity supply was lost, the cooling system of nuclear reactors in the FDNPP broke down, causing hydrogen explosions in the No. 1 and No. 3 reactor buildings on March 12 and 14, respectively. These explosions released a large amount of radioactive materials into the environment. The accident was classified on the INES (International Nuclear and Radiological Event Scale) at the maximum level of 7, the same as the Chernobyl accident. Radioactive nuclides, mostly produced by the nuclear fission of 235U and released into the atmosphere and oceans, were carried by global atmospheric and oceanic circulations all over the world [1–3]. Radioactive particles with different composition, size, shape and structure were identified worldwide in different biotic and abiotic media. The radionuclides detected included 131I, 137Cs, 134Cs, 132Te, 132I, 136Cs, etc. and radioactive noble gases (133Xe, 135Xe) [4, 5].

Lanzhou (36° N, 103° E, 1520 m a.s.l.), the capital of Gansu Province, is located on the upper reach of the Yellow River in north-west China, approximately 3200 km west of Fukushima, Japan. An atmospheric radionuclide monitoring station has been operated by the Gansu Center for Disease Prevention and Control (GSCDC) in Lanzhou. It belongs to the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) International Monitoring System (IMS) and provides atmospheric radioactivity detection data to the CTBTO International Data Center [6, 7]. The Fukushima radionuclides were first detected in the aerosol sample collected on March 26, 2011 by GSCDC.

MATERIALS AND METHODS

A. Materials

In order to quantitatively determine the radionuclide fallouts in Lanzhou due to the Fukushima accident, systematic analysis of aerosol samples was undertaken by GSCDC right after the accident. The samples were measured by a low-background HPGe detector operated in a low-level background shield made of lead, iron and copper. The detector was an ORTEC liquid nitrogen cooled P-type HPGe coaxial detector, with 70% relative efficiency, energy resolution (FWHM) of 1.92 keV at 1332 keV, and an integral background counting rate of 1.97 cps in the energy range of 5–2734 keV.

The aerosol samples were collected with a high volume air sampler (Snow White, Senya Oy, Finland). The sampling air flow was controlled by an inverter which controls the speed of the pump motor through changing the AC frequency. The initial sampling flow rate was about 700 m3/h, and the flow rate was >500 m3/h during a sampling period. The typical air volume passing through the filter in daily sampling was about 16,000 m3. The 3M filter product has a collection efficiency of >80% for particle sizes of 0.2 microns under operating conditions. The effective sampling area was about 420 mm × 550 mm.

B. Methods

The aerosol sampling was carried out at a sampling spot in a rural area north-east of Lanzhou. The air sampler was installed on the top of a concrete platform 6 m above the ground. The exposed filter was compressed into a Φ50 mm × 6 mm cylinder for gamma spectrum measurement.

The sampling frequency was daily. As per the operation procedure, the aerosol monitoring was initiated with 24 h filter exposure, followed by a cooling time of 24 h to let the short-lived radon progeny decay. The gamma spectrum measurement lasted for 24 h.

The gamma spectrometer was calibrated with a reference source, which is a cylinder of Φ50 mm × 5 mm. The calibration source was provided by the National Physical Laboratory, UK. The radionuclides in the source include 241Am, 109Cd, 57Co, 139Ce, 51Cr, 113Sn, 85Sr, 137Cs, 54Mn, 88Y, 65Zn and 60Co. The source substrate is a blank filter for collecting aerosol samples. No correction for self-absorption effects was performed, as the sample density is close to the calibration source. The spectra were analyzed using Gamma-Vision32 provided by ORTEC with the gamma spectrometer.

The peaks at 364.5 keV (131I), 477.6 keV (7Be), 661.6 keV (137Cs), 604.7 keV (134Cs) and 818.5 keV (136Cs) were used for activity determinations. The activity of 131I was decay corrected to the start time of sampling.

RESULTS AND DISCUSSION

A. Radionuclide concentration in aerosol

The aerosol in Lanzhou was monitored daily before the Fukushima accident, and background levels for fission isotopes such as 131I, 134Cs and 136Cs were invariably below the minimum detectable concentration (MDC). These represent anthropogenic radionuclides without natural contribution, which were released in large quantities after atmospheric nuclear weapons tests, the Chernobyl nuclear reactor accident, and authorized releases from nuclear reprocessing facilities. Under normal conditions, only 137Cs, due to its long half-life, is still present in the atmosphere [8].

Starting from March 26, radioisotopes released from nuclear reactors were detected, establishing the arrival time of Fukushima's radioactive cloud in Lanzhou at about 14 days after the accident. The radionuclide detected was 131I, with an activity concentration of 0.064 mBq/m3. On April 6, 2011, 131I, 137Cs, 134Cs and 136Cs in air were recorded at activity concentrations of 1.194, 0.231, 0.173 and 0.008 mBq/m3, respectively. Considering meteorological conditions, the maximum values correlate with rainfall which likely accelerated the radionuclides falling from upper to lower atmospheric layers. Two days later, the activity concentrations fell to about 50%, and they followed a general decreasing trend day by day. After May 2, 2011, all observed values were below the MDC. The average concentrations of Fukushima radionuclides measured by GSCDC were 0.23 $\pm$ 0.30 (131I), 0.03 $\pm$ 0.05 (137Cs) and 0.03 $\pm$ 0.04 (134Cs) mBq/m3.

As shown in [Figure 1: see original paper], the activity concentration of 131I is much higher than 137Cs and 134Cs in daily monitoring results, indicating that the radioactive cloud was richer in 131I, as iodine is a more volatile element than cesium. Results of radioactivity measurements in aerosols following the Fukushima accident showed that at least three radioactive plumes arrived at Lanzhou as indicated by 131I activity concentration, on March 28, April 1 and April 6.

The atmospheric transport model of the radioactive materials released from FDNPP was simulated by BGR (Bundesanstalt für Geowissenschaften und Rohstoffe) according to data from CTBTO. As shown in [Figure 2: see original paper], movements of the released radionuclides were initially controlled by westerly winds toward the Pacific and North America. Under the influence of the cyclonic system prevailing over the Bering Sea, an arm of the air masses moved toward the eastern part of Russia, then entered north-east China following the northerly flow along the western flank of the cyclonic circulation. Another arm of the air masses was rapidly transported across the Pacific Ocean, North America, Europe and Asia, and entered north-west China. After March 26, the fission products could be detected over the northern hemisphere.

According to the report of MEP (Ministry of Environmental Protection, the People's Republic of China), the average concentrations (in mBq/m3) of Fukushima radionuclides measured in mainland China were 0.60 $\pm$ 0.89 (131I), 0.16 $\pm$ 0.15 (137Cs) and 0.15 $\pm$ 0.14 (134Cs). The average concentrations (in mBq/m3) measured in northern Chinese provinces were 0.88 $\pm$ 1.11 (131I), 0.20 $\pm$ 0.17 (137Cs) and 0.19 $\pm$ 0.16 (134Cs). The average concentrations (in mBq/m3) measured in southern Chinese provinces were 0.30 $\pm$ 0.31 (131I), 0.11 $\pm$ 0.07 (137Cs) and 0.10 $\pm$ 0.06 (134Cs). The average concentrations measured by GSCDC were lower than the MEP data because lower activity concentrations (of the order of 10−6 Bq/m3) were not reported or detected by the monitoring stations of MEP.

As shown in [Figure 3: see original paper], average concentrations of the Fukushima radionuclides measured in different provinces in China show a significant decrease from north to south and from north-west to south-east, which complies with the BGR simulation that two main radionuclide transport pathways from the Fukushima accident entered mainland China.

Before the Fukushima fallout, however, 137Cs at activity concentrations close to MDC (of the order of 10−6 Bq/m3) was occasionally detected in aerosol samples due to residual radioactivity from atmospheric atomic bomb tests and the Chernobyl accident. Under normal background conditions, the main source of 137Cs in ground level aerosols is its resuspension from soil. This contamination had been monitored for a long time and no evidence of large fluctuations in air was found. In March 2013 ([Figure 4: see original paper]), 137Cs was detected with a significant increase in activity concentration in aerosol samples during a sand storm period. The 137Cs activity concentration was of the order of 10−5 Bq/m3, one order of magnitude higher than pre-Fukushima accident levels, indicating that the Fukushima fallout increased the activity concentration of 137Cs in soil in Lanzhou.

B. Fukushima radionuclides vs. cosmogenic 7Be

The cosmogenic 7Be, produced in the lower stratosphere and upper troposphere, has been widely used as a tracer of atmospheric processes [9]. Although the production of 7Be in the atmosphere is very different from 131I, 137Cs and 134Cs, its presence in the troposphere, and specifically its transport from the stratosphere/troposphere to ground-level air, may provide information on vertical transport of air masses and could help better understand the behavior of Fukushima radionuclides in the troposphere.

The 131I, 137Cs and 134Cs vs. 7Be aerosol activity record ([Figure 5: see original paper]) shows that the appearance of the 131I, 137Cs and 134Cs activity concentration maxima was usually accompanied by 7Be increases. However, the correlation coefficients of 131I, 137Cs and 134Cs vs. 7Be, which are r = 0.167 (p = 0.324), r = −0.064 (p = 0.732) and r = −0.073 (p = 0.736), respectively, differ from results studied by other authors. As $p > 0.05$ indicates no correlation between 7Be and 131I, 137Cs and 134Cs. Povinec [8] found the 131I vs. 7Be and 137Cs vs. 7Be correlation coefficients of r = 0.55 (p = 0.061) and r = 0.59 (p = 0.043), respectively, and Lujanienė [10] found a stronger correlation between 131I, 137Cs and 7Be (r = 0.69 and 0.75, respectively). The different correlation coefficients were probably due to the small data set.

C. Activity ratios

The 131I/137Cs activity ratio in aerosol samples varied from 0.3 to 133.5 with an average of 13.5 from March 27 to May 2 (Figure 6: see original paper). The average value is close to the 131I/137Cs activity ratio in total atmospheric release which was estimated to be 11 [11]. The 131I/137Cs ratios in mainland China ranged from 0.8 to 26.4 with an average of 6.3 according to the MEP data.

To compare the 131I/137Cs activity ratio at the initial release of radionuclides from the FDNPP, the 131I activity was decay-corrected to March 12, 2011 when the first emission of radionuclides occurred. As shown in Figure 6: see original paper, the decay-corrected 131I/137Cs activity ratios ranged from 18.9 to 486.3, with an average value of 122. From March 27 to April 20, the decay-corrected 131I/137Cs activity ratio showed a minimum value of 36 on April 5. However, the 131I/137Cs activity ratio seemed to return gradually to the values observed before April 5 after the minimum was reached. Similar variations in concentration and temporal decreases in the 131I/137Cs activity ratio were monitored in western regions of Japan such as Kanazawa, Nagoya, Osaka, and Tokushima [12].

The main long-lived constituents in the Fukushima fallout in Lanzhou are 137Cs and 134Cs, which have half-lives of 30.17 years and 2.06 years, respectively. These nuclides are utilized as time markers. The 134Cs/137Cs activity ratio can reveal information on radioisotope origin, such as distinguishing fallout from a bomb or fission products from a power reactor [13]. The relatively high activity concentrations of 137Cs and the very low values of activity ratio of 134Cs/137Cs, in combination with the absence of 134Cs in most cases, indicate a strong contribution from "older" 137Cs, probably from the Chernobyl accident and past global fallout [14]. The contribution of 134Cs was similar to 137Cs in the air from the FDNPP accident, as the 134Cs/137Cs activity ratio was close to 1 [15]. [Figure 7: see original paper] shows the 134Cs/137Cs activity ratios in aerosols taken in Lanzhou. The ratios of 134Cs/137Cs observed from March 27 to April 19 ranged from 0.3 to 1.4 with an average of 0.8, while the 134Cs/137Cs activity ratio based on the MEP data varied from 0.3 to 1.8 with an average of 0.9 in mainland China.

D. Back trajectories analysis

In order to simulate the pathway of air masses from Fukushima arriving at Lanzhou, China, the NOAA-ARL (National Oceanic and Atmospheric Administration-Air Resources Laboratory) HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model was used. Fukushima radionuclides were detected in Lanzhou on March 26, 14 days after the Fukushima nuclear accident. The maximum duration that can be simulated in the HYSPLIT application is 312 h (13 days). Thus, 13-day back-trajectories were used to study the transport of air masses from Japan to Lanzhou.

The back-trajectory analysis ([Figure 8: see original paper]) shows a direct transfer from Fukushima starting on March 13 across the Pacific Ocean, North America and Europe, arriving in Lanzhou at a height of 9000 m AGL (above model ground level). Therefore, we can conclude that the air masses released from the Fukushima accident on March 12 were transported to Lanzhou at a height close to 9000 m AGL.

E. Effective dose for inhalation

In the radioactivity from Fukushima, the ratio of gaseous to total 131I over Europe averaged 0.772 $\pm$ 0.136 based on pooled measurements of both 131I forms, with a noticeable constant value geographically and altimetrically [16]. Therefore, the total 131I concentration in the air mass over Lanzhou would be nearly four times the particulate 131I activity concentration measured on filters, i.e., at the peak it would be about 4.76 mBq/m3.

The effective dose for inhalation to the general population produced by the arrival of the radioactive plume to Lanzhou was calculated using the conversion factors set by the IAEA [17], an average inhalation rate of 22.2 m3/d per person, and the activity concentrations measured by GSCDC. The obtained values were 2.45 nSv and 8.86 nSv when the activity concentrations of 131I were corrected by the gaseous/total concentration ratio, respectively. The very low effective dose calculated from the data collected in the aerosol samples demonstrates that the FDNPP accident in Japan had no impact on the health of the population of Lanzhou.

CONCLUSION

The radioactivity from the FDNPP accident, triggered by the earthquake and tsunami of March 11, 2011, was detected in the ground surface air of Lanzhou 14 days after the steam venting and explosions. From March 26 to May 2, 2011, at least one kind of Fukushima radionuclide among 131I, 137Cs, 134Cs and 136Cs was detected above background level.

On the basis of radionuclide observations carried out in the air of Lanzhou following the accident at the FDNPP, we may summarize the obtained results as follows:

Results of radioactivity measurements in aerosols following the FDNPP accident show that at least three radioactive plumes arrived at Lanzhou as indicated by 131I activity concentration. The back-trajectory analysis simulated by NOAA-ARL HYSPLIT shows a direct transfer of the air masses released from Fukushima to Lanzhou across the Pacific Ocean, North America and Europe at a height close to 9000 m AGL. The aerosol monitoring data from 2013 shows the higher activity concentration of 137Cs was of the order of 10−5 Bq/m3, one order of magnitude higher than pre-Fukushima accident levels. This demonstrates that long-lived 137Cs from the FDNPP accident could be detected in ground level aerosols through its resuspension from soil and will exist in the environment for a long time.

The risk of effective dose for inhalation to human health produced by the arrival of the radioactive plume to Lanzhou can be compared to the dose limit to the general public of 1 mSv/year. The value of effective dose is close to one millionth of the annual limit. It can be concluded that at Lanzhou the dose increase due to the Fukushima accident is entirely negligible.

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Note: Figure translations are in progress. See original paper for figures.