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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/m^3) were: 1.194 (^{131}I), 0.231 (^{137}Cs), 0.173 (^{134}Cs) and 0.008 (^{136}Cs), detected on April 6, 2011. The average activity ratio of ^{131}I/^{137}Cs and ^{134}Cs/^{137}Cs in air were 13.5 and 0.78. The significant increase of ^{137}Cs 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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I. INTRODUCTION

On March 11, 2011, a powerful earthquake of 9 in the Richter scale hit the eastern part of mainland Japan. It triggered gigantic tsunami, with waves in heights of over 10 m, damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). When the backup electricity supply was lost, cooling system of nuclear reactors in the FDNPP was broken down, causing hydrogen explosions in No.1 and No. 3 reactor buildings on March 12 and 14, respectively. The explosions released a large amount of radioactive materials into the environment. The accident was classified on the INES (International Nuclear and Radiological Event Scale) scale at the maximum level of 7, the same as the Chernobyl accident. Radioactive nuclides, mostly produced by the nuclear fission of ^{235}U 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 ^{131}I, ^{137}Cs, ^{134}Cs, ^{132}Te, ^{132}I, ^{136}Cs, etc. and radioactive noble gases (^{133}Xe, ^{135}Xe) [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 Gansu Center for Disease Prevention and Control (GSCDC) in Lanzhou. It belongs to Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) International Monitoring System (IMS) and provides atmospheric radioactivity detection data to CTBTO International Data Center [6, 7]. The Fukushima radionuclides were first detected in the aerosol sample collected on March 26, 2011 by GSCDC.

II. 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 low-background HPGe detector operated in 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% relatively efficiency, energy resolution (FWHM) of 1.92 keV at 1332 keV, and the integral background counting rate of 1.97 cps in energy range of 5–2734 keV.

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

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B. Methods

The aerosol sampling was carried out at sampling spot in rural in northeast of Lanzhou. The air sampler was installed on the top of a concrete platform which is 6 m height above the ground. The exposed filter was compressed into a Φ50 mm $\times$ 6 mm cylinder for gamma spectrum measurement. The sampling frequency was daily.

As 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 reference source, which is a cylinder of Φ50 mm $\times$ 5 mm. The calibration source was provided by National Physical Laboratory, UK. The radionuclides in the source include ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{88}Y, ^{65}Zn and ^{60}Co. 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 GammaVision32 provided by ORTEC with the gamma spectrometer. The peaks at 364.5 keV (^{131}I), 477.6 keV (^{7}Be), 661.6 keV (^{137}Cs), 604.7 keV (^{134}Cs) and 818.5 keV (^{136}Cs) were used for activity determinations. The activity of ^{131}I was decay corrected to the start time of sampling.

III. RESULTS AND DISCUSSION

A. Radionuclide concentration in aerosol

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

Starting from March 26, radioisotopes release from nuclear reactors was detected. In this way, we could establish the Fukushima's cloud arrival time in Lanzhou, about 14 days after the accident. The radionuclide detected was ^{131}I, with the activity concentration of 0.064 mBq/m^3. On April 6, 2011, ^{131}I, ^{137}Cs, ^{134}Cs and ^{136}Cs in air were recorded, in activity concentrations of 1.194, 0.231, 0.173 and 0.008 mBq/m^3, respectively. Considering about the meteorological condition, the maximum values are correlated with rainfall which accelerated the radionuclides falling from upper to lower atmospheric probably. Two days after, the activity concentrations fell to about 50%, while they followed a general decreasing trend day by day. After May 2, 2011 all the observed values were below the MDC. The average concentrations of Fukushima radionuclides measured by GSCDC were 0.23 $\pm$ 0.30 (^{131}I), 0.03 $\pm$ 0.05 (^{137}Cs) and 0.03 $\pm$ 0.04 (^{134}Cs) mBq/m^3.

As shown in Fig. 1, the activity concentration of ^{131}I is much higher than ^{137}Cs and ^{134}Cs in daily monitoring results. This means the radioactive cloud was richer in ^{131}I, 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 ^{131}I activity concentration, on March 28, April 1 and April 6.

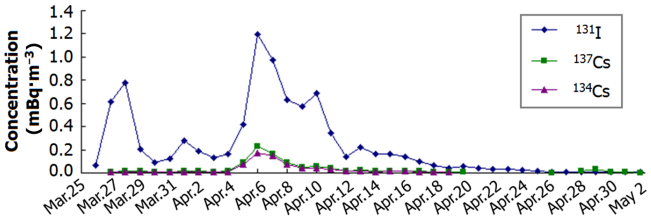

Fig. 1. (Color online) Daily average of measured activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs.

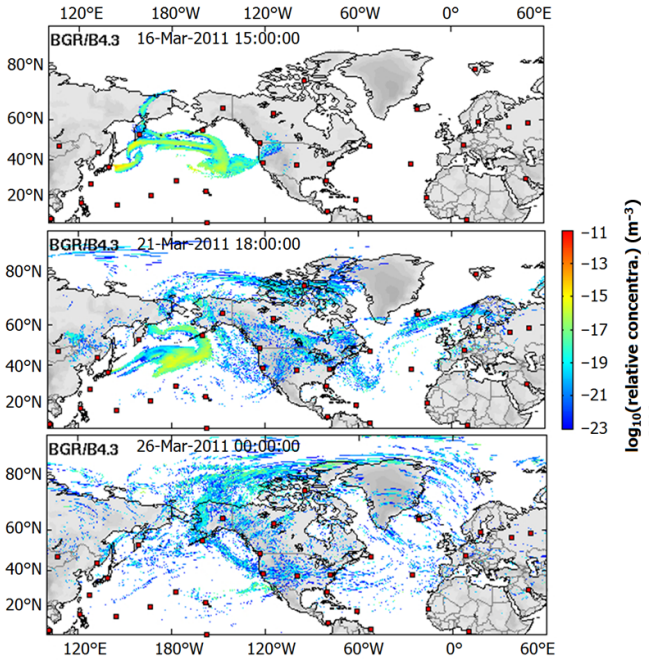

Fig. 2. (Color online) Simulation of the transport of air masses from Fukushima by BGR on March 16, 21 and 26, 2011.

The atmospheric transport model of the radioactive materials released from FDNPP was simulated by BGR (Bundesanstalt für Geowissenschaften und Rohstoffe) according to the data from CTBTO. As shown in Fig. 2, movements of the released radionuclides were initially controlled by the westerly winds toward the Pacific and North America. Under influence of the cyclonic system prevailing over the Bearing Sea, an arm of the air masses moved toward the eastern part of Russia. It 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/m^3) of Fukushima radionuclides measured in China mainland were 0.60 ± 0.89 (^{131}I), 0.16 ± 0.15 (^{137}Cs) and 0.15 ± 0.14 (^{134}Cs). The average concentrations (in mBq/m^3) measured in China northern provinces were 0.88 ± 1.11 (^{131}I), 0.20 ± 0.17 (^{137}Cs) and 0.19 ± 0.16 (^{134}Cs). The average concentrations (in mBq/m^3) measured in China southern provinces were 0.30 ± 0.31 (^{131}I), 0.11 ± 0.07 (^{137}Cs) and 0.10 ± 0.06 (^{134}Cs). The average concentrations measured by GSCDC were lower than the MEP data, because lower activity concentrations (of the order of 10^{-6} Bq/m^3) were not reported or detected by the monitoring station of MEP.

From Fig. 3, average concentrations of the Fukushima radionuclides measured in different provinces in China show significantly decrease from north to south and from north-west to south-east. It complied with the simulation of BGR that two main radionuclide transport pathways of the Fukushima accident entered mainland China.

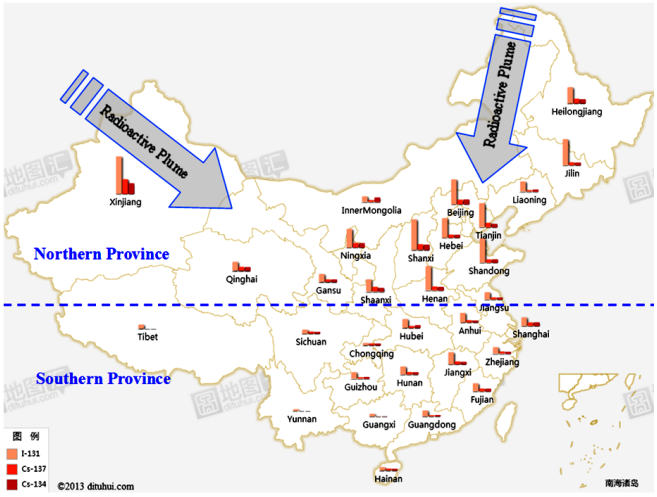

Fig. 3. (Color online) Average concentrations of the Fukushima radionuclides measured in mainland China.

Before the Fukushima fallout, however, ^{137}Cs in activity concentration closed to MDC (of the order of 10^{-6} Bq/m^3) was occasionally detected in aerosol samples due to the residual radioactivity of the atmospheric atomic bomb tests and of the Chernobyl accident. In contrast to normal background conditions when the main source of ^{137}Cs 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 (Fig. 4), ^{137}Cs was detected with significantly increase in activity concentration in aerosol samples during a sand storm period. The ^{137}Cs activity concentration was of the order of 10^{-5} Bq/m^3, one order of magnitude higher than pre-Fukushima accident level. This indicates that the Fukushima fallout increased the activity concentration of ^{137}Cs in soil in Lanzhou.

B. Fukushima radionuclides vs. cosmogenic ^{7}Be

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

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

C. Activity ratios

The ^{131}I/^{137}Cs activity ratio in aerosol samples varied from 0.3 to 133.5 with the average of 13.5 from March 27 to May 2 (Fig. 6(a)). The average value is closed to the ^{131}I/^{137}Cs activity ratio in total atmospheric release which was estimated to be 11 [11]. The ^{131}I/^{137}Cs ratios in mainland China ranged $0.8 - 26.4$ with an average of 6.3 according to the MEP data.

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

The main long-lived constituents in the Fukushima fallout in Lanzhou are ^{137}Cs and ^{134}Cs, which have half-lives of 30.17 years and 2.06 years, respectively. The nuclides are utilized as time markers. The ^{134}Cs/^{137}Cs activity ratio can reveal information on radioisotope origin, such as deciding

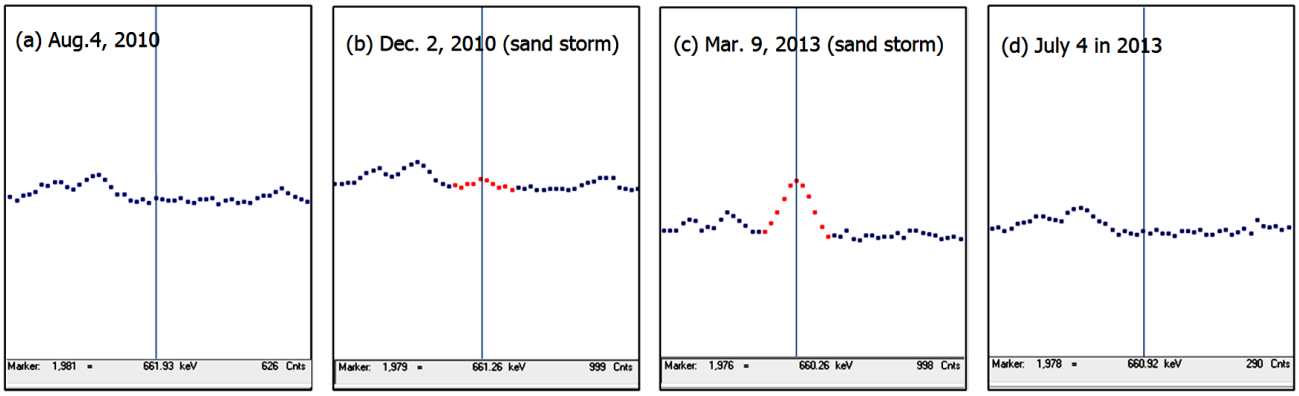

Fig. 4. (Color online) Gamma spectra of ^{137}Cs in aerosol samples collected on different days.

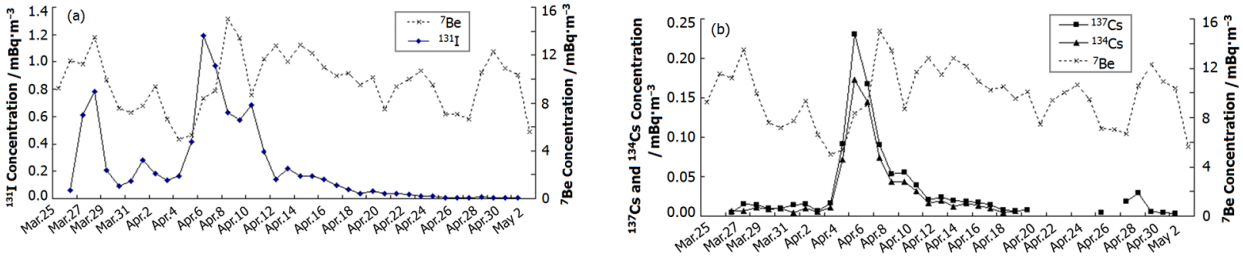

Fig. 5. (Color online) Comparison of ^{7}Be concentration with ^{131}I (a), and ^{137}Cs and ^{134}Cs (b).

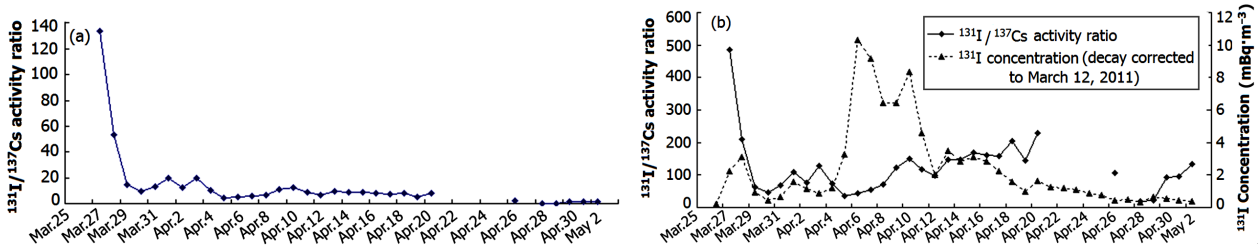

Fig. 6. (Color online) ^{131}I/^{137}Cs concentration ratio in aerosol (a) and the ratio with ^{131}I activity decay-corrected to March 12, 2011(b).

the fallout of a bomb or the fission products of a power reactor [13]. The relatively high activity concentrations of ^{137}Cs and the very low values of activity ratio of ^{134}Cs/^{137}Cs, in combination with the absence of ^{134}Cs in most of the cases indicate a strong contribution from "older" ^{137}Cs, probably from the Chernobyl accident and past global fallout [14]. The contribution of ^{134}Cs was similar to ^{137}Cs, as the ^{134}Cs/^{137}Cs activity ratio was close to 1 in the air from the FDNPP accident [15]. Figure 7 shows the ^{134}Cs/^{137}Cs activity ratios in aerosols taken in Lanzhou. The ratios of ^{134}Cs/^{137}Cs observed from March 27 to April 19 ranged 0.3–1.4 with an average of 0.8; while the ^{134}Cs /^{137}Cs activity ratio based on the MEP data varied from 0.3 to 1.8 with the average of 0.9 in mainland China.

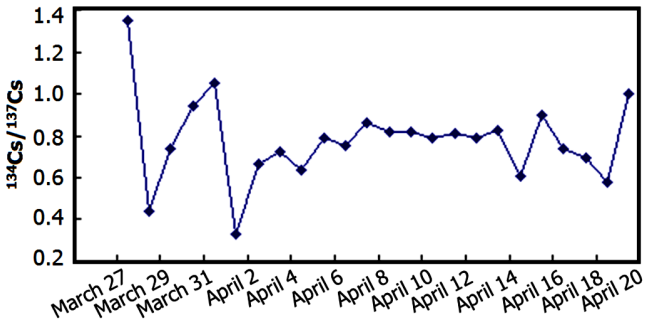

Fig. 7. (Color online) The ^{134}Cs/^{137}Cs concentration ratio in aerosol.

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. The Fukushima radionuclides were detected in Lanzhou on March 26, 14 days after the Fukushima nuclear accident. The maximum hours that can be simulated in the HYSPLIT application are 312 h (13 days). Thus, 13 days back-trajectories were used to study the transport of air masses from Japan to Lanzhou.

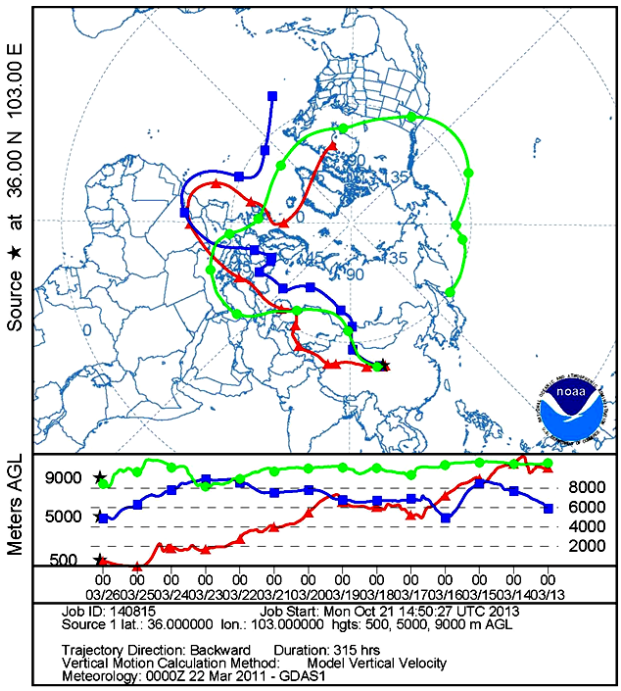

Fig. 8. (Color online) Thirteen-day back-trajectories, arrival at Lanzhou on March 26, 2011.

The back-trajectory analysis (Fig. 8) shows a direct transfer from Fukushima started on March 13 across the Pacific Ocean, North America and Europe arriving in Lanzhou at the height of 9000 m AGL (above model ground level). Therefore, we can know that the air masses released from Fukushima accident on March 12 was transported to Lanzhou at the height close to 9000 m AGL.

E. Effective dose for inhalation

Particulate ^{131}I collected on filter was only part of the total ^{131}I present in the air. In the radioactivity from Fukushima, the ratio of gaseous to total over Europe averaged 0.772 ± 0.136 based on pooled measurements of both ^{131}I forms, with a noticeable constant value, geographically and altimetrically [16]. Therefore, the total ^{131}I concentration in the air mass over Lanzhou would be nearly four times the particulate ^{131}I activity concentration measured on filters, i.e. at the peak it would be about $4.76\,\mathrm{mBq/m^3}$.

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 flux of air of $22.2\,\mathrm{m^3/d}$ per person and the results of the activity concentrations measured by GSCDC. The obtained values were 2.45 nSv and 8.86 nSv with the activity concentrations of ^{131}I were corrected by 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 over the health of the population of Lanzhou.

IV. 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 the Fukushima radionuclide within ^{131}I, ^{137}Cs, ^{134}Cs and ^{136}Cs 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 shows that at least three radioactive plumes arrived at Lanzhou as indicated by ^{131}I 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 the height close to 9000 m AGL.

The aerosol monitoring data of 2013 shows the higher activity concentration of ^{137}Cs was of the order of $10^{-5}\,\mathrm{Bq/m^3}$, one order of magnitude higher than pre-Fukushima accident levels. It demonstrates that long-lived ^{137}Cs from FDNPP accident could be detected in ground level aerosols as its resuspension from soil and it 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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