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Atmospheric pollutants transport tracks revealed from ^{131}I, ^{137}Cs, and ^{134}Cs leaked from Fukushima accident and ^{7}Be and ^{210}Pb observed at Guiyang of China

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Abstract A massive earthquake measuring 9.0 on the Richter scale that occurred on March 11, 2011, on Honshu Island, Japan, caused radioactivity leakage from the Fukushima Nuclear Power Plant, which led to the leakage of artificial nuclides (^{131}I, ^{137}Cs, and ^{134}Cs) and their global transportation by atmospheric circulation. This paper reports a systematic comparative observation on radioactive concentrations of natural nuclides (^{7}Be and ^{210}Pb) and artificial nuclides (^{131}I, ^{137}Cs, and ^{134}Cs) at the surface level, measured in weekly continuous aerosol sampling at Mount Guanfeng, Guiyang, China, from March 17, 2011 to April 28, 2011. During this period, the variations in the nuclide concentrations associated with their transport paths were analyzed with 315 hour back-trajectories of air mass initialized 500 m above the surface level at Guiyang. The results show that the pollutants of nuclear leakage from the Fukushima accident were transported to the Guiyang region of China via two significant pathways. In the first pathway the first wave of nuclear pollutants were transported from west to east in air masses at higher altitudes via global atmospheric circulation. The nuclear pollutants encircled the Earth almost once and after about 10 days to two weeks, between March 24 and March 31, 2011, intruded Guiyang from the northwestern region of China. In the second pathway, the nuclear pollutants from the Fukushima region arrived at Guiyang between April 7 and April 14, 2011, via air masses at lower altitudes that moved southwards because of the squeezing of the northeast Asian weather system and then by the influence, in succession, of the northeastern and southeastern air currents in the low-latitude region. The first transport pathway for atmospheric pollutants is on a global scale and based on air masses at higher altitudes, and the second transport pathway is on an eastern Asia regional scale and based on the air masses at lower altitude.

Key words Fukushima; nuclear leakage; ^{131}I-^{137}Cs-^{134}Cs-^{7}Be-^{210}Pb; aerosol transport; Guiyang

1 Introduction

A magnitude 9.0 earthquake occurred on Honshu, Japan, on March 11, 2011, and caused tsunamis and nuclear leakage (IAEA, 2011). The reactors of the Fukushima Nuclear Power Plant (141.033°E, 37.421°N) were damaged to different degrees, and radionuclides (^{131}I, ^{137}Cs, ^{134}Cs, etc.) were released. After the products of the nuclear leakage entered the atmosphere, radionuclides were successively detected at some monitoring stations in different regions of the northern hemisphere. They were first detected and reported by the monitoring station at Seattle, USA (Diaz Leon et al., 2011). Research on the air mass forward-trajectory (Lozano et al., 2011) showed that these radioactive nuclides arrived at the west coast of the USA, and then moved on and spread to the eastern region of the USA; afterwards, they crossed the Atlantic Ocean to reach the European continent. Several days later, the leaked nuclides were detected in northwestern German, in Milan (Italy) and in Thessaloniki, northeastern Greece, respectively (Clemenza et al., 2012; Manolopoulou et al., 2011; Pittauerová et al., 2011). On the Iberian Peninsula, ^{131}I was detected for the first time in the filter membrane samples taken from the Cáceres station from March 15 to 22, 2011

(CSN, 2011). Two weeks later, determinations conducted at two different stations (FIMERALL with an altitude of 300 m and IZAÑA with an altitude of 2400 m) on Tenerife (Canary Islands, Spain) showed that the specific radioactivity at the FIMERALL station was always higher than that at the IZAÑA station. This result indicates that the radioactive pollutants that reached this island were transported by the air masses at lower altitude (López-Pérez et al., 2013). Comparison of the ^{7}Be activity concentrations, observed weekly, also shows that these radioactive nuclides were transported via the air masses at lower altitude (Masson et al., 2011).

Recently, several observation results of Fukushima fallouts in the Asian regions, such as Korea (Kim et al., 2012; Lee et al., 2013), Beijing (Zhou et al., 2013), Xi'an (Liu et al., 2013), and Taiwan (Huh et al., 2012), Krasnoyarsk (Bolsunovsky and Dementyev, 2011) and West Siberia (Melgunov et al., 2012) of Russia, and Vietnam (Long et al., 2012), are reported. These observations demonstrate that the pollutants leaked from the Fukushima accident has been transported globally. Analyzing the global distribution of radionuclides is one method to trace the atmospheric transport pathways of the pollutants (Lee et al., 2004). However, this method is influenced and restricted by some factors (Lee and Feichter, 1995; Lee, 2004). The central Guizhou area is an inland area located in the central part of the Yunnan-Guizhou Plateau on the slope through which the Qinghai-Tibet Plateau transits to the hills and plains eastwards, and it has low latitude and higher altitude. The weather systems in this area are controlled by the southern branch of the westerlies and are also influenced by the southwesterly air current of the northern branch of the westerlies. Also, the southeast and southwest monsoons, the cold and warm air masses in the south-north direction, often meet here to form the Yunnan-Guizhou stationary front. Observing the concentrations in air and deposition fluxes of tracing nuclides (including ^{7}Be and ^{210}Pb) in the central Guizhou area is of special significance in tracking the global atmospheric transport of pollutants (Lee et al., 2004; Wan et al., 2005, 2008, 2010).

^{7}Be (half-life 53.3 d) is a radionuclide generated when cosmic rays impinge on atmospheric ^{14}N and ^{16}O target nuclei. ^{7}Be reaches its maximum value in the atmospheric layer at a height of about 20 km, and has a residence time of about 1 a in the stratosphere. With the weakening intensity of the cosmic rays radiating into the atmosphere, the yield of ^{7}Be decreases with increases in the thickness of the atmospheric layer and air density (Feely et al., 1989; Lal and Peters, 1967). After it is formed, ^{7}Be attaches to the surface of submicron aerosol particles rapidly, migrates with the atmospheric dynamic process and finally enters the ground environment through dry-wet deposition. The stratosphere, as the generation and accumulation source of ^{7}Be, can transport ^{7}Be to the troposphere through Brewer-Dobson circulation on a global scale; or when exchange occurs between the stratosphere and the troposphere, ^{7}Be enters the troposphere (Feely et al., 1989; Holton et al., 1995; Zanis et al., 1999). Therefore, the change in near-surface concentration of ^{7}Be is usually used as the most powerful tracer for vertical transport by air masses. The concentraion of ^{7}Be in tropospheric air increases with increasing ground altitude.

^{210}Pb (half-life 22.3 a) is an α-decay daughter of the intermediate decay product ^{222}Rn (half-life 3.8 d) of ^{226}Ra (half-life 1622 a) in the ^{238}U series. ^{222}Rn, as an inert gas, diffuses in the lower atmosphere and continuously decays after escaping from rock surfaces and soil particles. Owing to the differences in mineral composition of rock and soil, the escape-generation ratio of ^{222}Rn ranges from lower than 1% to higher than 30% (Dörr and Mūnnich, 1990; Krishnaswami and Seidemann, 1988). The ^{222}Rn in the lower atmosphere varies because of regional differences in the escape ratio and air-current transport. The average escape transport flux of ^{222}Rn from the land surface to the atmosphere is about 0.015 Bq/m^{2} a; the escape transport flux from the ocean surface and ice cover to the atmosphere is 2–3 orders of magnitude lower than that from the land surface (Dörr and Mūnnich, 1990; Turekian et al., 1977). Therefore, ^{210}Pb in the atmosphere is mainly from the land surface and has an obvious regional distribution difference (Lee et al., 2004; Wan et al., 2005, 2008, 2010).

According to the formation mechanisms of the ^{7}Be and ^{210}Pb concentrations in air near the ground surface, when the ^{7}Be concentration increases (or decreases), the sinking of the air mass from a higher altitude strengthens (or weakens); whereas, evidently, when the ^{210}Pb concentration in air increases (or decreases), the intrusion of continental air mass strengthens (or weakens). A strengthening intrusion of the oceanic air mass makes obvious a decrease in both ^{7}Be and ^{210}Pb concentrations (Wan et al., 2010).

Hong et al. (2012) and Hsu et al. (2012) noted the transport pathways of the pollutants released from the Fukushima accident using the ^{137}Cs/^{134}Cs observed in Korea and in Taiwan. In this paper, we compared the concentrations of ^{131}I, ^{137}Cs, and ^{134}Cs leaked from the Fukushima accident with the concentrations of natural nuclides (^{7}Be and ^{210}Pb) observed at Mt. Guanfeng of Guiyang, China, from March 17 to April 28, 2011. Then we calculated the air-mass distribution of 315 h back-trajectories at 500 m above ground in Guiyang (106°43′22.1″E, 26°34′19.3″N) every day in the XY plane, with the USA global reanalysis weather data from the National Centers for Environmental

Prediction (NCEP) with a goal of understanding the geographic track through which the air masses passing through the accident site traveled, and thus verified the atmospheric transport pathway of the nuclear pollutants.

2 Samples and experimental methods

The sampling device was a surface atmosphere sampling system (SASS, Model 500NE Aerosol Sampler) for measuring low-level nuclides and developed by the National Urban Security Technology Laboratory [formerly Environmental Measurements Laboratory (EML)] , USA. The sampling system was placed on the top of an experimental building of the State Key Laboratory of Environmental Geochemistry, about 30 m from the ground.

The (Model Fuji) sampler was equipped with a 20.3 cm × 25.4 cm rectangular filter, which had three layers of 100% polypropylene screen filter membrane as the filter material (Dynaweb, DW7301L) between two layers of 100% polyester protective cotton; it had an effective filtering area of 407 cm^2. The sampling flow velocity was 0.4–1.6 m^3/min. One sample was taken every week; in other words, the sampling time of every sample was about 168 h. The air volume collected on each sample ranged approximately from 0.9×10^4 to 1.1×10^4 standard cube meters (SCM), which was calibrated to the International SCM by observed daily air temperature and average local pressure measurements.

For the radioactivity of ^{131}I, ^{137}Cs, ^{134}Cs, ^{7}Be and ^{210}Pb, γ-spectrum determination was carried out using an S-100 series 16384-channel energy spectrometer manufactured by the Canberra Company, USA. The aerosol sample on a polypropylene screen filter membrane was packaged in a given shape and then placed directly into a GR2019 coaxial high-purity germanium (HPGe) detector (efficiency 20%) for counting. The energy spectrometer had good stability with no channel shift during tests, and the counting time of a single sample was 4.7×10^4–6.3×10^4 s. The counting peaks of ^{131}I, ^{137}Cs, ^{134}Cs, ^{210}Pb and ^{7}Be were at 0.364, 0.662, 0.605, 0.046 and 0.478 keV, respectively; the sample counting was controlled according to one standard deviation and the average errors of actual measurements were ±7.1% (^{131}I), ±11.9% (^{137}Cs), ±8.8% (^{134}Cs), ±1.9% (^{210}Pb) and ±1.6% (^{7}Be), respectively. The multi-nuclide standard source (Catalog No. 7500; Source No. 586-26-6) was provided by Isotope Products Laboratories, USA, and the activity of different nuclides was calculated from the efficiency curves.

Accurate measurements of low-level nuclides require not only a high-stability measurement device and the accurate calibration of energy scales and efficiency based on a multi-nuclide comprehensive standard source, but also on data comparison between international laboratories. To ensure international comparability of the measurement results, some aerosol samples on the polypropylene screen filter membrane were split equally at an early stage, and were then comparatively measured by both the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, and the formerly EML, USA. The scatter data points have very good consistency between two laboratories (Lee et al., 2004; Wan et al., 2005, 2008, 2010).

Fig. 1. Surface air sampling location at Guiyang site (circle) in China.

3 Results and discussion

3.1 Observational results

The observational results of the concentration changes in natural nuclides (^{7}Be, ^{210}Pb) and artificial nuclides (^{131}I, ^{137}Cs, and ^{134}Cs) in aerosols sampled weekly, both in air at near the ground surface at Mt. Guanfeng, Guiyang, since March 17, 2011 (Table 1) show that in the first week (March 17–24), the activity concentrations of ^{7}Be and ^{210}Pb were low, which indicates the effect of weak oceanic warm and wet air masses (Wan et al., 2010); and no ^{131}I, ^{137}Cs, and ^{134}Cs were detected within the first 10 days of the Fukushima nuclear accident. In the second week (March 24–31), the ^{7}Be concentration, which indicates the sinking of air masses from high altitude, reached 11.7 mBq/m^3; the ^{210}Pb concentration, which indicates the effect of inland air masses (Wan et al., 2010), was 3.4 mBq/m^3; and the ^{131}I, ^{137}Cs, and ^{134}Cs concentrations reached 0.22, 0.03 and 0.03 mBq/m^3, respectively. In the third week (March 31–April 7), the ^{7}Be and ^{210}Pb concentrations decreased normally, and those of ^{131}I and ^{137}Cs also decreased to 0.06 and 0.02 mBq/m^3, respectively. In the fourth week (April 7–14), the ^{7}Be

and ^{210}Pb concentrations changed little, and those of ^{131}I, ^{137}Cs, and ^{134}Cs increased to 0.07, 0.07, and 0.06 mBq/m^3, respectively; the ^{137}Cs and ^{134}Cs concentrations obviously increased. In the fifth week (April 14–21), although the ^{7}Be and ^{210}Pb concentrations still indicated a weak sinking of air masses from higher altitude and the effect of inland regions, the concentrations of artificial nuclides in the air had evidently decreased, with ^{131}I decreasing to 0.03 mBq/m^3 and ^{137}Cs and ^{134}Cs both decreasing to 0.02 mBq/m^3, which clearly indicates decay of ^{131}I and diffusion dilution of ^{137}Cs. In the sixth week (April 21–28), the ^{7}Be and ^{210}Pb concentrations still indicated the sinking of air masses from higher altitude and the effect of inland regions, respectively; however, the concentrations of artificial nuclides in air were evidently lower than the detection limits. From the second to fifth week, average of ratios of ^{137}Cs to ^{134} Cs is 0.92. The ratio of ^{131}I to ^{137}Cs decreased gradually from 8.1 to 1.9 due in part to the about four-half time decay of ^{131}I.

To further analyze the mutual relationships among the weekly changes in ^{131}I, ^{137}Cs, and ^{134}Cs concentrations observed in the atmospheric transport of the pollutants from the Fukushima nuclear accident, the distributions of air mass 315 h back-trajectories at 500 m above the ground in Guiyang over seven days of every week from March 17 to April 28, 2011, in the XY plane (Fig. 2) were calculated and plotted according to USA global reanalysis weather data from NCEP and the back-trajectories model (Draxler and Rolph, 2003). The reason why 315 h (nearly two weeks) was used is that the Fukushima Nuclear Power Plant is located leeward of Guiyang and that the time for the nuclear pollutants to be transported to China, probably on global scale, i.e., encircling the Earth once, might be about two weeks. In southwest China the minimal near-surface boundary layer mixing height is above 500 m from March to April each year, so the initial height of the air mass for the calculation of back-trajectories was selected as 500 m above the ground; also, as ^{7}Be and ^{210}Pb themselves are very good tracers in the vertical direction, the vertical transport process of air masses is not given for the calculations of back-trajectories in this paper.

3.2 Influence of nuclear leakage from Fukushima on Guiyang via global atmospheric circulation

In the first week (March 17–24, 2011), the ^{7}Be and ^{210}Pb concentrations, which indicate the effect of oceanic air masses, were low; and no ^{131}I and ^{137}Cs were detected, indicating that the pollutants did not reach Guiyang within 10 days of the Fukushima nuclear accident. As can be seen from Fig. 2a, the back-trajectories of the Guiyang station include two parts, i.e., the northwestern region of China, and the India Peninsula region of the South Asia subcontinent. These two fan regions were obviously not influenced by the air mass transport passing though the nuclear leakage point; meanwhile, as the air masses came from the region of South Asia with lower latitude, the ^{7}Be and ^{210}Pb concentrations in near-surface air were inevitably low in Guiyang.

In the second week (March 24–31, 2011), the ^{7}Be concentration (which indicates the sinking of air masses from higher altitude) observed in Guiyang reached 11.7 mBq/m^3; the ^{210}Pb concentration was 3.4 mBq/m^3, indicating the simultaneous normal effect of inland air masses. During this period, the concentrations of artificial nuclides ^{131}I, ^{137}Cs, and ^{134}Cs reached 0.22, 0.03 and 0.03 mBq/m^3, respectively, which indicates that the air mass in which the pollutants of nuclear leakage from Fukushima were first observed in Guiyang has the characteristics of "sinking from higher altitude" and "inland". The time when the pollutants were found was at least 13 d from the time of the accident, so the transportation has the characteristic of atmospheric circulation transport on a global scale.

Table 1 Concentration of natural (^{7}Be and ^{210}Pb) and artificial (^{131}I, ^{137}Cs, and ^{134}Cs) radionuclides in surface air at Guiyang, China[1]

Sampling day	Air volume[2]	^{7}Be	^{210}Pb	^{131}I	^{137}Cs	^{134}Cs	Ratio
	SCM	mBq/m^3	mBq/m^3	mBq/m^3	mBq/m^3	mBq/m^3	^{137}Cs/^{134}Cs
Mar.17, 2011–Mar.24, 2011	11076	3.1 ± 0.1	1.5 ± 0.1	＜MDA	＜MDA	＜MDA	--
Mar.24, 2011–Mar.31, 2011	10314	11.7 ± 0.1	3.4 ± 0.1	0.22±0.01	0.03±0.01	0.03±0.01	0.93
Mar.31, 2011–Apr.07, 2011	10587	5.1 ± 0.1	2.3 ± 0.1	0.06±0.01	0.02±0.01	0.03±0.01	0.74
Apr.07, 2011–Apr.14, 2011	10921	4.5 ± 0.1	2.7 ± 0.1	0.07±0.01	0.07±0.01	0.06±0.01	1.09
Apr.14, 2011–Apr.21, 2011	9753	8.7 ± 0.1	3.4 ± 0.1	0.03±0.01	0.02±0.01	0.02±0.01	0.92
Apr.21, 2011–Apr.28, 2011	9080	10.9 ± 0.1	3.6 ± 0.1	＜MDA	＜MDA	＜MDA	--

Note: [1] The measurement errors are expressed as 1 standard deviation; [2] The air volume is calibrated to the International SCM.

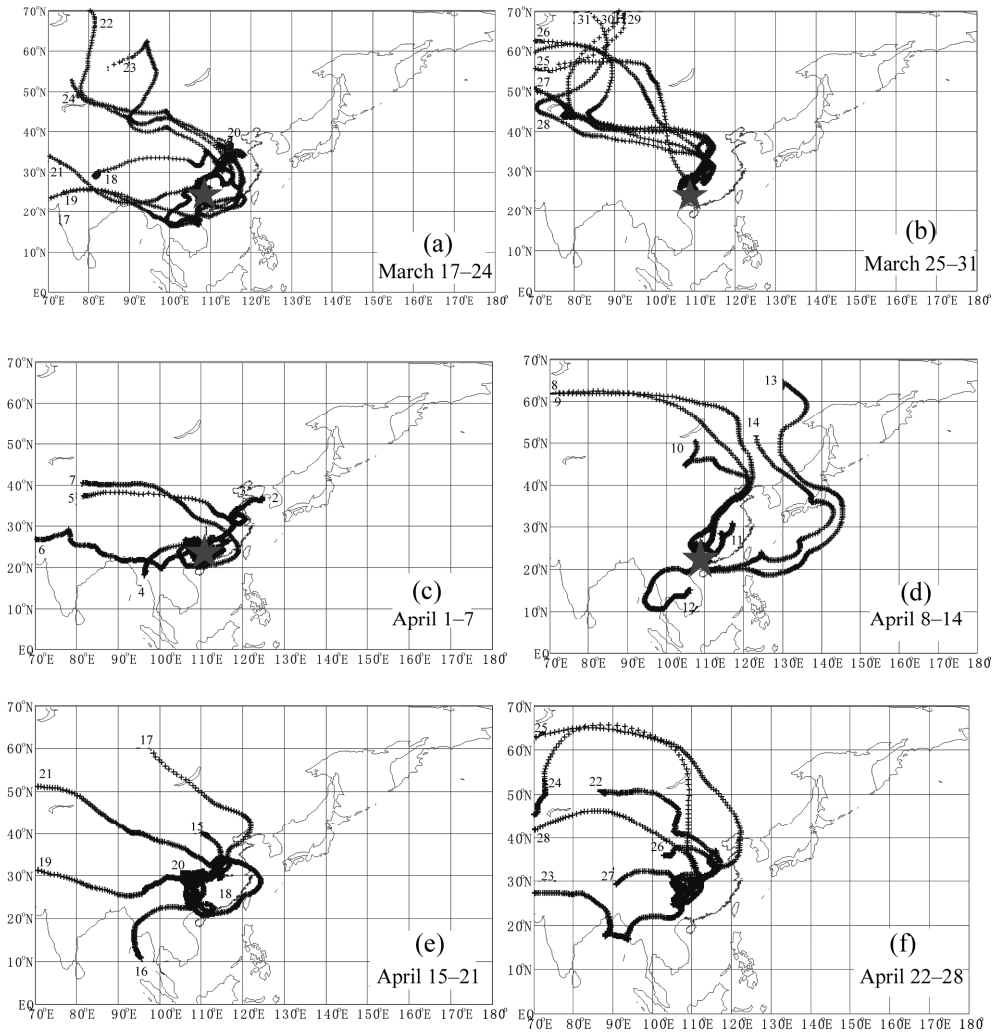

Fig. 2. Distribution of back-trajectories (2011.3.17−2011.4.28) over 315 hours at Guiyang site in China (a. 3.17–24; b. 3.25–31; c. 4.1–7; d. 4.8–14; e. 4.15–21; f. 4.22–28).

As can be seen from Fig. 2b, all the air masses on the back-trajectories in Guiyang during this period were from the regions of northwestern China and Middle Asia, crossing a latitude range of 40°N–60°N, so they have the typical weather characteristics of an upper-air low-pressure trough passing through (Keyser and Shapiro, 1986). These weather characteristics are shown in Figs. 3 and 4. There was obviously a sinking air current from higher altitude; this was verified by the high ^{7}Be concentration observed in Guiyang. During this week (March 24–31, 2011) ^{131}I was observed to have a high concentration, which indicates that from March 11, 2011, when the nuclear leakage occurred in Fukushima, the global diffusion transport of the first wave was completed within about 10 days to two weeks, and the pollutants then entered Guiyang from the northwestern region of China. The above observational results are comparable in time sequence with reported simulation and observational results (Qiao et al., 2011).

Fukushima, Japan, is located in a latitude range controlled by the upper level jet streams in the northern hemisphere in winter and spring. After the nuclear leakage occurred, a high concentration of ^{131}I entered the atmosphere; then, through sufficient mixing, diffusion or convection in the near-surface boundary layer, the ^{131}I reached the free atmosphere level; and finally, transported by a strong upper level jet stream; the ^{131}I was transported rapidly and globally along the latitude zone from west to east.

3.3 Direct transportation of nuclear leakage from Fukushima on the eastern Asia regional scale

From the change in back-trajectories (Fig. 2c), the air masses from low latitude South Asia dominated Guiyang during April 1–7, 2011, when the observed concentrations of artificial nuclides in Guiyang clearly decreased, and also concentrations of ^{7}Be and ^{210}Pb were relatively low. However, this status changed in the following week (April 7–14), during which period the air-current trajectory had passed over

Fukushima for two days (Fig. 2d). The ^{131}I, ^{137}Cs, and ^{134}Cs concentrations observed in Guiyang clearly increased again, especially that of ^{137}Cs, which increased significantly to 0.07 mBq/m^3. Meanwhile, the sinking air current from the higher altitude of northeast Asia obviously increased the concentrations of ^{7}Be and ^{210}Pb. From Fig. 2d and the change in concentrations of artificial nuclides observed in Guiyang, it can be seen that, although Fukushima is located leeward of China, the nuclear leakage from Fukushima was still transported to China directly in a specific weather system. This phenomenon has already observed in their previous diagnostic analysis of the back-trajectories of ozonezonide sounding observational data in Lin'an, China, that pollutants from Korea and/or Japan can influence the eastern region of China (Zheng et al., 2005). The observations and analysis reported here further demonstrate the possibility that such pollutants influence the southwestern region of China through direct transportation.

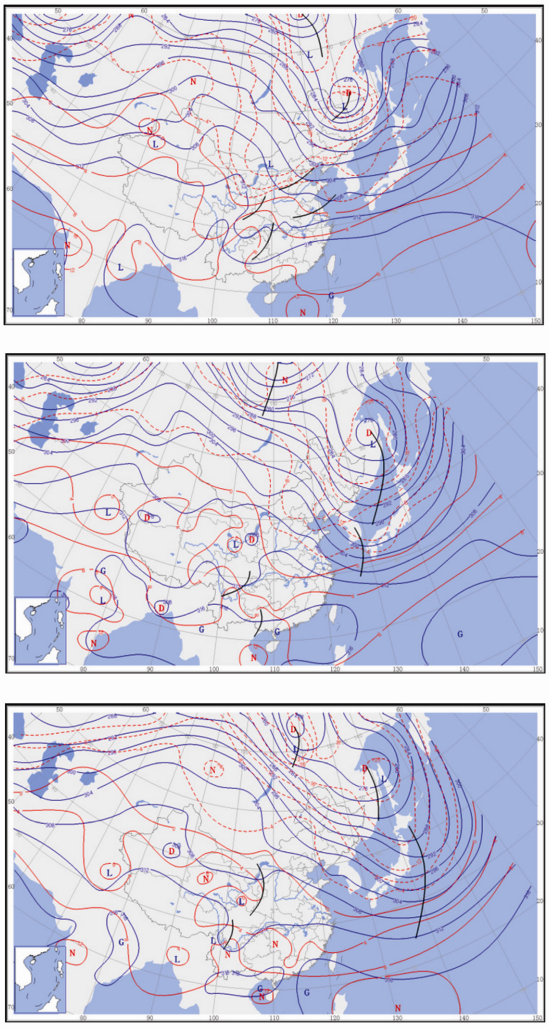

Fig. 3. Contours and isotherms for 700 hPa on April 10, 11 and 12, 2011.

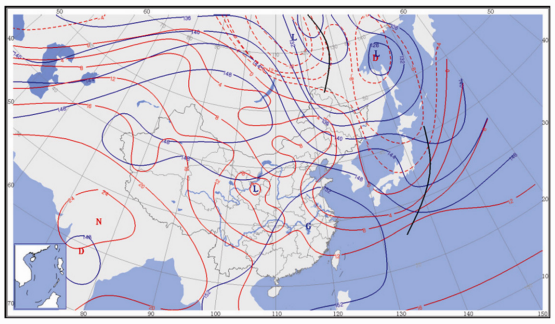

Fig. 4. Isobaric surface of 850 hPa on April 12, 2011.

This phenomenon also shows that ^{137}Cs, with a relatively long half-life, might be more significant in the second nuclear leakage from Fukushima (release of partial radioactive substances from the spent-fuel pool).

The observation in the fifth week (April 14–21, 2011) and later showed that although the ^{7}Be and ^{210}Pb concentrations are in relatively high levels, indicating there being influence of air masses from higher altitude and inland regions, respectively, the concentrations of artificial nuclides in air observed in Guiyang had evidently decreased with the change of air current (Fig. 2e and f) due to the dilution and a 4-half-life decay of ^{131}I and the diffusion dilution of ^{137}Cs.

4 Conclusions

The pollutants of nuclear leakage from Fukushima, Japan, in 2011 entered the southwestern region of China via two significant transport pathways, one on a global and the other on a regional scale.

(1) The first transport pathway is the atmospheric circulation transportation of the first wave of nuclear pollutants (^{131}I, ^{137}Cs, and ^{134}Cs) on a global scale from west to east, almost encircling the Earth once in about 10 days to two weeks, and the pollutants entered Guiyang from the upper level air of the regions of Middle Asia and northwester China during March 24–31, 2011. This fact shows that the nuclear pollutants that entered the atmosphere have a global effect through diffusion by atmospheric circulation transportation.

(2) The pollutants of nuclear leakage from Fukushima in the second wave arrived at Guiyang in the middle of April. The Fukushima nuclear leakage-polluted air mass moved southward through the northeast Asian weather system and was transported to a low-latitude region, and then arrived into the southwestern China region via air masses at a relatively low altitude under the influence of the southeastern warm and wet air current. This transportation was completed in a short time, and has significant characteristics of the eastern Asia regional system.

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References

Bolsunovsky A. and Dementyev D. (2011) Evidence of the radioactive fallout in the center of Asia (Russia) following the Fukushima Nuclear Accident [J]. *Journal of Environmental Radioactivity.* **102**, 1062–1064.

Clemenza M., Fiorini E., Previtali E., and Sala E. (2012) Measurement of airborne ^{131}I, ^{134}Cs and ^{137}Cs due to the Fukushima reactor incident in Milan (Italy) [J]. *Journal of Environmental Radioactivity.* **114**, 113–118.

CSN (2011) https://www.csn.es/images/stories/actualidad_datos/noticias/japon/tabla_deconcentraciones28marzo11i-131 [Z].

Dörr H. and Mūnnich K.O. (1990) ^{222}Rn flux and soil air concentration profiles in West-Germany—Soil ^{222}Rn as a tracer for gas transport in the unsaturated soil zone [J]. *Tellus Series B-Chemical and Physical Meteorology.* **42**, 20–28.

Diaz Leon J., Jaffe D.A., Kaspar J., Knecht A., Miller M.L., Robertson R.G.H., and Schubert A.G. (2011) Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA [J]. *Journal of Environmental Radioactivity.* **102**, 1032–1038.

Draxler R.R. and Rolph G.D. (2003) *HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory)* [Z]. Model access via NOAA ARL READY Website. NOAA Air Resources Laboratory, Silver Spring, MD, U.S.A. (Available online from http://www.arl.noaa.gov/ready/hysplit4.html).

Feely H.W., Larsen R.J., and Sanderson C.G. (1989) Factors that cause seasonal variations in Beryllium-7 concentrations in surface air [J]. *Journal of Environmental Radioactivity.* **9**, 223–249.

Holton J.R., Haynes P.H., and McIntyre E.M. et al. (1995) Stratosphere–troposphere exchange [J]. *Reviews of Geophysics*. **33**, 403–439.

Hong G.H., Hernández-Ceballos M.A., Lozano R.L., Kim Y.I., Lee H.M., Kim S.H., Yeh S.W., Bolívar J.P., and Baskaran M. (2012) Radioactive impact in South Korea from the damaged nuclear reactors in Fukushima: Evidence of long and short range transport [J]. *Journal of Radiological Protection*. **32**, 397–411.

Hsu S.C., Huh C.A., Chan C.Y., Lin S.H., Lin F.J., and Liu S.C. (2012) Hemispheric dispersion of radioactive plume laced with fission nuclides from the Fukushima nuclear event [J]. *Geophysical Research Letters*. **39**.

Huh C.A., Hsu S.C., and Lin C.Y. (2012) Fukushima-derived fission nuclides monitored around Taiwan: Free tropospheric versus boundary layer transport [J]. *Earth and Planetary Science Letters*. **319**, 9–14

IAEA (International Atomic Energy Agency) (2011) *Fukushima Nuclear Accident Update Log* (March 11, 2011) [Z]. http://www.iaea.org/newscenter/news/2011/fukushima 110311.html.

Keyser D. and Shapiro M.A. (1986) A review of the structure and dynamics of upper-level front zones [J]. *Monthly Weather Review.* **114**, 452–499.

Kim C.K., Byun J.I., Chae J.S., Choi H.Y., Choi S.W., Kim D.J., Kim Y.J., Lee D.M., Park W.J., Yim S.A., and Yun J.Y. (2012) Radiological impact in Korea following the Fukushima nuclear accident [J]. *Journal of Environmental Radioactivity.* **111**, 70–82.

Krishnaswami S. and Seidemann D.E. (1988) Comparative study of ^{222}Rn, ^{40}Ar, ^{39}Ar and ^{37}Ar leakage from rocks and minerals: Implications for the role of nanopores in gas transport through natural silicates [J]. *Geochimica et Cosmochimica Acta.* **52**, 655–658.

Lal D. and Peters B. (1967) Cosmic ray produced radioactivity on the Earth. In *Handbuch der Physik* (ed. Sitte K.) [M]. pp.551–612. Springer-Verlag, New York.

Lee H.N. (2004) Issues and challenges of using natural radionuclides as tracers for atmospheric studies. World Meteorological Organization (WMO)/Global Atmosphere Watch (GAW) Report No. 155, WMO TD No. 1201, 34–38.

Lee H.N. and Feichter J. (1995) An intercomparison of wet precipitation scavenging schemes and the emission rates of ^{222}Rn for the simulation of global transport and deposition of ^{210}Pb [J]. *Journal of Geophysical Research-atmospheres.* **100**, 23253–23270.

Lee H.N., Wan G.J., Zheng X.D., Sanderson C.G., Josse B., Wang S.L., Yang W., Tang, J., and Wang C.S. (2004) Measurements of ^{210}Pb and ^{7}Be in China and their analysis accompanied with global model calculations of ^{210}Pb [J]. *Journal of Geophysical Research-atmospheres.* **109**, D22203.

Lee S.H., Heo D.H., Kang H.B., Oh P.J., Lee J.M., Park T.S., Lee K.B., Oh J.S., and Suh J.K. (2013) Distribution of ^{131}I, ^{134}Cs, ^{137}Cs and ^{239,240}Pu concentrations in Korean rainwater after the Fukushima nuclear power plant accident [J]. *Journal of Radioanalytical and Nuclear Chemistry.* **296**, 727–731.

Liu Longbo, Wu Shan, Cao Junji, Xie Feng, Shi Quanlin, Zhang Changyun, Tang Hanbing, He Xiaobin, Zhang Rong, Chen Liyun, Wei Guanyi, Zhang Zhihong, Zhang Jiamei, and Dang Haijun (2013) Monitoring of atmospheric radionuclides from the Fukushima nuclear accident and assessing their impact on Xi'an, China [J]. *Chinese Science Bulletin.* **58**, 1585–1591.

Long N.Q., Truong Y., Hien P.D., Binh N.T., Sieu L.N., Giap T.V., and Phan N.T. (2012) Atmospheric radionuclides from the Fukushima Dai-ichi nuclear reactor accident observed in Vietnam [J]. *Journal of Environmental Radioactivity.* **111**, 53–58.

López-Pérez M., Ramos-López R., Nayra R., Perestelo X., Duarte-Rodriguez J.J., Bustos S., Alonso-Pérez E., Cuevas J., and Hernández-Armas J. (2013) Arrival of radionuclides released by the Fukushima accident to Tenerife (Canary Islands) [J]. *Journal of Environmental Radioactivity.* **116**, 180–186.

Lozano R.L., Hernández-Ceballos M.A., Adame J.A., Casas-Ruíz M., Sorribas M., San Miguel E.G., and Bolívar J.P. (2011) Radioactive impact of Fukushima accident in the Iberian Peninsula: Evolution and plume previous pathway [J]. *Environment International.* **37**, 1259–1264.

Manolopoulou M., Vagena E., Stoulos S., Ioannidou A., and Papastefanou C. (2011) Radioiodine and radiocesium in Thessaloniki, Northern Greece due to the Fukushima nuclear accident [J]. *Journal of Environmental Radioactivity.* **102**, 796–797.

Masson O., Baeza A., Bieringer J., Brudecki K., Bucci S., Cappai M., Carvalho E.P., Connan O., Cosma C., Dalheimer A., Didier D., Depuydt G., De Geer L.E., De Vismes A., Gini L., Groppi F., Gudnason K.,

Gurriaran R., Hainz D., Halldórsson O., Hammond D., Hnaley O., Holey K., Homoki Z.S., Ioannidou A., Isajenko K., Jankovic M., Katzlberger C., Kettunes M., Kierepko R., Kontro R., Kwakman P.J.M., Lecomte M., Leon Vinctro L., Leppanen A. P., Lind B., Lujaniene G., McGinnity P. , McMahon C., Malá H., Manenti S., Manolopoulou M., Mattila A., Mauring A., Mietelski J.W., Moller B., Nielsen S.P., Nikolic J., Overwater R.M.W., Pálsson S.E., Papastefanou C., Penev I., Pham M.K., Ponvinec P.P., Rameback H., Reis M.C., Ringer W., Rodriguez A., Rulík. P., Saey P.R.J., Samsonov V., Scholosser C., Sgorbati G., Silobritiene B.V., Soderstrom C., Sogni R., Solier L., Sonck M., Steinhauser G., Steinkopff T., Steinmann P., Stoulos S., Sýkora I., Todorovic D., Tooloutalaie N., Tositti L., Tschiersch J., Ugron A., Vagena E., Vargas A., Wershofen H., and Zhukova O. (2011) Tracking of airborne radionuclides from the damaged Fukushima Dai-Ichi nuclear reactors by European networks [J]. *Environmental Science & Technology.* **45**, 7670–7677.

Melgunov M.S., Pokhilenko N.P., Strakhovenko V.D., Sukhorukov F.V., and Chuguevskii A.V. (2012) Fallout traces of the Fukushima NPP accident in southern West Siberia (Novosi A.V. Birsk, Russia) [J]. *Environmental Science and Pollution Research.* **19**, 1323–1325.

Pittauerová D., Hettwig B., and Fischer H.W. (2011) Fukushima fallout in Northwest German environmental media [J]. *Journal of Environmental Radioactivity.* **102**, 877–880.

Qiao F.L., Wang G.S., Zhao W., Zhao J.C., Dai D.J., Song Y.J., and Song Z.Y. (2011) Predicting the spread of nuclear radiation from the damaged Fukushima Nuclear Power Plant [J]. *Chinese Science Bulletin.* **56**, 1890–1896.

Qiao Fangli, Wang Guansuo, Zhao Wei, Zhao Jiechen, Dai Dejun, Song Yajuan, and Song Zhenya (2005) Characteristics of U-pattern distribution of high-concentration ^{210}Pb in surface air at central Guizhou, China [J]. *Chinese Science Bulletin.* **50**, 1750–1755.

Turekian K.K., Nozaki Y., and Benninger L.K. (1977) Geochemistry of atmospheric radon and radon products [J]. *Annual Review of Earth and Planetary Sciences.* **5**, 227–255.

Wan Guojiang, Lee H.N., Wan Enyuan, Wang Shilu, Yang Wei, Wu Fengchang, Chen Jing'an, and Wang Changsheng (2008) Analyses of ^{210}Pb concentrations in surface air and in rain water at the central Guizhou, China [J]. *Tellus Series B—Chemical and Physical Meteorology.* **60**, 32–41.

Wan Guojiang., Zheng Xiangdong, Lee H.N., Bai Zhanguo, Wan Enyuan, Wang Shilu, Yang Wei, Su Fei, Tang Jie, Wang Changsheng, Huang Ronggui, and Liu Peng (2010) ^{210}Pb and ^{7}Be as tracers for aerosol transfers at center Guizhou, China: II. The explanation by weekly interval [J]. *Advances in Earth Science*. **25**, 492–504 (in Chinese with English abstract).

Zanis P., Schuepbach E., Gäggeler H.W., Hübener S., and Tobler L. (1999) Factors controlling beryllium-7 at Jungfraujoch in Switzerland [J]. *Tellus Series B—Chemical and Physical Meteorology.* **51**, 789–805.

Zheng Xiangdong, Wan Guojiang, Tang Jie, Zhang Xiaochun, Yang Wei, Lee H.N., and Wang Changsheng (2005) ^{7}Be and ^{210}Pb radioactivity and implications on sources of surface ozone at Mt. Waliguan [J]. *Chinese Science Bulletin.* **50**, 167–171.