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Impact of North Korean nuclear weapons test on 3 September, 2017 on inland China traced by ^{14}C and ^{129}I

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Abstract
Environmental impact of North Korea nuclear weapons testing on 3 Sept, 2017, is of key concern. In order to investigate whether there is radioactive leakage and whether it can be transported to inland China, ^{14}C and ^{129}I are determined in aerosol samples collected in a Chinese inland city before and after the test. Aerosol $\Delta^{14}C$ values before and after the test do not show any significant difference. In contrast, a four-fold increase of $^{129}I/^{127}I$ ratios was found after the test. The possible sources of ^{129}I in these atmospheric samples and the impact of the North Korea nuclear test are discussed.

Keywords North Korea underground nuclear weapons test · ^{14}C · ^{129}I · Environmental impact · Aerosol

Introduction

On 3 Sept, 2017, at 12:00 am local time, North Korea conducted the sixth nuclear test of a hydrogen bomb. This test is performed underground, which caused significantly detectable earthquake in both North Korea and the neighboring countries. The seismic record by University of Science and Technology of China and Chinese Academy of Sciences suggested that this explosion was located at 41.2982° N, 129.0742° E and the seismic magnitude was M_b 5.56 [1, 2]. This site refers to the Punggye-ri nuclear weapons test site, where the previous five nuclear weapons tests by North Korea were conducted. The estimated yield of this test is about 1.08×10^8 kg TNT, being the largest nuclear test among all six tests by North Korea, which is about 3–7.8 times bigger than that of "Fat Man" atomic bomb detonated over Nagasaki in 1945 [1].

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The emergency response of environmental monitoring was immediately initiated by China [3], South Korea and Japan [4] by monitoring the radioactivity level in the border area to North Korea using routine radioactive monitoring methods, but no measurable signals have been reported. The confirmation of the event is then considered more probable by investigating the radioactive signals directly released using a highly sensitive instrument, which will be also useful for the evaluation of any possible environmental impact.

^{14}C, with half-life of 5730 years, is produced as a neutron activation product through reactions of $^{14}N(n, p)^{14}C$, $^{13}C(n, \gamma)^{14}C$, $^{17}O(n, \alpha)^{14}C$ and $^{15}N(n, d)^{14}C$ in nuclear weapons tests and nuclear reactors, and often released to the atmosphere as gaseous forms (e.g. CO_2, CO). ^{129}I, a radioisotope of iodine with half-life of 15.7×10^6 years, is produced as a fission product of ^{235}U and ^{239}Pu, and released to the atmosphere as gaseous form (e.g. I_2) from nuclear weapons tests, nuclear accidents and nuclear spent fuel reprocessing. Levels of ^{14}C and ^{129}I in the environment have been raised by a few orders of magnitude in 1950–1980 due to intensive atmospheric nuclear weapons tests [5, 6]. ^{14}C and ^{129}I are two important radionuclides released from human nuclear activities such as nuclear weapons tests, nuclear accidents, spent nuclear fuel reprocessing plants, and therefore can be applied as excellent tracers for monitoring nuclear weapons testing and nuclear accidents/leakage. Accelerator mass spectrometry (AMS) is a very sensitive method for detecting long-lived radionuclides,

especially ^{14}C and ^{129}I, down to nBq level, therefore it can be used to detect very small releases of ^{14}C and ^{129}I to the environment from nuclear activities.

With highly sensitive AMS measurement of ^{129}I and ^{14}C in air samples, this work aims to explore the possible releases of radioactive substances from the sixth nuclear weapons test of North Korea, and to estimate its impact on environmental radioactivity in the Chinese inland area.

Experimental

Sampling

The aerosol samples were collected by a large volume sampler on glass fiber filter at the Xi'an AMS center (34.2069°N, 109.0000°E) in Xi'an, China (Fig. 1). The sampling flow rate is set as 1.5 m^3 min^{-1}, and sampling duration is 24 h for one glass fiber filter (Wuhan Tianhong instrument Ltd. China) with dimension of 200 mm × 220 mm and the sampling efficiency over 99.9% at the flow rate operated in this work. The total air volume of one filter is about 2000 m^3 under standard condition (0 °C and 101.325 kPa).

The Punggye-ri nuclear weapons test site (41.2982°N, 129.0742°E) is located in a mountain terrain, Kilju County, North Hamgyong Province in northeastern North Korea. The sampling site is approximately 2200 km southwest of the test site. The samples collected before and immediately after the North Korea sixth nuclear weapons test were analyzed for radioactive ^{14}C and ^{129}I.

Preparation of aerosol samples and AMS measurement for ^{14}C

The aerosol samples were cut into small pieces (< 2 × 2 mm), and put into a quartz tube for separation of carbon using pyrolysis. Carbon in aerosols was oxidized to carbon dioxide using high purity oxygen at 900 °C. Carbon dioxide collected during pyrolysis process were reduced to graphite using zinc metal powder in the presence of iron as a catalyst. The ^{14}C content in the prepared graphite was measured using 3MV AMS (HVEE, the Netherlands) in the Xi'an AMS Center, The measurement uncertainty of $^{14}C/^{12}C$ atomic ratio for the samples is better than 0.2% [7]. The minimum measurable ratio of $^{14}C/^{12}C$ is 3.1×10^{-16} [8].

The ^{14}C level in the aerosol samples is expressed as $\Delta^{14}C$, which is the deviation (in ‰) of the $^{14}C/^{12}C$ ratio of a sample with respect to modern carbon (standard sample) after correcting for the age and isotopic fractionation [9].

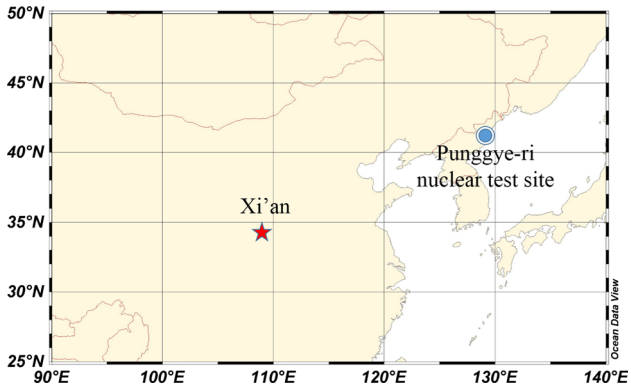

Fig. 1 Map showing the Punggye-ri nuclear test site of North Korea underground nuclear test on 3 Sept, 2017 (blue circle) and aerosol sampling location in Xi'an, China (red pentagram). (Color figure online)

Preparation of aerosol samples and determination of ^{129}I and ^{127}I

The aerosol samples were cut into small pieces (< 5 × 5 mm), and put into a quartz boat. ^{125}I was added as a chemical yield tracer. Iodine was separated from the aerosol filters using a combustion method by being oxidized to molecular iodine with oxygen at 800 °C in a tube furnace [10]. The released iodine was trapped into a mixture solution of 0.5 mol L^{-1} NaOH and 0.02 mol L^{-1} $NaHSO_3$. An aliquot of solution (1.0 mL) was taken for determination of ^{127}I using ICP-MS (Agilent 8800, USA) after 20-fold dilution with 0.5% tetramethylhydroxylamine. Cs^+ (CsCl) was used as an internal standard in the ICP-MS measurement of iodine. One mL of the solution was taken for measurement of ^{125}I by a NaI gamma counter (Model FJ2021, Xi'an Nuclear Instrument Factory, China) for calculation of chemical yield. Due to extremely small amounts of ^{129}I and ^{127}I in the trapping solution, addition of carrier is necessary to make sure a stable ^{127}I current in AMS measurement and operable amount of AMS target. Addition of iodine carrier into aerosol samples would decrease $^{129}I/^{127}I$ atomic ratio in the AMS target. In order to determine low-level ^{129}I concentration in Xi'an aerosols, amount of iodine added should be as low as possible, and chlorine is added to make sure the amount of target is sufficient. Based on the previous work [10], 0.2 mg ^{127}I carrier and 0.5 mg chloride were added to the remaining solution, and then nitric acid was added to pH 2. $AgNO_3$ solution was added to the solution to precipitate iodine and chloride. The formed AgI–AgCl precipitate was washed with 3% HNO_3 once and deionized water twice, then separated by centrifugation. After dried at 70 °C, the AgI–AgCl precipitate was well mixed with Nb metal powder (99.9%, 325 mesh, Alfa Aesar, USA) in a mass ratio of 1:5 and pressed into a cooper target holder. Chemical yields of iodine are over 80% in the whole procedure of sample preparation. ^{129}I in the AgI–AgCl precipitate was measured using a 3MV AMS in the Xi'an

AMS Center [10]. The procedural blank of $^{129}I/^{127}I$ was prepared using a blank glass fiber filter with the same procedure as the samples, and determined to be $< 2 \times 10^{-12}$ [11]. $^{129}I/^{127}I$ standard was prepared using NIST-SRM 4949c by dilution using ^{127}I carrier solution (prepared using iodine crystal with $^{129}I/^{127}I$ atomic ratio commonly considered to be less than 2×10^{-14}) in the same form as sample (AgI–AgCl precipitate), which is used for calibration of the measured $^{129}I/^{127}I$ ratio by AMS.

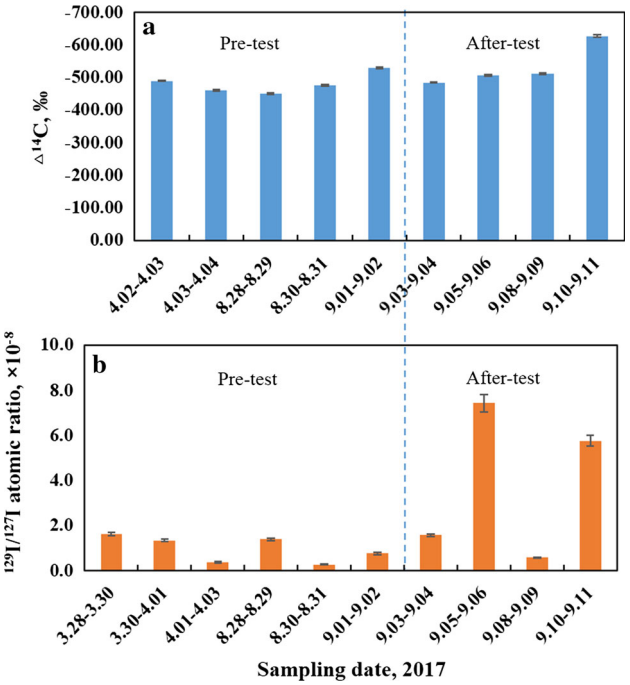

Fig. 2 Comparison of $\Delta^{14}C$ and $^{129}I/^{127}I$ levels in aerosols from Xi'an, China before and after the North Korea sixth nuclear weapons test

Results and discussion

Levels of $\Delta^{14}C$ and ^{129}I in the aerosols

The pre- and post-test aerosol samples were analyzed for both ^{14}C and ^{129}I. $\Delta^{14}C$ levels range from − 450‰ to − 530‰ for pre-test aerosols, while from − 485‰ to − 627‰ for post-test samples (Fig. 2a). No significant difference of $\Delta^{14}C$ ($p = 0.22$) between pre-test and post-test was measured. However, $\Delta^{14}C$ values in these aerosol samples are significantly lower than those observed in atmospheric CO_2 samples collected all over China with $\Delta^{14}C$ ranging from − 20‰ to − 30‰ from January to December, 2014 [12].

Concentrations of stable iodine (^{127}I) in aerosol were measured to be 1.2–6.0 ng m^{-3} (Table 1), which fell well within the common level of iodine in terrestrial aerosols [13]. ^{129}I concentrations range from 0.3×10^5 atoms m^{-3} to 4.6×10^5 atoms m^{-3} (Table 1).

The measured $^{129}I/^{127}I$ atomic ratios were in the range of $(0.3–1.6) \times 10^{-8}$ in April and August before the North Korean sixth nuclear weapons test, and $(0.6–7.4) \times 10^{-8}$ in early September after the test (Fig. 2b). It is reported that ^{129}I concentrations is in the order of 10^6–10^7 atoms/m^3 in northern Europe [14, 15], and much lower in Japan, Brazil, Seville and Southern Spain within a range of 10^4–10^5 atoms/m^3 [16–18]. The ^{129}I concentrations in Xi'an aerosols are comparable to those in Asia, America and southern Europe, while 1–2 orders of magnitude lower than those in northern Europe.

It is worthy to note that the average $^{129}I/^{127}I$ atomic ratio of post-test aerosol samples is 4.7 times higher than that of pre-test samples (Fig. 2b). High $^{129}I/^{127}I$ ratios were observed in two aerosol samples collected on 5–6 Sept and 10–11 Sept. However, statistical analysis suggests that there is no significant difference between pre-test $^{129}I/^{127}I$

Table 1 Analytical results of ^{127}I, ^{129}I concentrations and $^{129}I/^{127}I$ atomic ratios in Xi'an aerosols

No.	Sampling date 2017	$\Delta^{14}C$, ‰	Sampling date 2017	^{127}I conc. ng m^{-3}	^{129}I conc. $\times 10^5$ atoms m^{-3}	$^{129}I/^{127}I$ atomic ratio $\times 10^{-8}$
1			28–30 Mar	5.8 ± 0.2	4.5 ± 0.2	1.62 ± 0.07
2	2–3 Apr	− (489.37 ± 1.74)	30 Mar–01 Apr	2.8 ± 0.1	1.8 ± 0.1	1.34 ± 0.04
3	3–4 Apr	− (460.93 ± 1.88)	1–3 Apr	6.0 ± 0.1	1.1 ± 0.1	0.38 ± 0.04
4	28–29 Aug	− (450.06 ± 1.75)	28–29 Aug	1.2 ± 0.1	0.8 ± 0.1	1.38 ± 0.06
5	30–31 Aug	− (476.62 ± 2.09)	30–31 Aug	2.50 ± 0.1	0.3 ± 0.1	0.26 ± 0.01
6	1–2 Sept	− (529.94 ± 1.99)	1–2 Sept	3.5 ± 0.1	1.2 ± 0.1	0.75 ± 0.05
7	3–4 Sept	− (484.53 ± 1.88)	3–4 Sept	3.3 ± 0.1	2.5 ± 0.1	1.57 ± 0.05
8	5–6 Sept	− (505.84 ± 1.95)	5–6 Sept	2.1 ± 0.1	7.3 ± 0.3	7.43 ± 0.39
9	8–9 Sept	− (511.20 ± 2.57)	8–9 Sept	4.9 ± 0.1	1.3 ± 0.1	0.57 ± 0.03
10	10–11 Sept	− (627.01 ± 3.06)	10–11 Sept	1.7 ± 0.1	4.6 ± 0.1	5.75 ± 0.24

Uncertainties presented here is an extended uncertainty with a coverage of k = 1

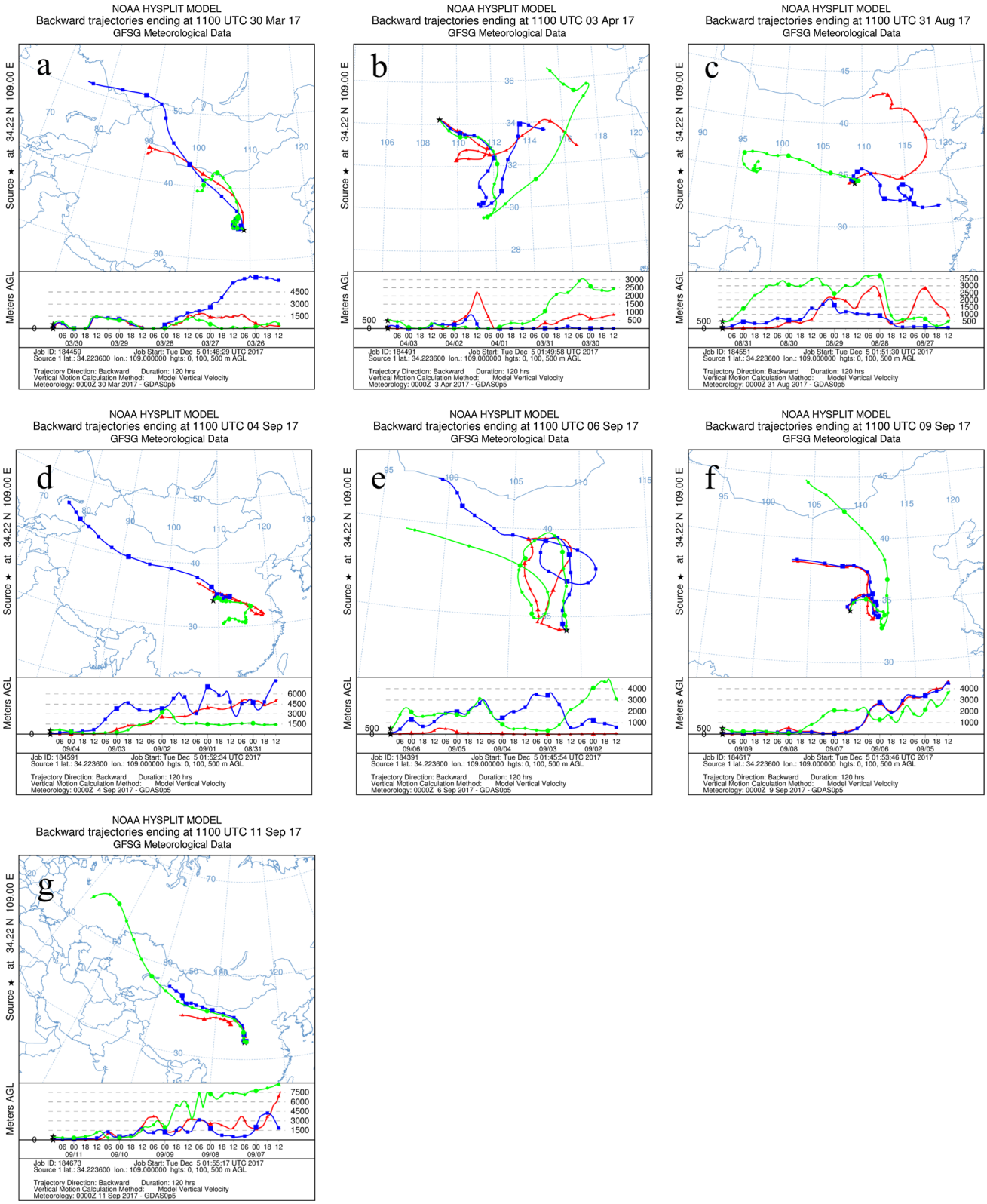

Fig. 3 Back trajectories analysis of air masses in Xi'an from 30 Mar to 11 Sept, 2017. The length of back trajectory is 120 h. Colors indicate the height of trajectory endpoint (0, 100, 500 m). Before North Korean nuclear weapons test on 3 Sept 2017 including **a** 26–30 Mar, **b** 31 Mar–3 Apr, **c** 27–31 Aug, and after the test including, **d** 31 Aug–4 Sept, **e** 2–6 Sept, **f** 5–9 Sept, **g** 7–11 Sept. (Color figure online)

ratios and those post-test ones ($p = 0.16$) because of high variation of $^{129}I/^{127}I$ ratios in these samples.

Sources of ^{14}C and ^{129}I in aerosols from Xi'an

No significantly statistical difference of ^{14}C concentrations in aerosols was noticed before and after North Korea sixth nuclear weapons test. In fact, the significantly low $\Delta^{14}C$ values (from − 450‰ to − 627‰) measured in these aerosol samples indicate that these aerosols contained high ^{14}C-depleted "old carbon" mainly from combustion of fossil fuel [19], which greatly reduces the atmospheric $\Delta^{14}C$ [12]. Therefore, the $\Delta^{14}C$ results suggest the ^{14}C signal of North Korea nuclear test, if it exists, would be negligible or completely masked by "old carbon" signals.

The increased $^{129}I/^{127}I$ ratios in the aerosol samples collected immediately after the nuclear weapons test was observed (Fig. 2), which is probably originated from the North Korean sixth nuclear weapons test. However, this is not in agreement with the results of radiation monitor along the board areas between China and North Korea, where no increased absorption dose rate was measured in air [3, 20].

To confirm the source of ^{129}I in the aerosol samples immediately after North Korean nuclear weapons test on 3 Sept, 2017, the pathway of the air masses in Xi'an in the periods for collection of these samples was investigated using transport and dispersion modelling (HYSPLIT) [21] (Fig. 3). Back trajectories analysis shows that the air masses at the sampling site in Xi'an, China during the sampling periods are dominantly transported from direction of west and northwest, and in a small portion from east and southeast but within the territory of China. Therefore, the high $^{129}I/^{127}I$ ratios and ^{129}I concentrations in these aerosol samples should be related to the air masses from west Asia and Europe during 26–30 Mar, 31 Aug–4 Sept and 10–11 Sept. Because the huge amount of ^{129}I have been released from the nuclear fuel reprocessing plants at Sellafield, UK and La Hague, France, level of aerosol $^{129}I/^{127}I$ ratios in Europe has been increased to $(15.6–102.0) \times 10^{-8}$ [15], which is 1–2 orders of magnitude higher than those in Xi'an. Consequently, the increased ^{129}I level should be attributed to ^{129}I-rich air masses which carry gaseous released ^{129}I and re-emission of liquid discharged ^{129}I from the European nuclear fuel reprocessing plants (Sellafield, UK and La Hague, France) [22], and it is unlikely related to the North Korea nuclear weapons test on 3 Sept, 2017.

Radiation impact of North Korea sixth nuclear test on Chinese inland

The analytical results of ^{14}C and ^{129}I in aerosol samples indicate that no radioactive substance from the North Korean sixth nuclear weapons test on 3 Sept, 2017 was detected in inland China, which agrees with the monitoring results along the boards areas between China and North Korea [3, 4].

The forward trajectory analysis shows that the air masses moved from the Punggye-ri nuclear test site on 3 Sept to north and northeast direction (Fig. 4). It indicates that even if there is any leakage of radioactive substances from the nuclear test, the radioactive pollutants should be transported northwards along Chinese northeast border (Jilin and Heilongjiang provinces) to southeast Russia, and should not be possible to reach Xi'an, China. It is also evident that the latest nuclear test would not cause an impact on the environment and human health in China.

Up to now, no radioactive substances has been detected in neighboring countries of North Korea (i.e. Japan, South Korea). However, owing to the collapse of nuclear test tunnel in late September [20], new radiation leakage through the mountain cracks is quite possible, which have attracted much attention. We continue to collect air samples and would further investigate its impact on Chinese environment, especially along the air mass movement directions.

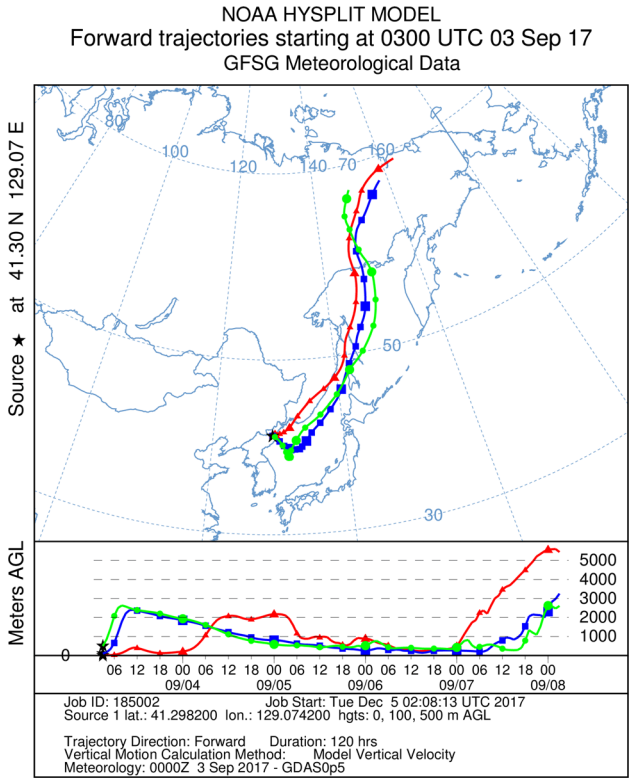

Fig. 4 Forward trajectory in 3–8 Sept, 2017 at Punggye-ri nuclear test site on 3 Sept, 2017

Conclusions

Based on the results obtained in this work and discussion above, it can be concluded that: (1) ^{14}C levels in the aerosols in Xi'an, China do not show any signals from the North Korean sixth nuclear weapons test. On the contrary, a much lower ^{14}C level was observed in the sampling period compared to ^{14}C level of atmospheric CO_2 all over China, which should be attributed to dilution effect of ^{14}C-depleted "old carbon" from combustion of fossil fuel; ^{14}C signals in a large city is expected to be significantly masked by the local dead carbon emission, thus it can be expected to be ineffective for monitoring purpose of nuclear safety. (2) A 4.7-fold increase of $^{129}I/^{127}I$ ratios were measured in the post-test samples compared to the pre-test ones. The back and forward trajectory analysis shows that the increased $^{129}I/^{127}I$ ratios in the aerosols collected immediately after the North Korean nuclear weapons test are attributed to the ^{129}I-enriched air masses contaminated by emission of radioiodine from the European nuclear reprocessing plants. The results of this work suggest no measurable leakage of radioactive substances in inland China from the North Korea sixth underground nuclear test, therefore no radiation impact to the environment and human health. In addition, sample collection along the air mass movement directions of a nuclear test event should be taken into consideration for future monitoring studies of such events.

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