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

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Impact of the North Korean nuclear weapon test on 3rd September 2017 in inland China traced by long-lived radionuclides (^{14}C and ^{129}I)

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Abstract

On 3rd Sept., 2017, the sixth nuclear weapons test detonated by North Korea at the Punggye-ri nuclear test site has attracted extensive attention, of which radioactive hazard releases are the key concern, in particular in its neighboring countries including China. The released radioactive substances might be quickly dispersed to a large area through atmospheric processes. Aerosol samples were collected in Xi'an, a Chinese inland city, and analyzed for two volatile and long-lived radionuclides, ^{14}C and ^{129}I, using highly sensitive accelerator mass spectrometry, to investigate possible leakage and level of radioactive substances from this nuclear weapons test. Values of $\Delta^{14}C$ in the post-test samples (-485‰ to -627‰) does not show any significant difference with those in the pre-test samples (-450‰ to -530‰), indicating no visible releases of radioactive ^{14}C from this nuclear weapons test. Compared to those observed in normal atmospheric CO_2 in China (-20‰ to -30‰), the highly negative values of $\Delta^{14}C$ in these aerosol samples can be attributed to the ^{14}C-depleted "old carbon" from combustion of fossil fuel in urban areas in Xi'an. A four-fold increase of $^{129}I/^{127}I$ ratios of (0.6-7.4) $\times 10^{-8}$ in the post-test samples than the pre-test ones ((0.4-1.6) $\times$ 10^{-8}) were observed. 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, Carbon-14, Iodine-129, environmental radioactivity, aerosol

Introduction

On 3rd Sept., 2017, at 12:00 am local time, North Korea conducted the sixth nuclear test of a hydrogen bomb. The seismic record by University of Science and Technology of China and Chinese Academy of Sciences suggested that this explosion is located at 41.2982° N, 129.0742° E and the seismic magnitude was M_b 5.56 (Wen, 2017; Zhao et al., 2017). This site refers to the Punggye-ri nuclear weapon test site, where the previous five nuclear weapons tests were conducted by North Korea. The estimated yield of this test is about 108 kt TNT, being the largest nuclear test among all 6 tests by North Korea, which is about 3 – 7.8 times bigger than that of "Fat Man" atomic bomb detonated over Nagasaki in 1945 (Wen, 2017).

The emergency response of environmental monitoring was immediately initiated by China (Ministry of Environmental Protection of the People's Republic of China, 2017), South Korea and Japan (The Nuclear Regulation Authority of Japam, 2017) by monitoring the radioactivity level in the border area to North Korea using routine radioactive monitoring methods, but no measurable signals have been reported. It is expected to confirm this event by investigating the radioactive signal directly released from this test by using a highly sensitive method, which will be also useful for evaluation of the possible environmental impact.

^{14}C with a half-life of 5730 years, is produced as a neutron activation product through reactions of $^{14}N(n, p)^{14}C$, $^{13}C(n, g)^{14}C$, $^{16}O(n, \alpha)^{14}C$ and $^{15}N(n, d)^{14}C$) in nuclear weapon tests and nuclear reactors, and often released to the atmosphere as gaseous form (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 releases to the atmosphere as gaseous form (e.g. I_2) from nuclear weapon tests, nuclear accidents and nuclear spent fuel reprocessing. ^{14}C and ^{129}I level in the environment have been raised by a few orders of magnitude in 1950-1980 due to intensive atmospheric nuclear weapons tests (Hou et al., 2009; Hua et al., 2013). ^{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 determination of 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.

Using a highly sensitive tool of AMS measurement for ^{129}I and ^{14}C in air samples, this work aims to explore the possible releases of radioactive substances from the 6th nuclear weapon test of North Korea, and estimate its impact on environmental radioactivity in the Chinese inland area.

Method

Sampling

The aerosol samples were collected by a large volume sampler (flow rateof 1.5 m^3/min) on glass fiber filter at the Xi'an AMS center (34°13'25"N, 109°0'0"E) in Xi'an, China (Fig. 1). The sampling site is approximately 2200 km southwest of the Punggye-ri nuclear weapon test site (41.2982° N, 129.0742° E) in mountain terrain, Kilju County, North

Hamgyong Province in northeastern North Korea. The samples collected before and immediately after the North Korea sixth nuclear weapon test were analyzed for radioactive ^{14}C and ^{129}I.

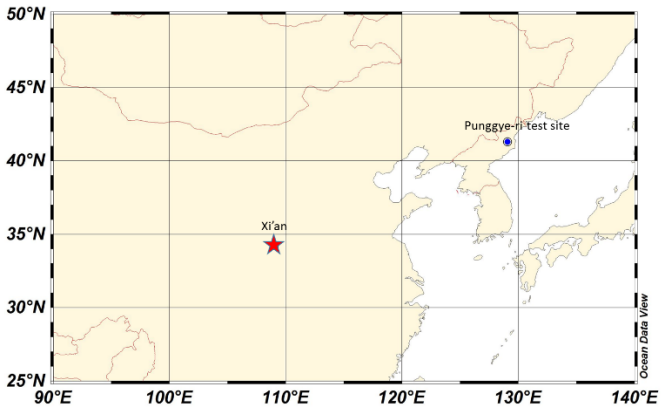

Figure 1. Map showing the Punggye-ri nuclear test site of the North Korea underground nuclear test on 3rd Sept., 2017 and aerosol sampling location in Xi'an, China

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 ℃. 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% (Cheng et al., 2013). The minimum measurable ratio of $^{14}C/^{12}C$ is 3.1×10^{-16} (Zhou et al., 2006).

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 (Stuiver and Polach, 1977).

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 chemical yield tracer. Iodine was separated from the aerosol filter using a combustion method by being oxidized to molecular iodine with oxygen at 800 ℃ in a tube furnace (Hou et al., 2010). The released iodine was trapped into a mixture solution of 0.5 mol/L NaOH and 0.02 mol/L $NaHSO_3$. An aliquot of solution (1.0 ml) was taken for determination of ^{127}I using ICP-MS (Agilent 8800, USA) after 10-fold dilution with deionized water of 18.2 MΩ cm, produced by a Cascada^{TM} Lab Water System (Pall Life Sciences, USA). 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). 0.2 mg ^{127}I carrier and 0.5 mg chloride were added to the remained solution, and then nitric acid was added to pH 2. $AgNO_3$ solution was added to the solution, and the formed AgI-AgCl precipitate was separated by centrifuge. After dried at 70°C, the AgI-AgCl precipitate was mixed with Nb metal powder (99.9%, 325 mesh, Alfa Aesar, USA) in mass ratio of 1:3 and pressed into a cooper target holder. ^{129}I in the AgI-AgCl precipitate was measured using 3MV AMS in the Xi'an AMS Center (Hou et al., 2010). The procedural blank of $^{129}I/^{127}I$ was prepared using a blank glass fiber filter with the same procedure as for the samples, and determined to be $<2\times10^{-13}$ (Zhou et al., 2010). $^{129}I/^{127}I$ 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

of less than 2×10^{-14}) in the same form as sample (AgI-AgCl precipitate), which is used for calibration/correction of the measured $^{129}I/^{127}I$ ratio by AMS.

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‰ (Niu et al., 2016).

Concentrations of stable iodine (^{127}I) in aerosol were measured to be 1.2-6.0 ng/m^3 (Table 1), which fell well in the common level of iodine in terrestrial aerosols (Saiz-Lopez et al., 2012). ^{129}I concentrations range from 0.3×10^5 atoms/m^3 to 4.6×10^5 atoms/m^3 (Table 1).

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

Sample Name	Sampling date, 2017	^{127}I, ng/m^3		^{129}I, $\times10^5$ atoms/m^3		$^{129}I/^{127}I$ atomic ratio, $\times10^{-8}$	
		Conc.	Uncertainty	Conc.	Uncertainty	Ratio	Uncertainty
AE002	3.28-3.30	5.8	0.2	4.5	0.2	1.62	0.07
AE003	3.30-4.01	2.8	0.1	1.8	0.1	1.34	0.04

AE004	4.01-4.03	6.0	0.1	1.1	0.1	0.38	0.03
AE005	8.28-8.29	1.2	0.1	0.8	0.1	1.38	0.06
AE006	8.30-8.31	2.50	0.1	0.3	0.1	0.26	0.01
AE007	9.01-9.02	3.5	0.1	1.2	0.1	0.75	0.05
AE008	9.03-9.04	3.3	0.1	2.5	0.1	1.57	0.05
AE009	9.05-9.06	2.1	0.1	7.3	0.3	7.43	0.39
AE010	9.08-9.09	4.9	0.1	1.3	0.1	0.57	0.03
AE011	9.10-9.11	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

The measured $^{129}I/^{127}I$ atomic ratios were in the range of (0.4-1.6) × 10^{-8} in April and August before the North Korean 6th nuclear weapon test, and (0.6-7.4) ×10^{-8} in early September after the test (Fig. 2b). This range is comparable to those in aerosols collected in Japan and Brazil, while 1-2 orders of magnitude lower than those in European (Englund et al., 2010; Santos et al., 2005; Toyama et al., 2013).

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

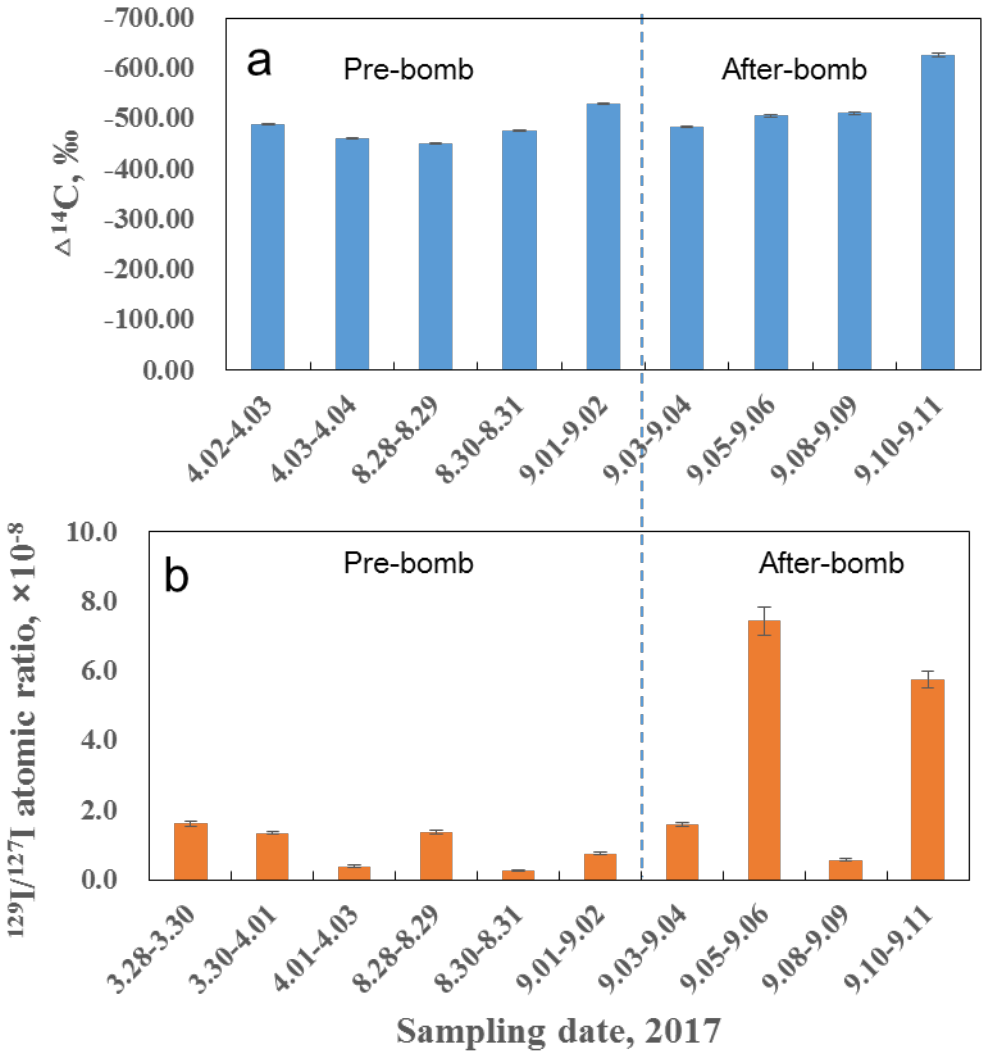

Figure 2. Comparison of $\Delta^{14}C$ and $^{129}I/^{127}I$ levels in aerosols from Xi'an, China before and after the North Korea 6th nuclear weapon test

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

No significantly statistical difference of ^{14}C concentrations in aerosols was noticed between before and after North Korea 6th nuclear weapon 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 (Paull et al., 1989) , which greatly reduce the atmospheric $\Delta^{14}C$ (Niu et al., 2016). 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 weapon test was observed (Fig. 2), which is probably originated from the North Korean 6th 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 (Ministry of Environmental Protection of the People’s Republic of China, 2017; Persio, 2017).

To confirm the source of ^{129}I in the aerosol samples immediately after North Korean nuclear weapon test on 3rd 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) (Draxler and Rolph, 2003) (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 26th - 30th Mar., 31st Aug. - 4th Sept., and 10th-11th Sept. Due to the huge amount of ^{129}I has being 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) $\times10^{-8}$ (Zhang et al., 2016), 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) (Zhang et al., 2017), and unlikely related to the North Korea nuclear weapon test on 3rd Sept. 2017.

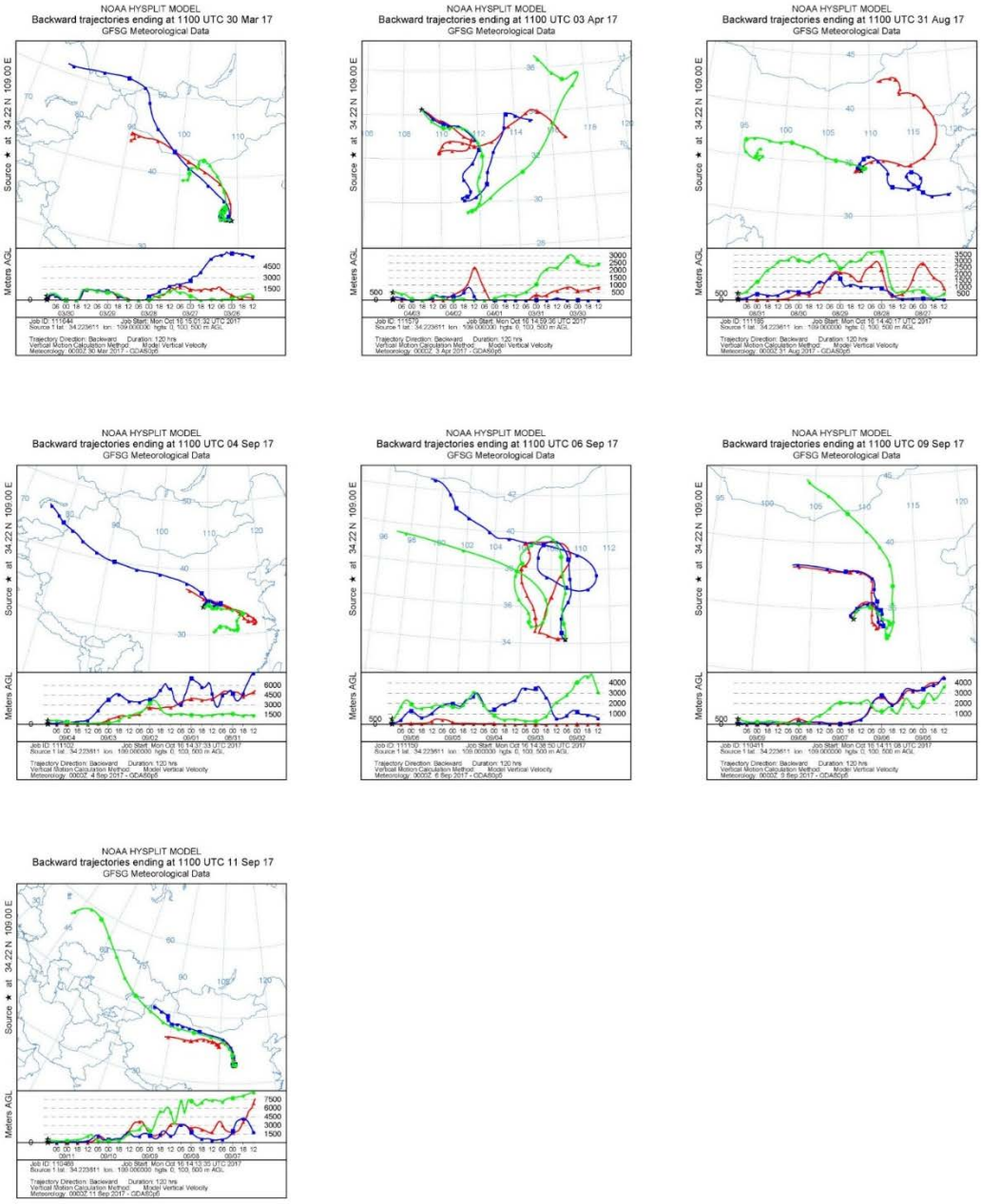

Figure 3. Back trajectories analysis of air masses in Xi'an from 30th Mar. to 11th Sept. 2017.

Radiation impact of North Korea 6th 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 6th nuclear weapons test on 3rd Sept. 2017 was detected in inland China, which agrees with the monitoring results along the boards areas between

China and North Korea (Ministry of Environmental Protection of the People's Republic of China, 2017; The Nuclear Regulation Authority of Japam, 2017).

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

Up to now, no radioactive substances have 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, 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.

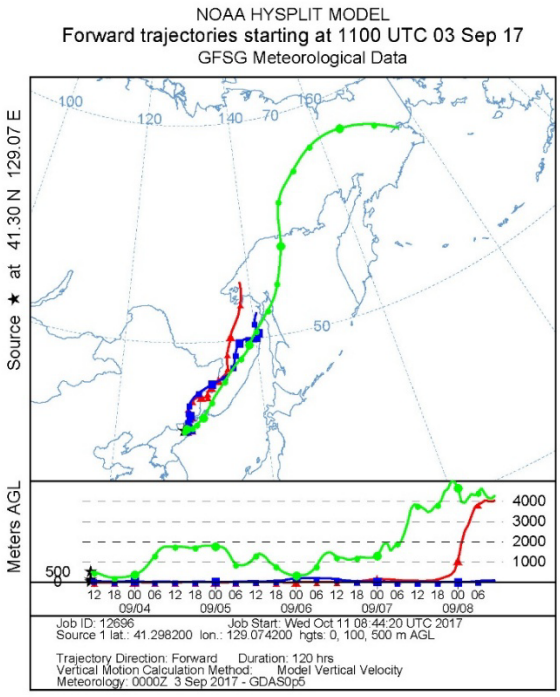

Figure 4. Forward trajectory in Punggye-ri nuclear test site on 3rd Sept., 2017

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 6th nuclear weapon 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; (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 show that the increased $^{129}I/^{127}I$ ratios in the aerosols collected immediately after the North Korean nuclear weapon 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 released from the North Korea 6th underground nuclear test, therefore no radiation impact to the environment and human health.

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