The fallout from Fukushima Daiichi nuclear accident profiles a new dating reference in ice and comparison with the Chernobyl accident

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Abstract

Absolute-agedating horizons play a pillar role in the reconstruction of an ice core chronology. In the modern era, these have included the global fallout from massive volcanic eruptions, atmospheric and marine thermonuclear weapons testing and nuclear accidents. After the occurrence of the Fukushima Daiichi nuclear accident (FDNA) on March 11 2011, the simulation of the radioactivity from the FDNA by a dispersion model (HYSPLIT) shows that the nuclides reached the study area in late March, consistent with the ground measurements in Xi’an, Lanzhou and Urumqi. To investigate the deposition of radioactivity resulting from the FDNA, we collected snowpack samples from four glaciers (i.e. Glacier No. 1, Glacier No. 72, Qiyi and Shiyi glaciers, respectively) in northwestern China and analysed them for total β activity (TBA). The measured TBA in the FDNA layers were increased by two to four times, compared with the averages in the non-FDNA layers. We revisited Glacier No. 1 in 2018 and studied a much deeper snow-pit profile for the TBA, seven years after the first-time investigation into a relatively shallow snow pit in 2011. The TBA concentrated in a dust layer and became more significant in 2018 compared to that in 2011. We compared the TBA in Glacier No. 1 with that in the Muztagata glacier from the Chernobyl accident in 1986, and the depositions of radioactivity in the two High-Asian glaciers were comparable. We conclude that the FDNA formed a distinctly new lasting reference in the snow, which could help date the snow and ice in the Northern Hemisphere.

1. Introduction

Radioactive horizons in glacial ice are an essential dating tool for establishing and validating ice core chronologies (Ambach et al 1987, Pourech et al 1988, Delmas et al 2004). These horizons are even more vital for Alpine glacier studies where the surface and shallow layers of the glaciers may be perturbed by melt and ablation (Haebelri et al 1988). In some extreme cases, modern upper sections of ice may be missing (Kehrwald et al 2008). Thus determining the presence or absence of a known horizon is the first step for developing a depthage relationship for an ice core.

The Fukushima Daiichi Nuclear Accident (FDNA) began on March 11, 2011, and resulted in a month-long discharge of radioactive materials into the atmosphere and ocean and soils (Chino et al 2011, IAEA 2011, Yasunari et al 2011, Behrens et al 2012). The environmental and health consequences from the FDNA fallout continue to concern the public (Normile 2011, Martin et al 2019). The total amount of radioactive substance released into the atmosphere has been initially estimated to be $\sim 1.5 \times 10^{17}$ Bq of $^{131}$I and $1.2 \times 10^{16}$ Bq of $^{137}$Cs (Commission 2011). Radionuclides from the event were detected in the atmosphere over much of the Northern Hemisphere.
Figure 1. The map of the study area using the World_Hillshade data provided by the ArcGisMapServer integrated into the QGIS v3.10 software (Team, 2020), where the global view is shown in the top-right corner, marked with Fukushima, Chernobyl and the study area. The red diamonds denote the sampling glaciers in this study; the green diamond denotes a reference ice core from the Muztagata Glacier (Tian et al. 2007); and the blue diamonds denote the four reference glaciers in Wang et al. (2015). The three related cities in this study are Urumqi, Lanzhou and Xi’an, respectively.

Table 1. Sampling sites, dates and other relevant information, where ‘weq’ denotes ‘water equivalent’.

| Mountain | Glacier             | Geographic location | Sampling date | Depth (m weq) |
|----------|---------------------|---------------------|---------------|--------------|
| Tianshan | Glacier No.72       | 41°48′ N, 80°12′ E, 4580 m | 26 May 2011 | 0.45         |
| Tianshan | Glacier No.1        | 43°06′ N, 86°48′ E, 4130 m | 22 May 2011 | 0.80         |
|          |                     |                     | 22 May 2011 | 0.80         |
|          |                     |                     | 30 August 2018 | 1.60        |
| Qilian   | Qiyi Glacier        | 39°15′ N, 97°45′ E, 4800 m | 7 May 2011 | 0.39         |
| Qilian   | Shiyi Glacier       | 38°12′ N, 99°52′ E, 4650 m | 4 May 2011 | 0.31         |

Hemisphere (Hosoda et al. 2011, Commission 2011, Masson et al. 2011, Steinhauser et al. 2014). The Japanese Nuclear and Industrial Safety Agency (JNISA) raised the FDNA to Level 7, the worst on the International Nuclear and Radiological Event Scale (INES); however, the agency also estimated that the release of radioactive material into the atmosphere was approximately one-tenth that of the Chernobyl accident, the only other accident to have an IAEA Level-7 rating (IAEA 2011).

The radioactive fallout from the FDNA has been detected in snow and ice samples in the mountain and polar regions. Two snowpit profiles in Greenland in 2012 showed the distinct $^{137}$Cs peaks sourced from the FDNA (Ežerinskis et al. 2014). Gabrielli et al. (2016) suggested that the radioactivity from the FDNA only formed a short-term glaciological reference, considering very low $^{137}$Cs values and the short half-life of $^{131}$I (8 d) in the four ice cores from the Alps. Wang et al. (2015) investigated four glaciers in the Tibetan Plateau (TP) for the total beta activity in May 2011 and observed significant horizons in the snow pits. We collected snowpack samples from four glaciers in the Tianshan and Qilian mountains to the north of the TP (figure 1). The array of sampling sites, distanced ~2000 km from west to east, were ideal for studying the transport and deposition of FDNA fallout. We expect the results to enlarge knowledge of the atmospheric transport and deposition of FDNA to glaciers in northwestern China, to verify the lastingness-preservation of the
new TBA horizon and to define a new reference for future ice-core dating.

2. Methods

2.1. Samples and measurements

From May 4 through 26 2011, we collected snow samples from the snow pits of the accumulation zones of four separate glaciers in northwestern China (figure 1). The geographic locations, sampling sites and other related information are shown in table 1. The sampled glaciers span from 80°–100° E, 38°–44° N and are located in the Tianshan and Qilian mountains. The snow pits were all located in the relatively flat area of the glaciers. There were automatic weather stations (AWS) (Campbell® CR1000 model) set up to monitor the surface temperature and other climatic elements on these glaciers. The temperature sensors (±0.1 °C) were mounted at 1.5 m above the snow surface. The 1.5 m air temperatures of the four sampling glaciers recorded by the AWS were below 0 °C between the FDNA starting and the fallout depositing, and they fluctuated at 0 °C during the sampling period (figure 2(a)), showing that there might be slight surface melting when sampling. The multi-yearly averaged daily surface temperatures on Glacier No. 1 show that surface melting could occur from June to September during 2011–2018 (figure 2(b)).

On 1st of August 2018, we revisited the Urumqi Glacier No. 1 and dug a much deeper snow pit (2.8 m, i.e. 1.6 m water equivalent) at the same site as in May 2011. The sampling procedure followed the same protocol as we used in 2011. Before sampling, the surface of the snow-pit wall was decontaminated by carefully removing the 5 cm-thick layers. Each snow
pit was sampled from top to bottom at a depth step of 10 cm to give a water volume of approximately 1.2 l per sample. All containers and tools used in the field and laboratory were acid-cleaned and sealed following the procedure for glacier samples described in Bilkiewicz (1978). Samples were kept frozen until analysis at the State Key Laboratory of Cryospheric Science (SKLCS).

The TBA was measured in the SKLCS in December 2011, using a Mini 20 Alpha-Beta Multidetector (Eurisys Mesures Company). The melted samples were passed through the anion- and cation-absorption membranes, which were dried before measuring. The duration of each sample’s measurement for the TBA was 86 400 s (1440 min or 24 h), reporting the result as average counts per minute (cpm). The background TBA in the environment was also measured for 24 h and was reported as 0.2 cpm, comparable to a standard of 0.4 cpm. A more detailed description of the TBA measurement for snow/ice samples can be referred to in Tian et al (2007). For comparison and expanding the geographic range of view, we also integrated the TBA data of Wang et al (2015) into our work. The amounts and size distributions of insoluble particles in the snow samples from Glacier No. 1 in 2018 were analyzed by an AccuSizer 780A (0.5–400 µm measurement range with an uncertainty less than 5%) (Ming et al 2016).

2.2. Dispersion simulation for the radioactive releases

We used the concentration module in the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (available at https://www.ready.noaa.gov/) (Draxler et al 1997, Stein et al 2015) to simulate the dispersion of the radioactive releases from the FDNA. The six-hourly Global Data Assimilation System (GDAS1) data of one-by-one square degree were used as the meteorological data to conduct the simulation. The fallout consisted of two dominant species, $^{131}$I and $^{137}$Cs, respectively. The recorded timeline of the FDNA shows that the initial radioactive release to air occurred around at 10:00 am (UTC + 9) on March 12 (BBC 2011). More than 99% of the total fallout was released to the air by March 31 (Company, 2012).

We assume that the accident was evenly emitting radioactive fallout to the air from March 12 to March 31. The total radioactive release to the air has been variously estimated by the Japan Atomic Energy Agency (Company 2012), Nuclear Safety Commission (NSC) of Japan (NSC 2011), INES (INES 2011), etc. We adopted the estimate and correction released by the Tokyo Electric Power Company (Company 2012), reporting that the accident released $\sim$ 500 PBq $^{131}$I and $\sim$ 10 PBq $^{137}$Cs into the air directly. Because
131I and 137Cs have different half-lives, we converted 137Cs concentration to 131I equivalent by multiplying it by a factor of 40 (Company 2012), which gives ~900 PBq 131I in total dispersed in the atmosphere from March 12 to March 31 from the source site (37.42° N, 141.03° E). We set the total modelling duration to 480 h (20 d) on a daily-average basis and ran the model.

3. Results and discussion

3.1. The spread of the FDNA fallout to central Asia

The hemispheric transport of the Fukushima radiation clouds by the westerlies displayed an exponential-like decrease eastward, with a dilution factor of at least five orders of magnitude following a full circuit around the globe (Hsu et al 2012). The radioactive particles resulting from the FDNA arrived in California on March 18 (EPA 2011) and in western Europe on March 19 (Masson et al 2011). However, the arrival of the radioactive particles from the exact accident in central Asia has been rarely reported previously. The simulated results by the HYSPLIT model show that they entered western China both at 5000 m (figure 3) and at 10,000 m (supplementary figure 1 (stacks.iop.org/ERL/15/084016/media) above sea level (a.s.l) in late March. The simulated daily 131I in the study area (70°–110° E and 30°–50° N) were flat before mid-March and thereafter ascended to higher concentration levels for both 5000 m and 10,000 m levels (figure 4). The arrival of the plume over the study area at 10 km height was earlier than that at 5 km height, which is consistent with a previous simulation by Povinec et al (2013) and due to rapid dispersion at 10 km height by jet cores (Meszaros et al 2016).

Figure 5 shows the values of air absorption dose in radiation environment automatic monitoring stations in Urumqi and Lanzhou from March 15 to May 31 2011, according to the report released by the Ministry of Environmental Protection, China (MEP 2011). The air absorbed dose rate (AADt) in Urumqi kept fluctuating around 105.7 nGy h⁻¹ before March 23rd and after that started to increase significantly and quickly reached 112.6 nGy h⁻¹ within a week (annotated by the orange dashed rectangle). The abrupt increase of the AADt in Urumqi implies that the first wave of the radioactive particles arriving in central Asia was detected on March 23, around two weeks after the occurrence of the FDNA. This is consistent with the previous simulation and an earlier study, which suggested that the journey of the radioactive particles around the globe took ~18 d (Hsu et al 2012).

In a more eastern inner city, Xi’an, the 131I measured in the air shows a significant peak around March 25 (Liu et al 2013), consistent with the prediction of the previous simulation (Hsu et al 2012). The air absorbed dose rate in Lanzhou did not show a significant increase in late March of 2011. An outstandingly high record showed around 25 May, which was much later than the global dispersion of the fallout and should be considered as an outlier; however, the aerosol 131I over the Lanzhou city measured by Wu

![Figure 4](image-url)
Figure 5. The air absorbed dose rate in nanoGray per hour (nGy h$^{-1}$) recorded by the local radiation-environment automatic monitoring stations in the Urumqi and Lanzhou cities during March 15 to May 31, 2011 and released by MEP (2011), and the concentration of $^{131}$I in megabequerel per cubic meter (mBq m$^{-3}$) in the air over Xi’an (adapted from figure 1 in Liu et al (2013)) and Lanzhou (adapted from figure 1 in Wu et al (2013)). The blue dashed lines show the results of the abrupt-change-point analysis for the AADR in Urumqi and Lanzhou, applying the method introduced by Hammer et al (2001) and Gallagher et al (2011).

Figure 6 shows a consistent variation with that over Xi’an during the same period. The consistency between modelling and ground measurements suggests that the fallout of the FDNA dispersed in western China and that the mountain glaciers may have acted as the receptors of the fallout.

The ground measurements in Xi’an, Lanzhou and Urumqi (figure 5) are more consistent in radioactivity shown for the modeling results at 5 km height than for that at 10 km (figure 4); this suggests that the radiative fallout received in the study area might primarily be transported through a long distance from the source in the mid- and low- troposphere rather than the upper troposphere and stratosphere.

3.2. The trace of the FDNA fallout in the high-Asian glaciers and new dating horizons

Figure 6 shows the TBA profiles in the snow pits of the four sampling glaciers in 2018 (a) and 2011 (b), (c), (d) and (e). The TBAs in the instantly-depositing layers are significantly higher than in the layers below them by ~2–4 times, and even higher ratios of the TBAs between the TBA-maximum layer and non-FDNA layers can be observed (table 2). The results show that all four sampling glaciers had received significant radioactive-fallout deposition from the FDNA.

We revisited the Urumqi Glacier No. 1 in August 2018 and dug a much deeper snowpit (2.8 m, i.e. 1.6 m weq) at the same site as in May 2011 (figure 6(a)). At the depth of 125 cm to 132 cm weq in the snowpit, we observed a significant horizon of the TBA concentration, which is four times larger than the average of the other layers. The dust events around Glacier No. 1 occur in the springtime and then intense melt in the following summer (figure 2(b)) would accumulate dust in a distinct layer in the snow, generally with only one dust layer forming every single year (Wang et al 2006). The peaks in the dust profile show that the TBA horizon around 130 cm of the snowpit can be traced back to 2011 (figures 6(a)–(b)). The TBA received from the FDNA was not lost during the last seven years, but instead was reserved in the layer accumulated from 31 cpm kg$^{-1}$ in 2011 to 42 cpm kg$^{-1}$ in 2018 (table 2). The accumulation of TBA in the dust layer is probably due to the densification and melt-refreezing of snow after deposition (Wang et al 2006) and the fact that nuclides are tightly bound to insoluble particulate matter in ice, which has been revealed in an Alps ice core by Di Stefano et al (2019).
Figure 6. The TBA profiles in the snow pits of Glacier No. 1 (Gl_1, the red line) in (a) 2018 and (b) 2011 (the blue line), and the in-situ pictures of the snow pits are shown in the Supplementary Figure 2, and of (c) Glacier No. 72 (Gl_72, the blue line), (d) Qiyi Glacier (Gl_Qiyi, the blue line) and (e) Shiyi Glacier (Gl_Shiyi, the blue line) in 2011. The annual dust layers in the Gl_1 snow pit in 2018 (a) are described as the measured dust concentrations (the grey dashed line) with year marked for the peaks. The layers instantly receiving the FDNA fallout are shaded in grey.

Table 2. The ratios of the total beta activity (TBA) in the FDNA and non-FDNA layers of the snow pits of the sampling glaciers.

| Glacier No. | Year | FDNA layers | Non-FDNA layers | Maximum TBA layer |
|-------------|------|-------------|-----------------|-------------------|--------------------|
|             |      | TBA (cpm kg⁻¹) | TBA (cpm kg⁻¹) | Depth (cm weq) | Depth (cm weq) | TBA (cpm kg⁻¹) |
| Gl_1_2011   | 2018 | 1.9–24.9 | 13.2–18.8 | 27.8 | 47.1 | 2 | 4 |
| Gl_1_2011   | 2011 | 1.8–18.1 | 11.6–18.1 | 21.5 | 34.9 | 3 | 4 |
| Gl_1_2011   | 2011 | 1.6–16.6 | 10.4–16.6 | 15.0 | 19.6 | 2 | 2 |
| Gl_1_2011   | 2011 | 1.5–19.8 | 14.6–19.8 | 18.1 | 30.8 | 2 | 3 |
| Gl_1_2018   | 2018 | 124.7–132.2 | 124.7–132.2 | 41.7 | 41.7 | 4 | 4 |

3.3. Comparison with the fallout trace in the Muztagata glacier received from the Chernobyl accident

The Chernobyl accident in 1986 is the only other nuclear accident to be ranked at Level 7 besides the FDNA by the IAEA. The total-radioactive-activity release from the Chernobyl accident was estimated as 5300 PBq (1 PBq = 10¹⁵ Bq) in a most cited source (UNSCEAR 2000), while the best estimate of the release from the FDNA was 520 PBq (340–800 PBq) (Steinhauser et al 2014), approximately one-tenth of the former. It is challenging to track the trace of ¹³¹I from the FDNA in the sampling glaciers given its short half-life, while it is promising to retrieve the robust signal of ¹³⁷Cs with a relatively long half-life (~30 years) in the layer depositing seven years ago. The variation of the radionuclides deposition in Alpine areas may be significant. For example, an investigation into the radioactivity in the Swiss Alps showed that totally-deposited activity ranged from ~500 to 14 000 Bq m⁻² (Haebler et al 1988). It is risky to directly evaluate the regional environmental
impacts from the Chernobyl accident and FDNA by comparing the TBA records in two glacial sites. However, the comparison could provide us with a clue as to the difference between the influence from the Chernobyl accident and FDNA, which are both over 4000 km away from the study area.

Tian et al (2007) identified and confirmed a significant horizon of TBA (∼ 7.5 cpm kg$^{-1}$) from the CNPP accident in an ice core from the Muztagata Glacier (figure 1). To compare the radioactivity from the FDNA and the Chernobyl accidents depositing on the study area, we selected the Muztagata Glacier as the reference. Because of radioactivity decay, the beta-activity concentration (i.e. TBA) is corrected to the depositing time for comparison.

One approach to calculating the original deposition of the radioactivity is to use the model by Ambach et al (1988). The decay can be approximated by a power function $A(t) = A_0 * t^{-0.226}$, where $A$ denotes activity and $t$ is time in days. The corrected TBA depositing in the surface of the Muztagata Glacier in 1986 is 54.1 cpm kg$^{-1}$, taking into account that there are 17 years between the depositing time and the TBA measurement. The limitation of the model is that the power of 0.226 is only valid for the time when the activities were measured (Ambach et al 1988).

Another approach for calculating the original TBA is simply to use the half-life of $^{137}$Cs and the decay constant, if we consider that the main component of the TBA is $^{137}$Cs. During 1986–2003 the decay should be about 30%, and the original TBA is supposed to be ∼10.7 cpm kg$^{-1}$, which is much lower than the result from the first approach.

The TBAs depositing in the four sampling glaciers in this work range from 20 cpm kg$^{-1}$ to 47 cpm kg$^{-1}$, and similar TBAs (figure 7) were measured in the other four glaciers of the TP by Wang et al (2015) (figure 1). The FDNA had comparable amounts of radioactivity depositing in the High-Asian glaciers to that of the Chernobyl accident.

4. Conclusions

On 11 March 2011, a Level-7 radioactive-release accident occurred in the Fukushima-Daiichi nuclear power plant. In about two weeks, the radioactive nuclides arrived in central Asian cities (Urumqi, Xi’an and Lanzhou), according to the reported measurements and our simulation with the HYSPLIT dispersion model. We collected snow-pit samples from four glaciers in the Tianshan Mountain and Qilian Mountain in May 2011 and analysed the total beta activity (TBA). Compared with the non-FDNA snow layers, the TBA in the FDNA layers was enriched by up to ∼2–4 times, and the maximum-TBA layer had even higher TBA ratios relative to the non-FDNA layers. The TBAs in the snow-pit profiles in 2011 show the radioactive horizons received from the fallout of the FDNA.

We revisited Glacier No. 1 and studied a deeper snowpit at the same site for dust and TBA in August 2018. We found the TBA accumulated in a dust layer, due probably to densification and
melt-refreezing processes and bonding to insoluble particles, and it increased from 30.8 cpm kg\(^{-1}\) in 2011 to 41.7 cpm kg\(^{-1}\) in 2018. The TBA concentrated in the dust layer of snow confirms that the TBA layer in the glacier from the FDNA is not short-term but lasting, and could be applied as a reference for future ice/firn-core dating.

We also compared the TBAs from the FDNA depositing in the glaciers in our study area with those from the Chernobyl accident. The comparison suggests that the depositions of the TBA in the glaciers in Central Asia and the Tibetan Plateau from the two accidents are comparable. As the radioactivity depositions from the FDNA were detected and verified for significance widely in the snow and ice of Greenland, the Tibetan Plateau and Qilian and Tianshan mountains, this new reference may help date snow and ice in the Northern Hemisphere.

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Data availability statements

It is currently shared by communities that the dataset would be publicly available upon acceptance of publication. Please directly contact the corresponding author (wangfeiteng@lzb.ac.cn) or the second author (petermingjing@hotmail.com) for the data repository and the authors will respond according to the statements.

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