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# High-resolution $^{129}$I bomb peak profile in an ice core from SE-Dome site, Greenland

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ABSTRACT

$^{129}$I in natural archives, such as ice cores, can be used as a proxy for human nuclear activities, age marker, and environmental tracer. Currently, there is only one published record of $^{129}$I in ice core (i.e., from Fiescherhorn Glacier, Swiss Alps) and its limited time resolution (1–2 years) prevents the full use of $^{129}$I for the mentioned applications. Here we show $^{129}$I concentrations in an ice core from SE-Dome, Greenland, covering years 1956–1976 at a time resolution of ~6 months, the most detailed record to date. Results revealed $^{129}$I bomb peaks in years 1959, 1962, and 1963, associated to tests performed by the former Soviet Union, one year prior, in its Novaya Zemlya test site. All $^{129}$I bomb peaks were observed in winter (1958.9, 1962.1, and 1963.0), while tritium bomb peaks, another prominent radionuclide associated with nuclear bomb testing, were observed in spring or summer (1959.3, and 1963.6; Iizuka et al., 2017). These results indicate that $^{129}$I bomb peaks can be used as annual and seasonal age markers for these years. Furthermore, we found that $^{129}$I recorded nuclear fuel reprocessing signals and that these can be potentially used to correct timing of estimated $^{129}$I releases during years 1964–1976. Comparisons with other published records of $^{129}$I in natural archives showed that $^{129}$I can be used as common age marker and tracer for different types of records. Most notably, the 1963 $^{129}$I bomb peak can be used as common age marker for ice and coral cores, providing the means to reconcile age models and associated trends from the polar and tropical regions, respectively.

## 1. Introduction

Iodine-129 is a long-lived radionuclide that primarily comes from human nuclear activities (HNA), such as nuclear bomb testing, nuclear fuel reprocessing, and nuclear accidents (Aldahan et al., 2007; Hou et al., 2013; Reithmeier et al., 2010, 2006; UNSCEAR, 2000). It is considered to be a good environmental tracer because its half-life is long (i.e., 15.7 Ma), its sources are known, its behavior in the environment is conservative, and its biogeochemical cycling is generally known (Fabryka-Martin et al., 1985; Muramatsu et al., 2004). $^{129}$I has been used in seawater (He et al., 2013), surface water (Schwehr et al., 2005; Snyder et al., 2010), groundwater (Herod et al., 2015), atmospheric particulates (Moran et al., 1999; Reithmeier et al., 2010), and soil (Honda et al., 2015) to understand associated natural processes in these systems and to trace movement and transport of HNA-derived radionuclides across the environment.

In addition, $^{129}$I has good applications to studies involving natural archives. Natural archives, such as coral and ice cores, are recorders of past environmental conditions and events. $^{129}$I in coral cores can be used to reconstruct, in good detail, past HNA impacts on areas where corals grew. This information can be further used to understand atmosphere and ocean transport of HNA-derived radionuclides and to establish coral age dates or chronologies (Bautista et al., 2016; Chang et al., 2016). These capabilities of $^{129}$I are applicable not only in coral cores, but likewise in other types of natural archives, such as ice cores.

Currently, the only published $^{129}$I record in ice core is that from the Fiescherhorn glacier, Swiss Alps (Reithmeier et al., 2006; Wagner et al., 1996). Similar to $^{129}$I in coral cores, $^{129}$I in Fiescherhorn features signals from different HNA, including bomb testing and nuclear fuel reprocessing. However, time resolution of the Fiescherhorn record is only

*Abbreviations:* HNA, Human Nuclear Activities; NFR, Nuclear Fuel Reprocessing

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about 1–2 years, which may be insufficient to reveal seasonal and, in some cases, annual events. This time resolution also limits the capabilities of $^{129}$I in Fiescherhorn as HNA proxy, environmental tracer, and age marker.

Currently, one of the most widely used age marker for ice cores is the 1963 tritium bomb peak. This is used to identify which part of the ice core corresponds to year 1963. This peak was produced by aboveground nuclear bomb tests, which occurred between years 1945–1980. In terms of bomb fission yield, 93% of these tests were performed between 1951 and 1962, peaking in year 1962. This event caused a maximum in environmental tritium concentration in year 1963 (Reithmeier et al., 2006; UNSCEAR, 2000). However, the 1963 tritium bomb peak can be used as age marker only for a single year. Additional age markers before or after 1963 are needed to provide additional age anchor points and to enhance certainty of ice core chronologies.

In this study, we report $^{129}$I concentrations measured in an ice core from SE-Dome site, Greenland. The SE-Dome ice core has high accumulation rate (i.e., average 1.02 m/yr), allowing measurement of $^{129}$I at a time resolution of about 0.5 year or 6 months. The resulting time series features the most detailed historical reconstruction of $^{129}$I deposition to date, revealing annual and some seasonal variations. The current dataset spans years 1956–1976, covering years of aboveground nuclear bomb testing. Our objectives here are (1) to explore $^{129}$I in SE-Dome as HNA proxy, particularly looking at consistencies and discrepancies between measured $^{129}$I in SE-Dome versus historical estimations of $^{129}$I releases from different HNA sources; (2) to explore $^{129}$I in SE-Dome as age markers, comparing it with tritium concentrations previously measured in the same ice core (Iizuka et al., 2017) and possibly providing additional age markers for years before or after 1963; and (3) to investigate annual and seasonal variations in $^{129}$I concentration, providing a detailed historical record of $^{129}$I deposition, which will be useful for future environmental tracer studies.

## 2. Material and methods

### 2.1. Study site, sampling, sample description and age model

The ice core analyzed in this study was drilled at the SE-Dome site (67.18°N, 36.37°W; 3170 masl; Fig. 1), 185 km north of Tasiilaq, Greenland. Climate in SE-Dome, based from observations in Tasiilaq, is characterized by unusually high amounts of precipitation, peaking in January at about 120 mm and lowest in July at about 47 mm (Cappelen et al., 2001). This results to an accumulation rate of about 1.02 m/yr of water equivalent for the ice core. Sampling was done from May 22 to 27, 2015 using a lightweight electromechanical ice-core drilling system developed in Hokkaido University (Matoba et al., 2014). Complete details of the sampling is reported by Iizuka et al. (2016). The total length of the ice core is 90.82 m. However, for this study, only sections from 90.72 up to 66.93 m were analyzed, which corresponds to years 1956.1–1976, respectively. Age model of the SE-Dome ice core was constructed up to sub-annual resolution (i.e., with estimated accuracy in the order of months) by matching $\delta^{18}$O variations in the ice core to simulations from isotope-enabled climate models (Furukawa et al., 2017).

### 2.2. Sample treatment and $^{129}$I measurement

Ice core samples were stored and processed in −50 °C and −20 °C facilities, respectively, in the Institute of Low Temperature Science, Hokkaido University in Hokkaido, Japan and in the National Institute of Polar Research, Tachikawa, Tokyo, Japan. Use of these low-temperature facilities was necessary to prevent any melting and to preserve the integrity of ice core samples. Subsampling at 0.5 m intervals (i.e., roughly corresponding to a time resolution of 0.5 year) were done using electric saws, with subsequent surface cleaning step using ceramic knives. Samples were then put inside clean (with 1% $HNO_3$), capped, HDPE containers, and then weighed and stored until $^{129}$I pretreatment.

About 200 mL of each sample was used for $^{129}$I measurement by accelerator mass spectrometry (AMS). AMS target preparation involved solvent extraction and back-extraction using $CCl_4$ and $NaNO_2$ for iodine purification (detailed schematic of solvent extraction procedure is shown in Fig. S1 in Supporting Information). 0.75 mg of low-ratio Woodward iodine carrier ($^{129}$I/$^{127}$I ratio = $1.5 \times 10^{-14}$) was added to form enough AgI precipitate. AgI precipitation from the solution was done through the addition of $AgNO_3$ in acidic conditions. AgI was then pressed with Nb in aluminum cathodes and analyzed by AMS (MALT facility, the University of Tokyo; details of AMS measurement is reported by Matsuzaki et al., 2015). Note that this analytical method only measures inorganic forms of iodine and is thus slightly underestimated (i.e., Hou et al., 2009 estimate > 90% of iodine in precipitation is inorganic).

Results are reported as $^{129}$I atoms/L of $H_2O$. This is calculated from the results of AMS measurement ($^{129}$I/$^{127}$I of the sample + carrier) multiplied only with the known amount of iodine carrier added (i.e., 0.75 mg), then divided with the volume of the sample. We note that this is a valid estimation because we determined that $^{127}$I concentration in the SE-Dome samples were negligible compared to the amount of iodine carrier added (i.e., $^{127}$I concentrations in SE-Dome are below our ICP-MS instrument detection limit of 0.089 ppb. This means that a 200-mL sample will have less than $1.78 \times 10^{-5}$ mg of $^{127}$I, which is negligible compared to the 0.75 mg of iodine carrier added to the samples).

### 2.3. Simulation model for $^{129}$I from nuclear bomb tests

For years 1958–1962, bomb-derived $^{129}$I, as observed in SE-Dome, was simulated using the equation:

$$I_y = B + S_{y-1}w(1-x) + US_{y-1}y(1-z) + S_{y-2}wx + US_{y-2}yz \quad (1)$$

where, $I_y$ is the measured $^{129}$I (in $10^6$ atoms/L of $H_2O$) in year, $y$; $B$ is the baseline value, which we designate as 3 (based on the lowest point in the actual $^{129}$I record in SE-Dome). $S$ and $US$ are fission yields (in Mt; data from UNSCEAR, 2000) of bomb tests performed by the Former Soviet Union and the United States, respectively; $w$ and $y$ are conversion factors from Mt yield to $10^6$ atoms of $^{129}$I/L of $H_2O$ for Soviet and US tests, respectively; and $x$ and $z$ are fractions of the signal that goes to secondary peak for Soviet and US tests, respectively. For years 1963–1965, instead of using Eq. (1), Soviet tests in year 1962 were instead split into three peaks, one year after the other (i.e., years 1963–1965), with 45%, 33%, and 22% of the contributions, respectively. The simulation calculations are discussed in detail in Section 3.2.

## 3. Results and discussion

Fig. 2 shows $^{129}$I time series in ice core from SE-Dome site, Greenland (hereafter referred to as “SE-Dome”) and tritium (previously reported by Iizuka et al., 2017), spanning years 1957.3–1976.0 and 1956.1–1976.0, respectively. Note that $^{129}$I and tritium measurements were performed at similar resolutions (i.e., 0.5 m or about 0.5 year) and using the exact same ice core segments to make comparisons direct and simple.

$^{129}$I exhibits concentration maxima or “peaks” during the following years (peak magnitudes are given inside parentheses in $10^6$ atoms of $^{129}$I per L of $H_2O$): 1958.9 (15.84), 1962.1 (21.78), 1963.0 (35.99), 1964.2 (39.79), 1967.6 (11.09), 1972.2 (21.08), and 1974.9 (21.59). Meanwhile, tritium exhibits three peaks in years 1959.3 (41.8 tritium units or TU), 1963.6 (254.4 TU), and 1964.6 (147.3 TU). We note that $^{129}$I and tritium are expressed as activities rather than flux because our objective is to associate observed signals with source emissions from human nuclear activities (HNA). It is difficult and misleading to make this association when looking at fluxes because flux is largely influenced by ice core accumulation rate and this skews the resulting time series trends (see Fig. S2 in Supporting Information for $^{129}$I fluxes and