Temporal markers in a temperate ice core: insights from ^{3}H and ^{137}Cs profiles in the Adamello glacier

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Abstract. The article discusses the use of ^{3}H and ^{137}Cs as temporal markers in ice cores extracted from temperate glaciers. We present a complete tritium (^{3}H) profile for a 46 m ice core drilled from Adamello glacier, a temperate glacier in the Italian Alps, and compare it to ^{137}Cs profile from the same ice core. Our analysis reveals contamination of tritium between 19 and 32 m of depth, which can be attributed to the worldwide radioactive contamination caused by atmospheric nuclear bomb testing in the 1950s and 1960s. Results show that the radioactive peak associated with 1963 is not coincident for ^{3}H and ^{137}Cs, but the ^{3}H peak occurs 1.5 m above the ^{137}Cs one. Our hypothesis is that this misalignment is caused by meltwater-induced postdepositional processes that affect ^{137}Cs, which is more sensitive to percolation than ^{3}H. The total inventory of ^{137}Cs in this ice core is also among the lowest ever reported, providing additional evidence that meltwater disturbed its distribution into the ice. On the contrary, the total tritium inventory is comparable to what is reported in literature, making it a more reliable temporal marker for temperate glaciers.

1 Introduction

Due to its sensitivity to temperature variations, the cryosphere provides some of the most evident indications of climate change, with dramatic ice losses reported globally. In particular, mountain glaciers are globally losing mass at accelerating rates (Zemp et al., 2019) and according to future projections, including the most optimistic ones, this trend will continue throughout this century (Hock et al., 2019). European alpine glaciers are amongst the fastest declining ones, with an estimated average ice loss of -0.74 $\pm$ 0.20 m w.e. yr^{-1} for the period 2000-2016 (Davaze et al., 2020). As a result of increasing temperatures, glaciers are not only retreating, they are also changing with respect to their thermal regime. Atmospheric warming and more frequent melting events are rapidly modifying the temperature distribution of glaciers, increasing ice temperature (Gilbert et al., 2010, 2020). Finding accumulation basins with temperatures constantly below the pressure melting point is becoming rarer and rarer in many mountain ranges as they are turning from cold to temperate conditions (Gabrielli et al., 2010). Temperate ice is defined as ice that because of its thermodynamic state can contain meltwater inclusions (Lliboutry, 1971). Moreover, the rise of the Equilibrium Line Altitude (ELA), defined here as the altitude at which a glacier has a zero mass balance, is leading to a gradual reduction in glacier size, ultimately eradicating the accumulation basins. For the Alps, it is expected that by the

end of the 21st century at least 69% of the glaciers in the Alps will completely lie below the ELA (Zebre et al., 2021). As a consequence of increased temperature and subsequent percolation of meltwater, it is becoming increasingly difficult to retrieve reliable climatic records from mountain glaciers through ice core drilling. In the future, the role of glaciers as paleoclimatic and environmental archives will thus depend on our ability to read signals from temperate glaciers. Determining if and to what degree such glaciers can provide climatic and environmental records is an urgent issue that ice core science must address. Meltwater percolation is in fact a potential threat to the preservation of proxy signals in the ice, smoothing the temperature isotopic signal (Thompson et al., 1993), relocating impurities (Pavlova et al., 2015) and altering trace elements records. Since meltwater-induced postdepositional effects are not consistent for all chemical species but are influenced by several factors (Avak et al., 2018; Moser et al., 2023) including solubility, the position inside the ice lattice, and the concentration, it can be sometimes difficult to evaluate to what degree the signal has been altered. In general, elements and compounds which are not soluble in water, are present in the form of particles or are well incorporated in the ice lattice will be less prone to elution and relocation due to meltwater (Eichler et al., 2001; Wong et al., 2013), but microparticles may still accumulate at melt surfaces leading to the enrichment of elements bound to particulate matter (Pavlova et al., 2015; Niu et al., 2017).

Among the many contaminants we can find in ice cores there are radionuclides of anthropogenic origin. During the 1950s and 1960s, atmospheric nuclear bomb testing carried out mainly by USA and USSR led to worldwide radioactive contamination of the environment. Atmospheric thermonuclear explosions can reach the stratosphere, where the circulation allows the contamination to rapidly extend to the whole hemisphere. The majority of tests was carried out in the northern hemisphere, which therefore suffers from the highest contamination, although radionuclides generated from atmospheric nuclear tests have been found in the Antarctic continent as well (Picciotto and Wilgain, 1963; Jouzel et al., 1979). In 1963 the atmospheric yield of nuclear bomb testing reached its maximum (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000), before the entering into force of the Partial Test Ban Treaty, which introduced a ban on atmospheric nuclear testing for USA, USSR, and UK. This maximum has been registered in paleoclimatic archives and has since been used as a temporal reference horizon (Mikhalenko et al., 2015; Eichler et al., 2000; Clemenza et al., 2012). Likewise, radioactive layers relative to the 1986 Chernobyl accident (United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; International Atomic Energy Agency, 2006) and the 2011 Fukushima accident (Steinhauser et al., 2014) have been found in environmental records. Among the main radionuclides which were injected into the environment following such events are ^{137}Cs and ^{3}H, and the presence of both these radionuclides have been widely documented in glaciers and ice caps. The radioactive layers corresponding to 1963 and 1986 can aid the dating of ice cores, if the signal is well preserved, and can shed some light on melting processes inside the glacier (Kang et al., 2015).

Here we present the complete tritium (^{3}H) profile for a 46 m ice core extracted from the Adamello glacier, a low altitude temperate glacier. To assess the integrity of signals in temperate ice, the tritium record is also compared with ^{137}Cs and hyperspectral data related to dust content. Finally, a comparison is made with similar data from a cold ice core (Colle del Lys).

1.1 Tritium

Tritium is a pure beta emitter with a maximum emission energy of 18 keV and a half life of 12.33 years (Lucas and Unterweger, 2000). It is naturally produced in the upper atmosphere as a result of the interaction between cosmic rays and atmospheric gases, mainly nitrogen, with a production rate of 0.320 atoms $cm^{-2} s^{-1}$ (Masarik and Beer, 2009), resulting in a low natural background detectable in precipitations; this was estimated to be <1 $Bq L^{-1}$ in the United States (Thatcher, 1962; Kaufman and Libby, 1954), although measurements before the 1950s, when the artificial production of this radionuclide began, are quite scarce. Reconstructions of the natural tritium background are available from ice core data from Fiescherhorn glacier (Schotterer et al., 1998a).

^{3}H was a key component in thermonuclear bombs, being one of the hydrogen isotopes used as thermonuclear fuel. During the bomb testing period, tritium concentrations in precipitations in the Northern Hemisphere reached several thousands of TU. Figure 1 shows plots of monthly concentration of tritium in precipitation in Vienna and Ottawa, obtained from the Global Network of Isotopes in Precipitation (GNIP) Database (International Atomic Energy Agency and World Meteorological Organization, 2021). The trends shown here can be taken as representative of the Northern Hemisphere: a gradual increase during the 1950s leading up to a maximum in 1963 and a subsequent gradual decrease following the entering into force of the Partial Test Ban Treaty. The tritium activity concentration shows a strong seasonality, with a maximum in spring-summer each year. This mechanism has been referred to as 'Spring Leak' (Michel, 2005; Harms et al., 2016) and is due to enhanced air exchange between the troposphere and the stratosphere, i.e. the main reservoir for this radionuclide, during spring. ^{3}H has been extensively used as a temporal horizon marker for 1963 in ice cores (Eichler et al., 2000; Qiao et al., 2021; Mikhalenko et al., 2015; Gabrieli et al., 2011; Kang et al., 2015) and, as long as activities were high, to reveal seasonal signals and aid annual layer counting in ice cores (Schwikowski et al., 1999; Schotterer et al., 1998b; Yasunari et al., 2007). Since tritium has a relatively short half-life, as more time passes since its emission into the environment, its detection is gradually becoming more challenging.

1.2 Adamello Ice core

The ADA16 ice core was drilled during the spring of 2016 at Pian di Neve (WGS84: 10°31'22" E, 46°8'51" N), the former accumulation basin for the Adamello Glacier; this glacier has experienced several years of negative mass balance (Ranzi et al., 2010) and is now entirely located below the ELA. The first chronology for this ice core has estimated a surface age of 1993, indicating no accumulation was preserved on this glacier for the last 20 years (Festi et al., 2021). Due to its relatively low altitude (3100 m asl) and prevailing climatic conditions, Adamello glacier has a temperate regime: the body of the glacier is at melting point and during summer is massively subject to melting, which could alter the climatic signal preserved in the ice. The ice core was drilled where the maximum ice thickness of the glacier was estimated (268 ± 5 m) (Picotti et al., 2017) and reached a total depth of 46 m. During drilling, ice core chips were also collected. These are residual materials produced during the mechanical drilling by the contact between the ice and the rotating blade and are usually discarded due to potential chemical contamination, as the ice has been in direct contact with the drill. Nonetheless, they are suitable for radionuclide analysis, as
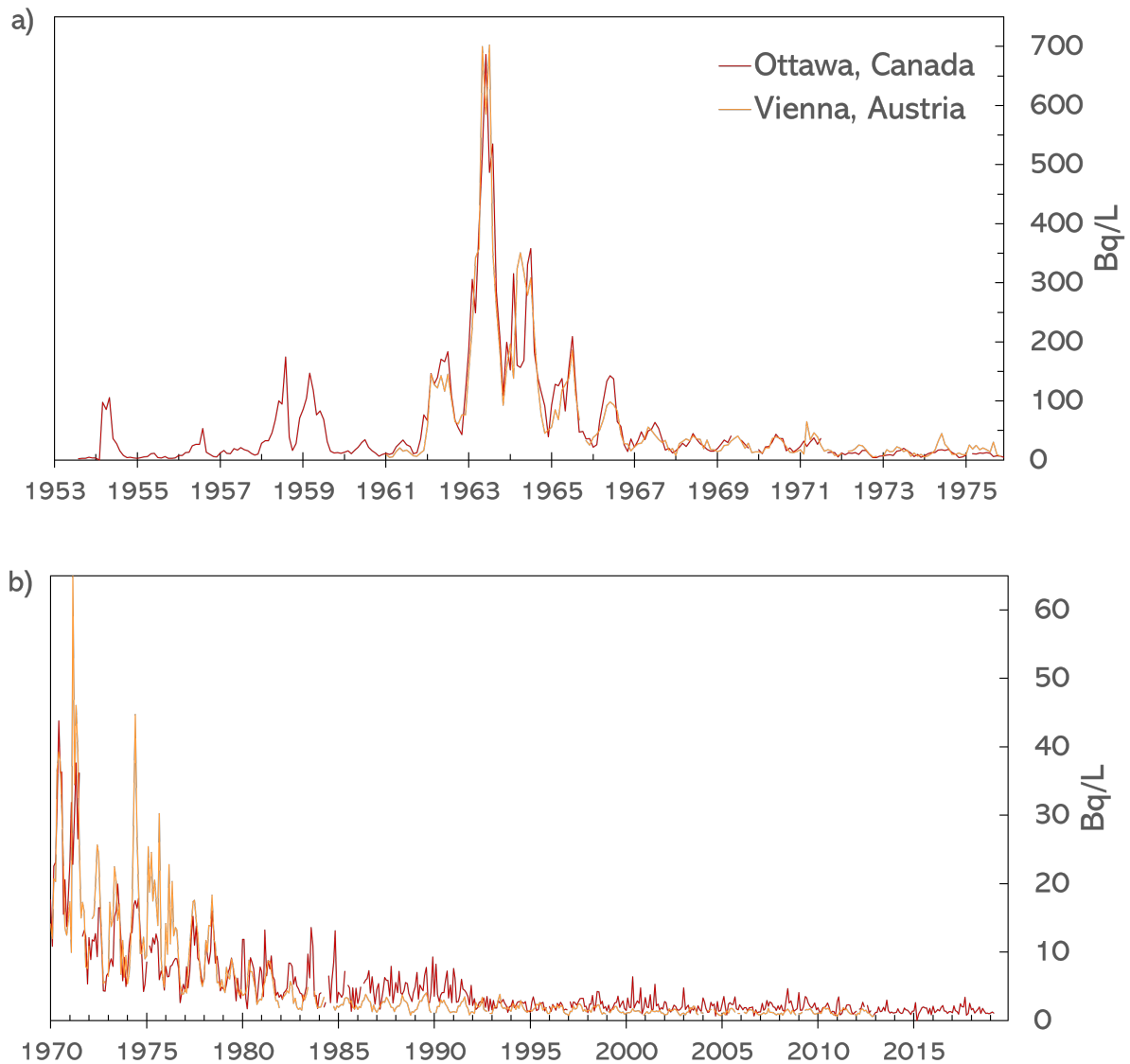
Figure 1. Monthly concentration of tritium in precipitation in Vienna (Austria) and Ottawa (Canada) from 1953 to 1975 (a) and from 1971 to 2019 (b). Data from the Global Network of Isotopes in Precipitation (GNIP) Database (International Atomic Energy Agency and World Meteorological Organization, 2021), converted to $Bq\,L^{-1}$ (where 1 Tritium Unit = 0.118 $Bq\,L^{-1}$). Please note the different scale of Y-Axes in the two panels.

radioactive contamination during drilling operations is virtually impossible, although they have a poor depth resolution which
is limited to the entire length of the single ice core sections extracted during drilling runs.

Analyses were performed both on the chips and on ice samples from the ice core. The chips were divided into 112 runs, some of which were stored together, for a total of 71 samples covering each a mean thickness of 59.4 cm. Ice samples were taken from 27 m to 35 m of depth to obtain a higher temporal resolution for this part of the ice core, with samples taken every 5 to 10 cm.

1.3 Lys Ice core

To have a reference from a cold alpine glacier, we also analyzed 38 samples belonging to an ice core drilled at Colle del Lys (7°51'4" E, 45°55'13" N), which is the uppermost part of the Lys glacier, located in the Monte Rosa massif at 4240 m asl. This is one of the few glacierized areas presenting cold ice in the Alps: thanks to their elevation, glaciers in the upper part of the Monte Rosa massif are only occasionally subject to melting (Hoelzle et al., 2011). Thus, several undisturbed ice cores were obtained from this area, at Colle del Lys and Colle Gnifetti (Smiraglia et al., 2000; Villa et al., 2006; Wagenbach et al., 2012). For the aims of the present work, we analyzed samples remaining from the ^{137}Cs analysis that was carried out by Clemenza et al. (2012).

2 Materials and Methods

2.1 Sample Preparation and Analysis

Samples from ADA16 were thawed in a clean room inside precleaned polyethylene bottles and then filtered using a vacuum-driven filtration system and polycarbonate filters (47 mm diameter, 0.45 μm cutoff), then passed through Eichrom's Tritium Column, to eliminate all possible radioisotopic disturbances present in the sample. Tritium columns were first conditioned using 10 ml of ultrapure water, before adding 25 ml of sample, of which the first 5 ml were discarded while the rest was collected for further preparation. At last, samples were prepared for liquid scintillation counting by mixing 8 ml of sample with 12 ml of scintillation cocktail (UltimaGold ullT) in a 20 ml polypropylene low diffusion vial.

Samples from Lys glacier were directly prepared for liquid scintillation and measured using a total beta protocol. Thus, data belonging to this ice core will be reported as counts per minute (cpm) as it is not possible to perform a calibration curve for total beta analysis.

The liquid scintillation counter used is the Quantulus 1220 produced by Perkin Elmer. Quantulus is equipped with two dual multi-channel analyzers, enabling simultaneous measurement of 4 spectra, each with 1024 channel resolution. In our analysis we considered a counting window between 50 and 250, which covers the emission energy of tritium.

2.2 Calibration curves and Detection Limit

For conversion of cps to Bq L[-1] we constructed a calibration curve for each dataset (low resolution chips and high resolution ice core) by measuring tritium standards of known activity, as can be seen in Figure 2. Standards were prepared from NIST Hydrogen-3 Radioactivity Standard 4361C. 3 sets of standards (Set1-2-3) were prepared following the same procedure which
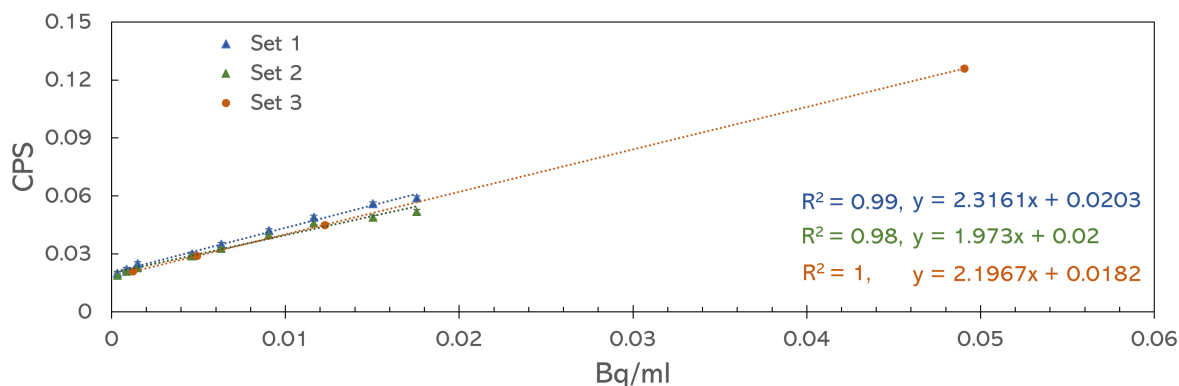
Figure 2. Calibration curves calculated for the 3 sets of tritium standards. Errors on cps are 0.001.

was used for the samples, the only exception being that Set2 was the only one that passed through tritium columns. Set1 and Set2 were prepared with identical activities to estimate the potential influence of the elution columns on sample activities. As the difference between Set1 and 2 was below 15% (in terms of CPS), we decided not to elute the tritium standards. Set3 was repeatedly measured in between sample runs to check for instrumental stability. Set1 was used for calibration of the low
125 resolution dataset, while Set3 was employed for the high resolution dataset.

Following Currie (1968) we calculated a Critical Level as $LC = 1.64\sqrt{2Rb/t}$ where Rb is the background rate and t is the measurement time; when the net sample rate is higher than the critical level, we can consider the sample contribution (Rs-Rb) to be distinct from the background, and the Specific Activity (Bq L^{-1}) was calculated using the calibration coefficient from the standard measurements. All activities were decay corrected to 01 January 2016. When (Rs-Rb) <LC, an upper limit
130 was calculated as three times the counting error on the sample measurement. The Detection Limit for our measurement is 1.5 Bq L^{-1}, calculated as 3.3 σ where σ is the standard deviation of 10 independent blanks (Currie, 1968).

2.3 Hyperspectral imaging spectroscopy

Hyperspectral imaging spectroscopy is a technique used to characterize surfaces and materials on the basis of their optical properties. In the visible part of the electromagnetic spectrum (from 300 to 750 nm), snow and ice show high reflectance;
135 ice core characteristics such as the impurity content (e.g., mineral dust, volcanic ash, black carbon, algae) affect the ice's capability to absorb and reflect electromagnetic radiation in the visible range, allowing the extraction of a continuous record of reflectance and derived parameters along the ice core. Ice core sections were placed on a fixed ice-core support, illuminated using a halogen stable light source (600 or 1000 W, LOT Quantum Design), and scanned with a HeadWall spectrometer (400 - 1000 nm, Hyperspec VNIR, HeadWall Photonics). The Hyperspectral imaging scanner was set in motion step by step
140 from the top to the bottom of sections, using a motion speed of 0.9 mm s^{-1} and an exposure time per frame of 39.8 ms. The

obtained high resolution images were thus processed to obtain reflectance curves considering a portion of 20 pixels centered on a central axis along ice-core sections. The technique, developed in collaboration between the European Cold Laboratory (EuroCold) and the Remote Sensing of Environmental Dynamics Laboratory (LTDA) both at University of Milano-Bicocca, is described in detail in Garzonio et al. (2018). For this study, we analyzed a portion of the ice core about 7 m long (from 26.8 m to 33.9 m), corresponding to the area around the main ^{137}Cs peak. Our aim was to verify if the presence of impurities could alter the radionuclides' distribution in the ice, as past studies have shown a strong bond between cesium and particulate matter (Di Stefano et al., 2019). Finally, we used the Snow Darkening Index (SDI), calculated as the normalized ratio between the reflectance in the red (i.e., from 640 to 670 nm) and green (i.e., from 550 to 590 nm) wavelengths, which gives information regarding the concentration of mineral dust in the ice core (Di Mauro et al., 2015).

3   Results and Discussion

3.1 ^{3}H Activity

The tritium profile for the Adamello ice core is reported in Figure 3. The low resolution dataset covered the whole depth of the ice core, but only the portion between 19 and 32 m displayed a tritium signal, implying these layers can be attributed to the period between the 1950s and the 1970s.

The tritium signal was further investigated by analyzing the high resolution dataset between 27 and 35 m. In general, good accordance between the two datasets was found, both concerning the measured specific activities and the general trend, with the exception of the main peak which was found at 29 ± 0.6 m in the low resolution dataset and at 30.530 ± 0.025 m for the high resolution dataset. We consider the low resolution dataset to be less reliable due to its broader depth coverage which may lead to inaccurancies when the signal exhibits peaks and valleys compressed in a narrow depth range, as shown by the high resolution dataset. Activity values are generally lower than those reported for other alpine glaciers (Eichler et al., 2000; Schotterer et al., 1998b, 1977; Oerter and Rauert, 1982) even if we account for decay. Discrepancies between tritium activities as reported in precipitation at the time of the radioactive fallout and tritium detected in ice cores can be explained by i) immediate loss of a fraction of the initial deposited signal due to summer snow melt, as proposed by Oerter and Rauert (1982); ii) present loss due to melting and percolation of water inside the ice, which is expected to be more extensive in the Adamello ice core than in ice cores extracted from cold glaciers.

3.2 ^{137}Cs and ^{3}H profile comparison

It is interesting to compare the tritium signal with the ^{137}Cs profile from the same ice core, as reported in Di Stefano et al. (2019). ^{137}Cs is also a byproduct of nuclear atmospheric tests, and since the transport mechanisms of these two radionuclides are almost the same (Pourchet and Pinglot, 1979), it is expected to find the 1963 peak at the same depth for ^{137}Cs and ^{3}H, as observed in ice cores from cold glaciers (Pinglot et al., 2003).
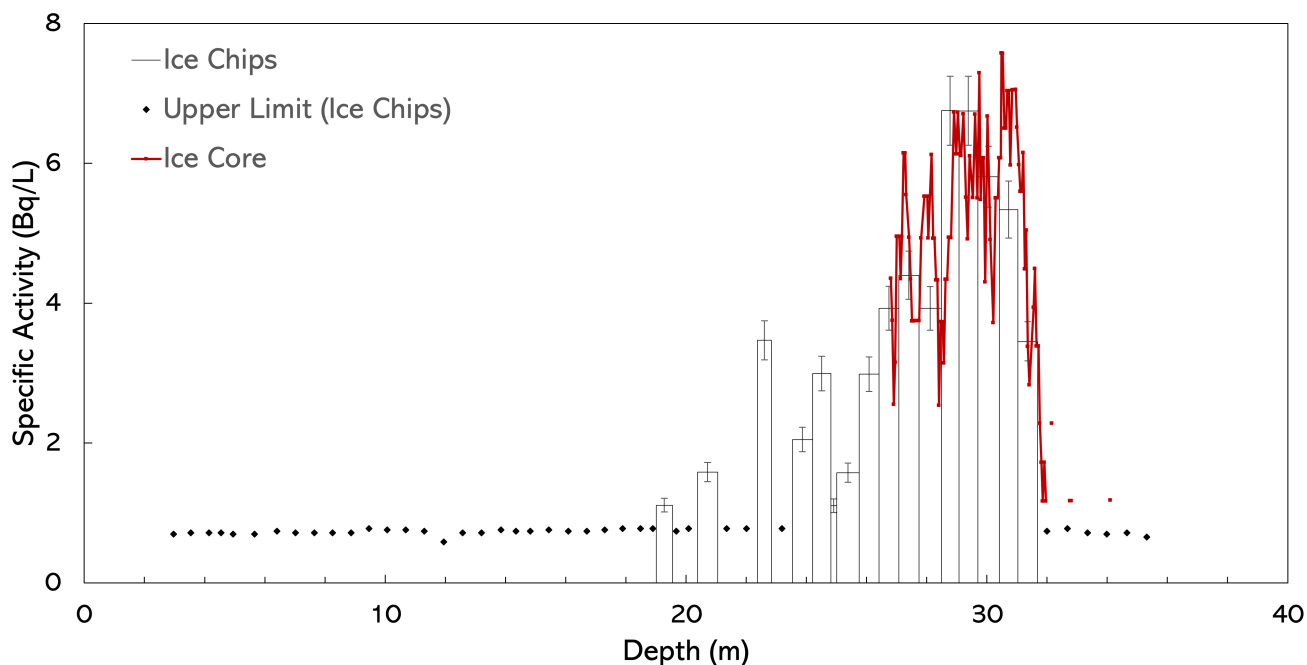
Figure 3. Tritium activity in the Adamello temperate ice core. Data reported in black are related to low resolution measurements (ice chips), diamonds indicate upper limits, bars observed activities. In red results from high-resolution analyses conducted on ice core sections.

In the Adamello ice core, the respective peaks are misaligned by at least 1.5 m if we consider the high resolution dataset for tritium, as can be seen in Figure 4 (b,d). Following the dating proposed by Festi et al. (2021), this would correspond to a time lag of approximately 2 years between the two peaks, which in terms of deposition would be difficult to explain. Our hypothesis is thus that the ^{137}Cs signal has been altered by post-depositional processes. We must note that Festi et al. (2021) used data from Di Stefano et al. (2019) as a tie point for 1963, therefore the established timescale may be slightly inaccurate regarding the layer counting between the 1986 and 1963 tie points, as it heavily relies on the implication that the^{137}Cs peak corresponded to the 1963 unaltered signal. The inaccuracy would introduce an additional error of 1 /2 years.

As shown in Figure 4c, SDI data display a well-defined impurity peak at 33 m of depth indicating a layer rich in mineral dust. This layer, located in section 77-78, is approximately 1 m deeper than the main ^{137}Cs peak (section 74-75). The presence of a major impurity layer below the ^{137}Cs peak suggests the latter is not the product of the enrichment of dust into a single layer as a result of melting. Impurities may in fact accumulate into a single layer if a hidden melting surface is present inside the glacier and this may also lead to radionuclide concentration (Baccolo et al., 2020). Since the impurity and the radioactive layer don't overlap, we ruled out this possibility. Dislocation of cesium particles inside the ice cores has undoubtedly happened, and the fact that a dust-rich layer with no cesium was found just below the cesium peak may be an indication that even a small amount of particulate (small enough to not be identified as a dust layer in the stratigraphy) can lead to a high cesium peak.
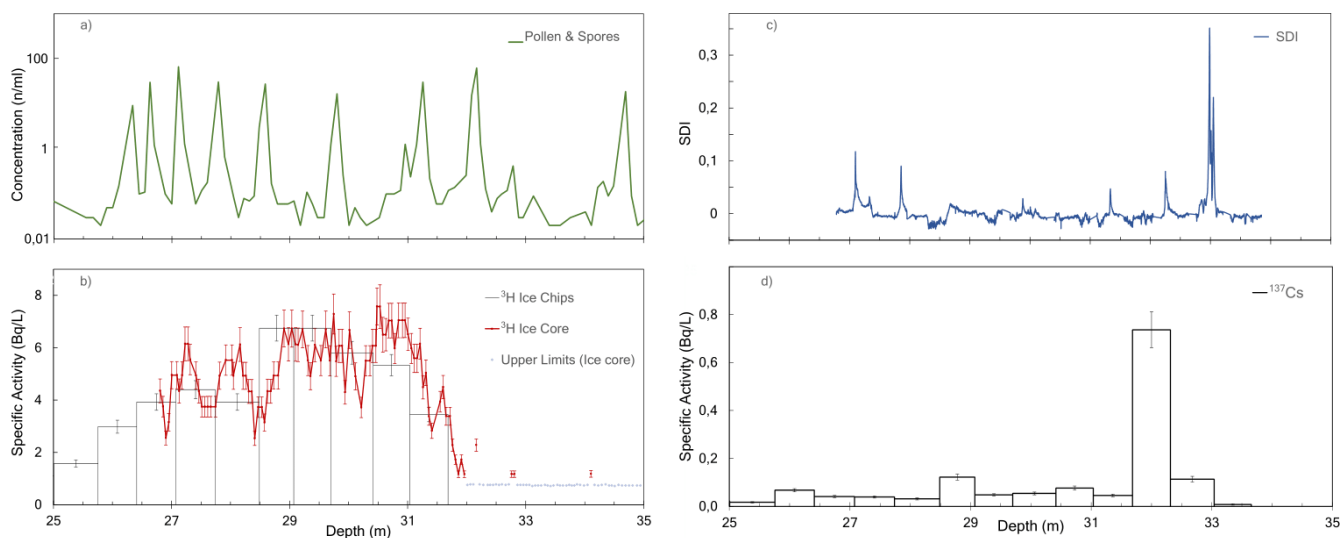
Figure 4. Detailed overview of the ice core in the portion where the the tritium and ^{137}Cs 1963 peaks have been observed (between 25 and 35 m deep). a) Pollen and spores concentration taken from Festi et al. (2021). Each peak of pollen and spores concentration reflects one flowering year (February–September). b) Tritium profiles.Specific activity in Bq L^{-1} of ^{3}H for the ice chip low resolution dataset (in black) and the ice core high resolution (in red). Diamonds indicate upper limits calculated for the high resolution data set. c) SDI record, a hyperspectral index related to the impurity content of ice. d) Specific activity in Bq L^{-1} of ^{137}Cs taken from Di Stefano et al. (2019)

The explanation we propose to explain the lag between the ^{3}H and the ^{137}Cs signals is the presence of meltwater-induced postdepositional processes altering the signal embedded in the ice. ^{137}Cs, mainly bound to particulate matter (Qin et al., 2012; Tanaka et al., 2013; Di Stefano et al., 2019), is more sensitive to percolation than ^{3}H (Pinglot et al., 2003), which is incorporated in the water molecule and can be regarded as a matrix signal. This result was unexpected since particle-bound signals have

190 generally been conserved inside this ice core and we thus expected cesium to behave accordingly. Particulate matter impurities are infact usually among the last proxies to be affected by meltwater relocation processes (Meyer and Wania, 2011; Moser et al., 2023). The exact mechanism behind the relocation of cesium is beyond the scope of this study and further research will need to investigate the relationship between cesium and meltwater presence in ice cores.

Vertical relocation of cesium has been reported in literature: Pinglot et al. (2003) found a similar delay between cesium and

195 tritium in ice cores extracted from low altitude glaciers in Svalbard, although the difference between the ^{3}H peak and the ^{137}Cs was less marked (<1 m). This can be expected since Adamello glacier has experienced much more melting than Svalbard and more time has passed since the deposition of radioactive species on the glacier. Similarly, Schotterer et al. (1977) reported a high concentration of ^{137}Cs 3 meters below the tritium bomb level in an ice core extracted from a relatively low altitude glacier in the Swiss Alps (Plaine Morte).

200 Our hypothesis is further supported by findings we obtained on total beta measurements from an ice core extracted at Colle del Lys in 1996(Smiraglia et al., 2000). Figure 5 shows that in this ice core the main beta (^{3}H) peak and the main ^{137}Cs coincide.
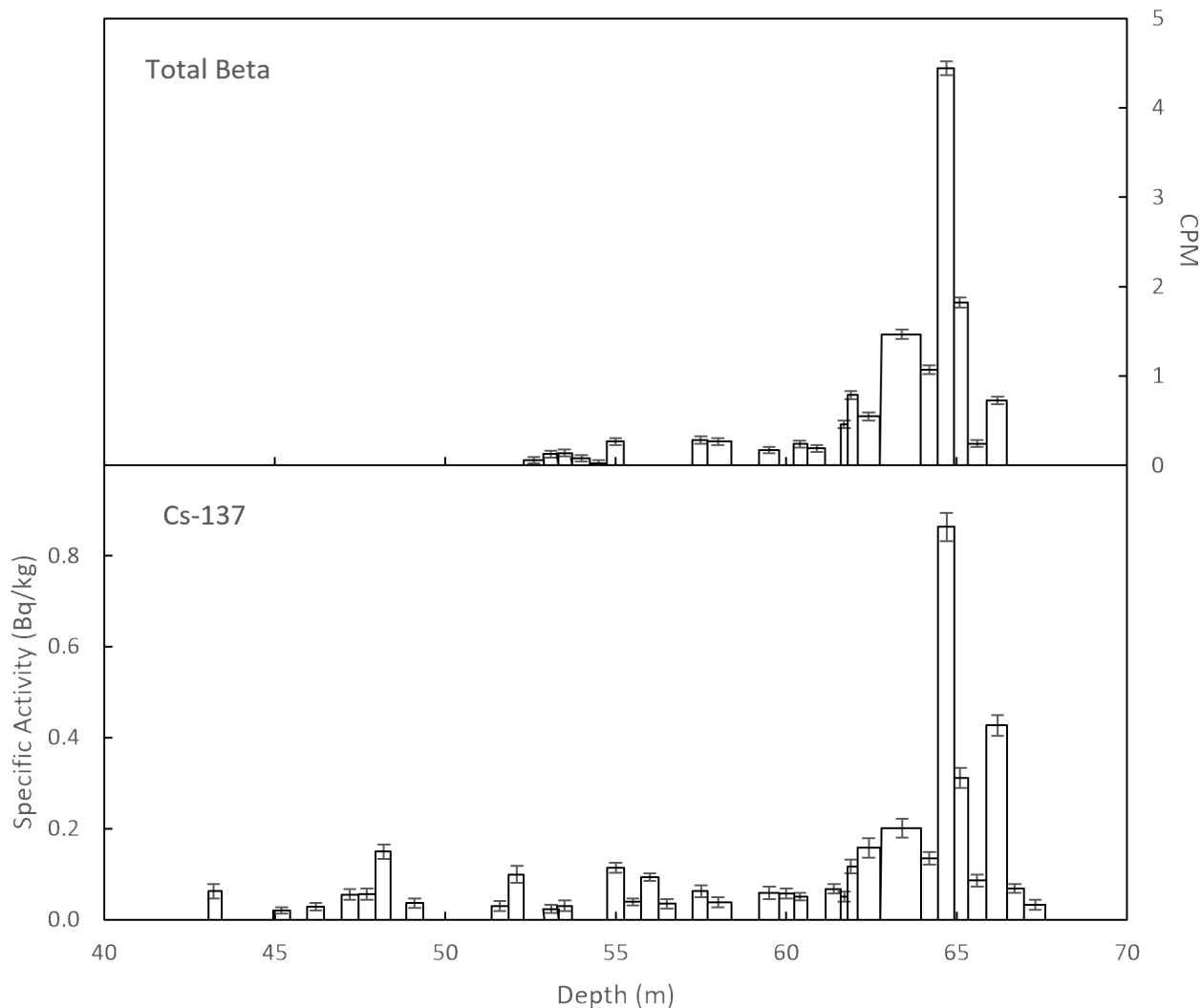
Figure 5. Total beta activity and ^{137}Cs profiles from Lys ice core. [137]Cs data taken from Clemenza et al. (2012)

Due to its position and high elevation, Lys glacier is characterized by a cold regime, as proven by the borehole temperature measurements taken at the time of drilling (Smiraglia et al., 2000). This allows the signal to be preserved in the ice without any relocation. This result is in agreement with another report from an alpine cold glacier, Grenzgletscher, where the 1963 peak for
205 beta/tritium and for cesium were found at the same depth (Eichler et al., 2000). Grenzgletscher and Lys drilling sites are less than 15 km apart.

3.3 ^{137}Cs and ^{3}H total inventories

For the Adamello ice core, we calculated the integrated activity of both ^{137}Cs and ^{3}H related to the 1963 fallout events. To do so, data in Bq L^{-1} was converted to Bq m^{-2} by considering the thickness of each sample and the ice density of 0.9 g cm^{-3}. This data was also decay corrected to 1963. Regarding ^{137}Cs, as we know there is a migration of the signal, and in order to be conservative in our calculations, we considered everything from 28 m deep downward to be possibly related to the nuclear weapons fallout; taking this into account we obtained a total inventory of ^{137}Cs of 2597 ± 400 Bq m^{-2}, where the main contribution is given by a single sample (1519 ± 228 Bq m^{-2}). This value is at the lower limit with respect to what observed in other alpine glaciers (3500 ± 800 Bq m^{-2})(Eichler et al., 2000) and in European soils (e.g. Bossew et al. (2001) reports a value of ^{137}Cs from bomb fallout of 2300 Bq m^{-2} in 1986, which corresponds to approximately 3800 Bq m^{-2} if decay corrected to 1963). These results indicate partial signal loss due to meltwater drainage for the ^{137}Cs signal. Looking at the high resolution tritium dataset, we calculated a total inventory, corrected to 1963, of 71000 ± 7000 Bq m^{-2} for the 1963 event (considering from 30,5 to 31 m), with the peak at 30.5 m corresponding to 7300 ± 700 Bq m^{-2}. As a comparison, Oerter and Rauert (1982) in a study of two ice cores extracted from Vernagtferner Glacier (Austrain Alps), sampled at a similar thickness resolution than ours, found the highest tritium values, corrected to 1963, to be 4500 and 5300 Bq m^{-2}. In our low resolution dataset, the single highest sample corresponds to 75000 ± 5500 Bq m^{-2}. In order to roughly compare our data to literature, we converted data reported in literature to Bq m^{-2} and decay corrected it to 1963, as seen in Table 1. Our data is comparable if not higher than reported tritium activities in other ice cores, suggesting that tritium in the Adamello ice core was not lost due to melting, although the region around the main peak appears to be broader than what seen in literature, leading to activity values in Bq L^{-1} to be lower than expected, likely because of diffusion processes.

Table 1. Activity values for the 1963 tritium peak at different locations. Data taken from literature were converted to Bq m^{-2} considering an ice density of 0.9 g cm^{-3} and decay corrected to 1963, but are to be considered rough approximations. Reported data refers to the single highest sample if not stated otherwise.

Ice Core Location	Reference	1963 Tritium Inventory (Bq m^{-2})
Geladaindong, Tibetan Plateau	Kang et al. (2015)	84000*
Grenzglestcher, Swiss Alps	Eichler et al. (2000)	25700
Jungfraujoch, Swiss Alps	Schotterer et al. (1977)	47800
Adamello, Italian Alps	This work (Ice core)	71000**
Adamello, Italian Alps	This work (Ice chips)	75000

*Sum of 3 different datapoints

**Sum of all datapoints between 30.5 and 31 m

3.4 Signal Preservation in the Adamello ice core

Evidence from the Adamello ice core has shown that proxies in temperate glacier must be approached with caution. In Festi et al. (2021) it is observed that peaks in pollen concentration and black carbon concentration with exceptional high values may represent multiple years condensed in one single layer as a result of negative mass balance years and enrichment of the impurities at the exposed surface. This poses an issue for annual layer counting. Despite this, due to the alignment of many palynomorphs and BC peaks, the seasonality of the signal seems to be preserved. On the contrary, in Festi et al. (2021) the ^{210}Pb profile did not show a clear exponential decrease in activity concentrations with increasing depth, as it is typically observed in glacier ice. ^{210}Pb concentrations have shown in literature large fluctuations connected with dirt horizons (Gaggeler et al., 1983) indicating that ^{210}Pb may be transported with water, thus preventing a meaningful dating of temperate glaciers with this nuclide. ^{210}Pb thus shows a behaviour in ice similar to what observed for cesium: these highly unsoluble elements remain bound to particulate matter and are prone to relocation and loss of signal when meltwater is present. Unexpectedly, in this ice core the cesium peak was not coincident with a dust layer. Furthermore, by taking into account the highest ^{137}Cs peak and comparing it to activity levels attributed to 1963 found at Colle Gnifetti (Eichler et al., 2000), we found a loss of expected signal of more than 50%. On the other hand, looking at tritium the record seems to be mostly preserved at least for the period post 1960s (as the 1954 and 1958 peaks were not detected in this ice core), because the shape of the tritium profile matches the tritium precipitation record and because comparing the total inventory, no loss in tritium activity is found; this is further confirmation about the better preservation of tritium compared to cesium. We thus believe that Adamello glacier can still function as a paleoclimate archive but to correctly interpret the information thereby contained, it is necessary to consider melting and disturbances processes affecting the climatic signals.

4 Conclusions

We analyzed tritium with Liquid Scintillation Counting in the entire 2016 Adamello ice core with a resolution of approximately 0.5 m, and with an increased resolution for the portion between 27 and 35 m of depth. Our analysis showed contamination of tritium between 19 and 32 m of depth, which can be attributed to the worldwide radioactive contamination caused by atmospheric nuclear bomb testing in the 1950s and 1960s. Additionally, we were able to compare the ^{3}H profile with another byproduct of nuclear bomb testing, ^{137}Cs. The analysis revealed that the radioactive peak associated with 1963 is not coincident for the two artificial radionuclides. The ^{3}H peak occurs 1.5 m above the ^{137}Cs one. As many records agree showing that the deposition of the two radioactive species was concurrent, the shift of the peaks is only explainable assuming that the ^{137}Cs peak has been subject to a downward relocation triggered by meltwater percolation in temperate ice. Tritium was not affected by the process as it is present in the ice matrix. While the total tritium inventory associated with the 1963 peak in the Adamello ice core is comparable to what is reported in literature for cold glaciers not affected by melting, the same is not for ^{137}Cs. The total inventory of ^{137}Cs calculated at the 1963 peak for this radionuclide is among the lowest ever reported, providing additional evidence that meltwater disturbed its distribution into the ice, partially washing it away.

Such finding poses issues about the established application of radionuclides for dating mountain ice cores. While for cold glacier it is reasonable to assume that radioactive signals are well preserved in the ice column, for temperate ice cores this is not the case. To avoid potential dating inaccuracies, it is preferable to reconstruct the entire profile of ^{3}H to obtain a stronger indication of the true 1963 signal depth. In general, when disturbances are present in the reconstructed profile, matrix signals such as tritium are more easily preserved. Additional studies will be needed to investigate the relocation mechanism of cesium in the presence of meltwater.

Author contributions. ED and MC conceived the idea of this work. ED carried out the sample preparation for liquid scintillation counting. DB and RG acquired and analyzed the Hypersectral Data. ED, GB and BD intepreted the data. ED prepared the manuscript with contributions from all co-authors. MS and VM provided additional editing and review.

Competing interests. The authors declare that they have no conflict of interest

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References

Avak, S. E., Schwikowski, M., and Eichler, A.: Impact and implications of meltwater percolation on trace element records observed in a high-Alpine ice core, J. Glaciol., 64, 877–886, 2018.

Baccolo, G., Łokas, E., Gaca, P., Massabò, D., Ambrosini, R., Azzoni, R. S., Clason, C., Di Mauro, B., Franzetti, A., Nastasi, M., Prata, M., Prati, P., Previtali, E., Delmonte, B., and Maggi, V.: Cryoconite: An efficient accumulator of radioactive fallout in glacial environments, Cryosphere, 14, 657–672, https://doi.org/10.5194/tc-14-657-2020, 2020.

Bossew, P., Ditto, M., Falkner, T., Kienzl, K., and Rappelsberger, U.: Contamination of Austrian soil with caesium-137, J. Environ. Radioact., 55, 187–194, 2001.

Clemenza, M., Cucciati, G., Maggi, V., Pattavina, L., and Previtali, E.: Radioactive fallouts as temporal makers for glacier ice cores dating, Eur. Phys. J. Plus, 127, 68, 2012.

Currie, L. A.: Limits for qualitative detection and quantitative determination. Application to radiochemistry, Anal. Chem., 40, 586–593, 1968.

Davaze, L., Rabatel, A., Dufour, A., Hugonnet, R., Arnaud, Y., and Braithwaite, R. J.: Region-Wide Annual Glacier Surface Mass Balance for the European Alps From 2000 to 2016, 8, 1–14, https://doi.org/10.3389/feart.2020.00149, 2020.

Di Mauro, B., Fava, F., Ferrero, L., Garzonio, R., Baccolo, G., Delmonte, B., and Colombo, R.: Mineral dust impact on snow radiative properties in the European Alps combining ground, UAV, and satellite observations, J. Geophys. Res., 120, 6080–6097, https://doi.org/10.1002/2015JD023287, 2015.

Di Stefano, E., Clemenza, M., Baccolo, G., Delmonte, B., and Maggi, V.: 137Cs contamination in the Adamello glacier: Improving the analytical method, J. Environ. Radioact., 208-209, 106 039, https://doi.org/10.1016/j.jenvrad.2019.106039, 2019.

Eichler, A., Schwikowski, M., Gäggeler, H. W., Furrer, V., Synal, H.-A., Beer, J., Saurer, M., and Funk, M.: Glaciochemical dating of an ice core from upper Grenzgletscher (4200 m asl), J. Glaciol., 46, 507–515, 2000.

Eichler, A., Schwikowski, M., and Gäggeler, H. W.: Meltwater-induced relocation of chemical species in Alpine firn, Tellus, Ser. B Chem. Phys. Meteorol., 53, 192–203, https://doi.org/10.3402/tellusb.v53i2.16575, 2001.

Festi, D., Schwikowski, M., Maggi, V., Oeggl, K., and Jenk, T. M.: Significant mass loss in the accumulation area of the Adamello glacier indicated by the chronology of a 46m ice core, Cryosphere, 15, 4135–4143, https://doi.org/10.5194/tc-15-4135-2021, 2021.

Gabrieli, J., Cozzi, G., Vallelonga, P., Schwikowski, M., Sigl, M., Eickenberg, J., Wacker, L., Boutron, C., Gäggeler, H., Cescon, P., and Barbante, C.: Contamination of Alpine snow and ice at Colle Gnifetti , Swiss / Italian Alps , from nuclear weapons tests, Atmos. Environ., 45, 587–593, https://doi.org/10.1016/j.atmosenv.2010.10.039, 2011.

Gabrielli, P., Carturan, L., Gabrieli, J., Dinale, R., Krainer, K., Hausmann, H., Davis, M., Zagorodnov, V., Seppi, R., Barbante, C., Fontana, G. D., and Thompson, L. G.: Atmospheric warming threatens the untapped glacial archive of Ortles mountain, South Tyrol, J. Glaciol., 56, 843–853, https://doi.org/10.3189/002214310794457263, 2010.

Gaggeler, H., Von Gunten, H. R., Rossler, E., Oeschger, H., and Schotterer, U.: 210Pb dating of cold alpine firn/ ice cores from Colle Gnifetti, Switzerland., J. Glaciol., 29, 165–177, https://doi.org/10.1017/S0022143000005220, 1983.

Garzonio, R., Di Mauro, B., Cogliati, S., Rossini, M., Panigada, C., Delmonte, B., Maggi, V., and Colombo, R.: A novel hyperspectral system for high resolution imaging of ice cores: Application to light-absorbing impurities and ice structure, Cold Reg. Sci. Technol., 155, 47–57, https://doi.org/10.1016/j.coldregions.2018.07.005, 2018.

Gilbert, A., Wagnon, P., Vincent, C., Ginot, P., and Funk, M.: Atmospheric warming at a high-elevation tropical site revealed by englacial temperatures at Illimani, Bolivia (6340 m above sea level, 16°S, 67°W), J. Geophys. Res. Atmos., 115, 1–10, https://doi.org/10.1029/2009JD012961, 2010.

315 Gilbert, A., Sinisalo, A., R. Gurung, T., Fujita, K., B. Maharjan, S., C. Sherpa, T., and Fukuda, T.: The influence of water percolation through crevasses on the thermal regime of a Himalayan mountain glacier, Cryosphere, 14, 1273–1288, https://doi.org/10.5194/tc-14-1273-2020, 2020.

Harms, P. A., Visser, A., Moran, J. E., and Esser, B. K.: Distribution of tritium in precipitation and surface water in California, J. Hydrol., 534, 63–72, https://doi.org/10.1016/j.jhydrol.2015.12.046, 2016.

320 Hock, R., Bliss, A., Marzeion, B. E. N., Giesen, R. H., Hirabayashi, Y., Huss, M., and C, V. R.: GlacierMIP – A model intercomparison of global-scale glacier mass-balance models and projections, 65, 453–467, https://doi.org/10.1017/jog.2019.22, 2019.

Hoelzle, M., Darms, G., Lüthi, M. P., and Suter, S.: Evidence of accelerated englacial warming in the Monte Rosa area, Switzerland/Italy, Cryosphere, 5, 231–243, https://doi.org/10.5194/tc-5-231-2011, 2011.

International Atomic Energy Agency: Environmental Consequences of the Chernobyl Accident and their Remediation: Twenty years of
325 Experience, Tech. rep., https://doi.org/10.1007/bf02850918, 2006.

International Atomic Energy Agency and World Meteorological Organization: Global Network of Isotopes in Precipitation. The GNIP Database. Accessible at: https://nucleus.iaea.org/wiser, 2021.

Jouzel, J., Merlivat, L., Pourchet, M., and Lorius, C.: A Countinuous Record of Artificial Tritium Fallout at the South Pole (1954-1978), Earth Planet. Sci. Lett., 45, 188–200, 1979.

330 Kang, S., Wang, F., Morgenstern, U., Zhang, Y., Grigholm, B., Kaspari, S., Schwikowski, M., Ren, J., Yao, T., Qin, D., and Others: Dramatic loss of glacier accumulation area on the Tibetan Plateau revealed by ice core tritium and mercury records, Cryosph., 9, 1213–1222, 2015.

Kaufman, S. and Libby, W. F.: The natural distribution of tritium, Phys. Rev., 93, 1337–1344, https://doi.org/10.1103/PhysRev.93.1337, 1954.

Lliboutry, L.: Permeability, Brine Content and Temperature of Temperate Ice, J. Glaciol., 10, 15–29,
335 https://doi.org/10.3189/s002214300001296x, 1971.

Lucas, L. L. and Unterweger, M. P.: Comprehensive Review and Critical Evaluation, 105, 541–549, 2000.

Masarik, J. and Beer, J.: An updated simulation of particle fluxes and cosmogenic nuclide production in the Earth's atmosphere, J. Geophys. Res. Atmos., 114, 1–9, https://doi.org/10.1029/2008JD010557, 2009.

Meyer, T. and Wania, F.: Modeling the elution of organic chemicals from a melting homogeneous snow pack, Water Res., 45, 3627–3637,
340 https://doi.org/10.1016/j.watres.2011.04.011, 2011.

Michel, R. L.: Tritium in the hydrologic cycle, in: Isot. Water Cycle Past, Present Futur. a Dev. Sci., pp. 53–66, https://doi.org/10.1007/1-4020-3023-1_5, 2005.

Mikhalenko, V., Sokratov, S., Kutuzov, S., Ginot, P., Legrand, M., Preunkert, S., Lavrentiev, I., and Kozachek, A.: Investigation of a deep ice core from the Elbrus western plateau , the Caucasus , Russia, Cryosph., pp. 2253–2270, https://doi.org/10.5194/tc-9-2253-2015, 2015.

345 Moser, D. E., Thomas, E. R., Nehrbass-Ahles, C., Eichler, A., and Wolff, E.: Melt-Affected Ice Cores for (Sub-)Polar Research in a Warming World, Egusph. Prepr., pp. 1–48, https://doi.org/10.5194/egusphere-2023-1939, 2023.

Niu, H., Kang, S., Shi, X., Paudyal, R., He, Y., Li, G., Wang, S., Pu, T., and Shi, X.: In-situ measurements of light-absorbing impurities in snow of glacier on Mt. Yulong and implications for radiative forcing estimates, Sci. Total Environ., 581-582, 848–856, https://doi.org/10.1016/j.scitotenv.2017.01.032, 2017.

350 Oerter, H. and Rauert, W.: Core drilling on Vernagtferner (Oetztal Alps, Austria) in 1979: tritium contents., Zeitschrift fur Gletscherkd. und Glazialgeol., 18, 13–22, 1982.

Pavlova, P. A., Jenk, T. M., Schmid, P., Bogdal, C., Steinlin, C., and Schwikowski, M.: Polychlorinated biphenyls in a temperate alpine glacier: 1. Effect of percolating meltwater on their distribution in glacier ice, Environ. Sci. Technol., 49, 14 085–14 091, 2015.

Picciotto, E. and Wilgain, S.: Fission Products in Antarctic Snow, A Reference Level for Measuring Accumulation, J. Geophys. Res., 68, 5965–5972, 1963.

Picotti, S., Francese, R., Giorgi, M., Pettenati, F., and Carcione, J. M.: Estimation of glacier thicknesses and basal properties using the horizontal-to-vertical component spectral ratio (HVSR) technique from passive seismic data, J. Glaciol., 63, 229–248, https://doi.org/10.1017/jog.2016.135, 2017.

Pinglot, J. F., Vaikmäe, R. A., Kamiyama, K., Igarashi, M., Fritzsche, D., Wilhelms, F., Koerner, R., Henderson, L., Isaksson, E., Winther, J.-G., Others, Vaikmae, R. A., Kamiyama, K., Igarashi, M., Fritzsche, D., Wilhelms, F., Koerner, R., Henderson, L., Isaksson, E., Winther, J.-G., and Others: Ice cores from Arctic sub-polar glaciers: chronology and post-depositional processes deduced from radioactivity measurements, J. Glaciol., 49, 149–158, 2003.

Pourchet, M. and Pinglot, F.: Determination of the stratospheric residence time from the total beta activity of Antarctic and Greenland snows, Geophys. Res. Lett., 6, 6–8, 1979.

365 Qiao, J., Colgan, W., Jakobs, G., and Nielsen, S.: High-Resolution Tritium Pro fi le in an Ice Core from Camp Century, Greenland, https://doi.org/10.1021/acs.est.1c01975, 2021.

Qin, H., Yokoyama, Y., Fan, Q., Iwatani, H., Tanaka, K., Sakaguchi, A., Kanai, Y., Zhu, J., Onda, Y., and Takahashi, Y.: Investigation of cesium adsorption on soil and sediment samples from Fukushima prefecture by sequential extraction and EXAFS technique, Geochem. J., 46, 297–302, https://doi.org/10.2343/geochemj.2.0214, 2012.

370 Ranzi, R., Grossi, G., Gitti, A., and Taschner, S.: Energy and mass balance of the mandrone glacier (Adamello, Central Alps), Geogr. Fis. e Din. Quat., 33, 45–60, 2010.

Schotterer, U., Finkel, R. C., Oeschger, H., Wahlen, M., Bart, G., and Gäggeler, H. W.: Isotope measurements on firn and ice cores from alpine glaciers., Iahs, pp. 232–236, 1977.

Schotterer, U., Schwarz, P., and V. Rajner.: FROM PRE-BOMB LEVELS TO INDUSTRIAL TIMES A complete tritium record from an alpine ice core and its relevance for environmental studies, 1998a.

Schotterer, U., Schwarz, P., and V. Rajner.: From pre-bomb levels to industrial times: A complete tritium record from an alpine ice core and its relevance for environmental studies, Isot. Tech. study Environ. Chang., 1998b.

Schwikowski, M., Briitsch, S., G, H. W., Schotterer, U., and Scherrer, P.: A high-resolution air chemistry record from an Alpine ice core : Fiescherhorn glacier , Swiss Alps suitable glaciers are often small in the horizontal and hindered by atmospheric stability [Baltensperger 380 extension , thus limiting the accessible timescale, Europe, 104, 709–719, 1999.

Smiraglia, C., Maggi, V., Novo, A., Rossi, G., Johnston, P., Sr, A., Maggi, V., Gc, R., and Johnston, P.: Premilinary Results of Two Ice Core Drillings on Monte Rosa (Colle Gnifetti and Colle del Lys), Italian Alps, Geogr. Fis. e Din. Quat., pp. 165–172, 2000.

Steinhauser, G., Brandl, A., and Johnson, T. E.: Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts, Sci. Total Environ., 470-471, 800–817, https://doi.org/10.1016/j.scitotenv.2013.10.029, 2014.

385 Tanaka, K., Sakaguchi, A., Kanai, Y., Tsuruta, H., Shinohara, A., and Takahashi, Y.: Heterogeneous distribution of radiocesium in aerosols, soil and particulate matters emitted by the Fukushima Daiichi Nuclear Power Plant accident: Retention of micro-scale het-

erogeneity during the migration of radiocesium from the air into ground and rive, J. Radioanal. Nucl. Chem., 295, 1927–1937, https://doi.org/10.1007/s10967-012-2160-9, 2013.

Thatcher, L. L.: The distribution of tritium fallout in precipitation over north america, Int. Assoc. Sci. Hydrol. Bull., 7, 48–58,
390   https://doi.org/10.1080/02626666209493255, 1962.

Thompson, L. G., Mosley-Thompson, E., Davis, M., Lin, P. N., Yao, T., Dyurgerov, M., and Dai, J.: "Recent warming": ice core evidence from tropical ice cores with emphasis on Central Asia, Glob. Planet. Change, 7, 145–156, https://doi.org/10.1016/0921-8181(93)90046-Q, 1993.

United Nations Scientific Committee on the Effects of Atomic Radiation: Sources and Effects of Ionizing R adiation, Tech. rep., www.
395   unscear.org/docs/reports/1993/1993c{_}pages91-120.pdf{%}0Awww.unscear.org/docs/reports/annexc.pdf, 2000.

United Nations Scientific Committee on the Effects of Atomic Radiation: Sources And Effects of Ionizing Radiation, Tech. Rep. 4, https://doi.org/10.1093/oxfordjournals.rpd.a079988, 2008.

Villa, S., Negrelli, C., Maggi, V., Finizio, A., and Vighi, M.: Analysis of a firn core for assessing POP seasonal accumulation on an Alpine glacier, Ecotoxicol. Environ. Saf., 63, 17–24, https://doi.org/10.1016/j.ecoenv.2005.05.006, 2006.

400   Wagenbach, D., Bohleber, P., Preunkert, S., Wagenbach, D., Bohleber, P., and Preunkert, S.: Cold , alpine ice bodies revisited : what may we learn from their impurity and isotope content ?, Geogr. Ann. Ser. A, Phys. Geogr. ISSN, 3676, https://doi.org/10.1111/j.1468-0459.2012.00461.x, 2012.

Wong, G. J., Hawley, R. L., Lutz, E. R., and Osterberg, E. C.: Trace-element and physical response to melt percolation in Summit (Greenland) snow, Ann. Glaciol., 54, 52–62, https://doi.org/10.3189/2013AoG63A602, 2013.

405   Yasunari, T. J., Shiraiwa, T., Kanamori, S., Fujii, Y., Igarashi, M., Yamazaki, K., Benson, C. S., and Hondoh, T.: Intra-annual variations in atmospheric dust and tritium in the North Pacific region detected from an ice core from Mount Wrangell, Alaska, J. Geophys. Res. Atmos., 112, 1–16, https://doi.org/10.1029/2006JD008121, 2007.

Zebre, M., Colucci, R. R., Giorgi, F., Glasser, N. F., Racoviteanu, A. E., and Del Gobbo, C.: 200 years of equilibrium-line altitude variability across the European Alps (1901-2100), Clim. Dyn., 56, 1183–1201, https://doi.org/10.1007/s00382-020-05525-7, 2021.

410   Zemp, M., Huss, M., Thibert, E., Eckert, N., Mcnabb, R., Huber, J., Barandun, M., and Machguth, H.: Global glacier mass changes and their contributions to sea-level rise from 1961 to 2016, pp. 2–37, 2019.