



Evidence of radionuclide fractionation due to meltwater percolation in a temperate glacier

Elena Di Stefano^{1,2}, Giovanni Baccolo³, Massimiliano Clemenza², Barbara Delmonte¹, Deborah Fiorini¹, Roberto Garzonio¹, Margit Schwikowski³, and Valter Maggi¹

¹Environmental and Earth Sciences Department, University Milano-Bicocca, 20126 Milano, Italy

²Milano-Bicocca Section, Istituto Nazionale di Fisica Nucleare, 20126 Milano, Italy

³Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Correspondence: Elena Di Stefano (elena.distefano@unimib.it)

Abstract. The article discusses the use of ³H and ¹³⁷Cs as temporal markers in ice cores extracted from temperate glaciers. We present a complete tritium (³H) profile for a 46 m ice core drilled from Adamello glacier, a temperate glacier in the Italian Alps, and compare it to ¹³⁷Cs 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 ³H and ¹³⁷Cs, but the ³H peak occurs 1.5 m above the ¹³⁷Cs one. This misalignment is caused by meltwater-induced postdepositional processes that affect ¹³⁷Cs, which is more sensitive to percolation than ³H. The total inventory of ¹³⁷Cs 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 ± 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 (Gabielli et al., 2010). Temperate ice is defined as ice that because of its thermodynamic state can contain meltwater inclusions (Liboutry, 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
25 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
30 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) such as 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
35 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 contamina-
tion of the environment. Atmospheric thermonuclear explosions can reach the stratosphere, where the circulation allows the
40 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,
45 USSR, and UK. This maximum has been registered in paleoclimatic archives and has since been used as a temporal reference
horizon (Mikhalevko 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 ^3H , and the pres-
50 ence 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 (^3H) 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
55 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 $0.320 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Masarik and Beer, 2009), resulting in a low natural background detectable in precipitations; this was estimated to be $<1 \text{ Bq L}^{-1}$ 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).

^3H 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. ^3H has been extensively used as a temporal horizon marker for 1963 in ice cores (Eichler et al., 2000; Qiao et al., 2021; Mikhaleenko 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: $10^{\circ}31'22'' \text{ E}$, $46^{\circ}8'51'' \text{ N}$), 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 ($268 \pm 5 \text{ m}$) (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

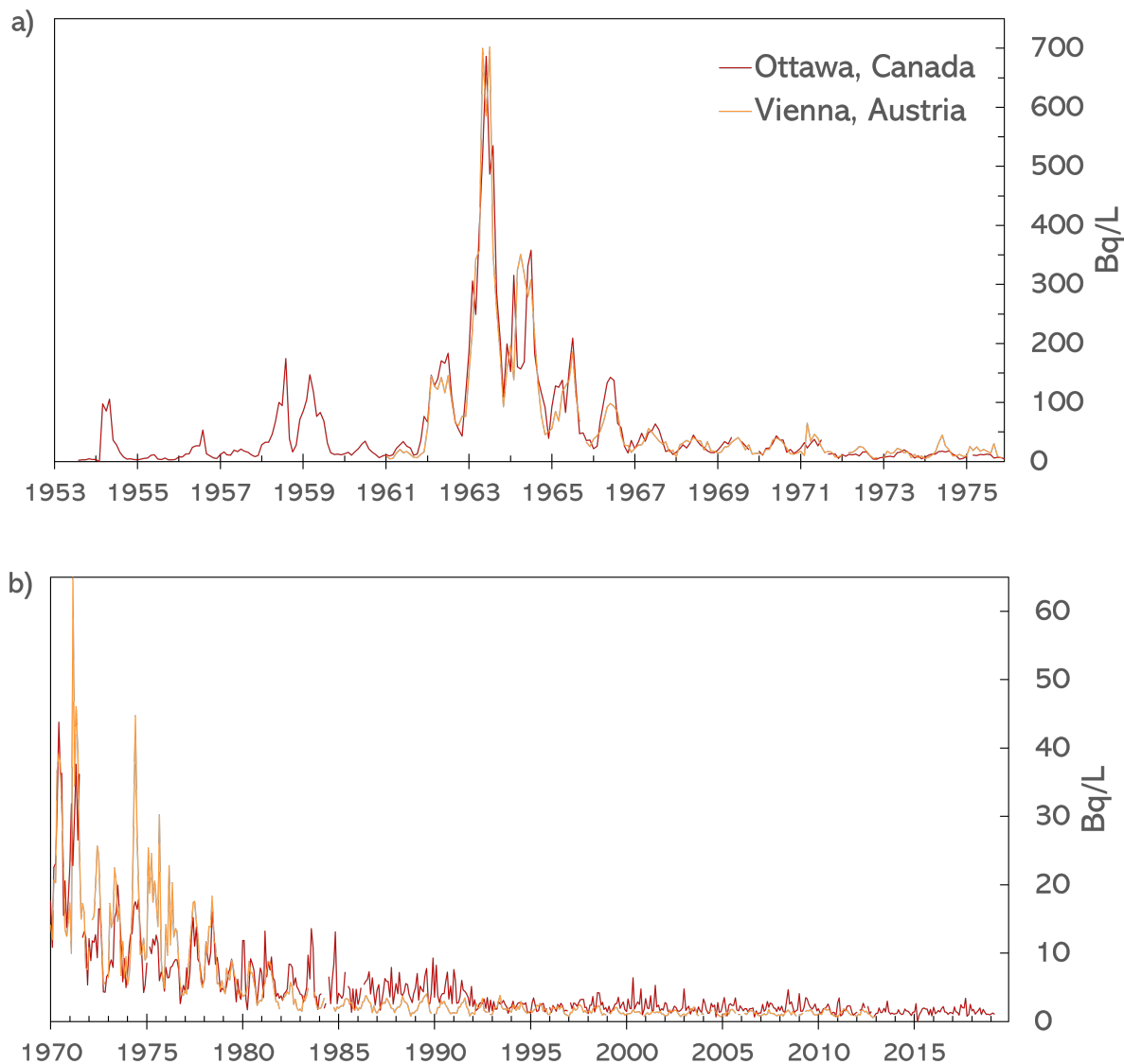


Figure 1. Monthly concentration of tritium in precipitation in Vienna (Austria) and Ottawa (Canada) from 1953 to 1975 (a) and from 1971 to 2019 (b). Data from the Global Network of Isotopes in Precipitation (GNIP) Database (International Atomic Energy Agency and World Meteorological Organization, 2021), 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.

95 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
100 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 ¹³⁷Cs analysis that was carried out by Clemenza et al. (2012).

2 Materials and Methods

2.1 Sample Preparation and Analysis

105 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
110 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
115 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
120 Hydrogen-3 Radioactivity Standard 4361C. 3 sets of standards (Set1-2-3) were prepared following the same procedure which

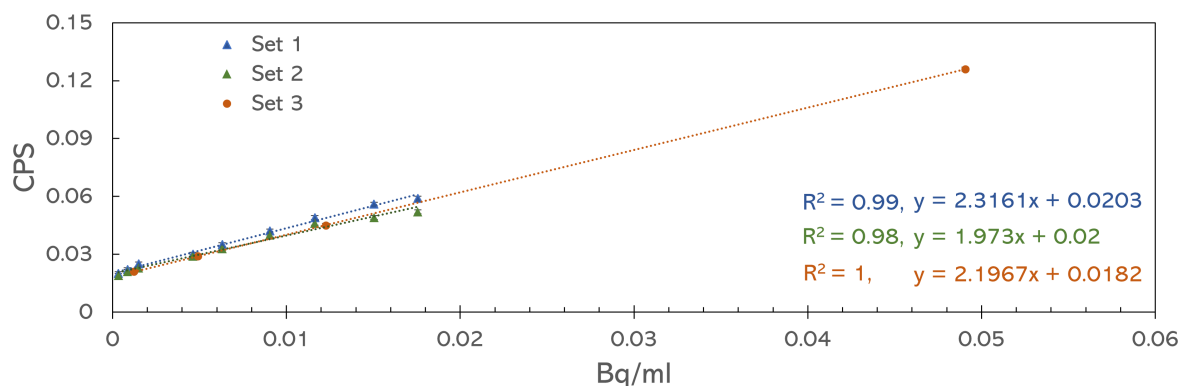


Figure 2. Calibration curves calculated for the 3 sets of tritium standards. Errors on cps are 0.001.

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 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 ($R_s - R_b$) 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 $(R_s - R_b) < LC$, an upper limit 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 3.3σ 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; 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 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).

150 3 Results and Discussion

Results are presented in Figures 3 and 4. 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.

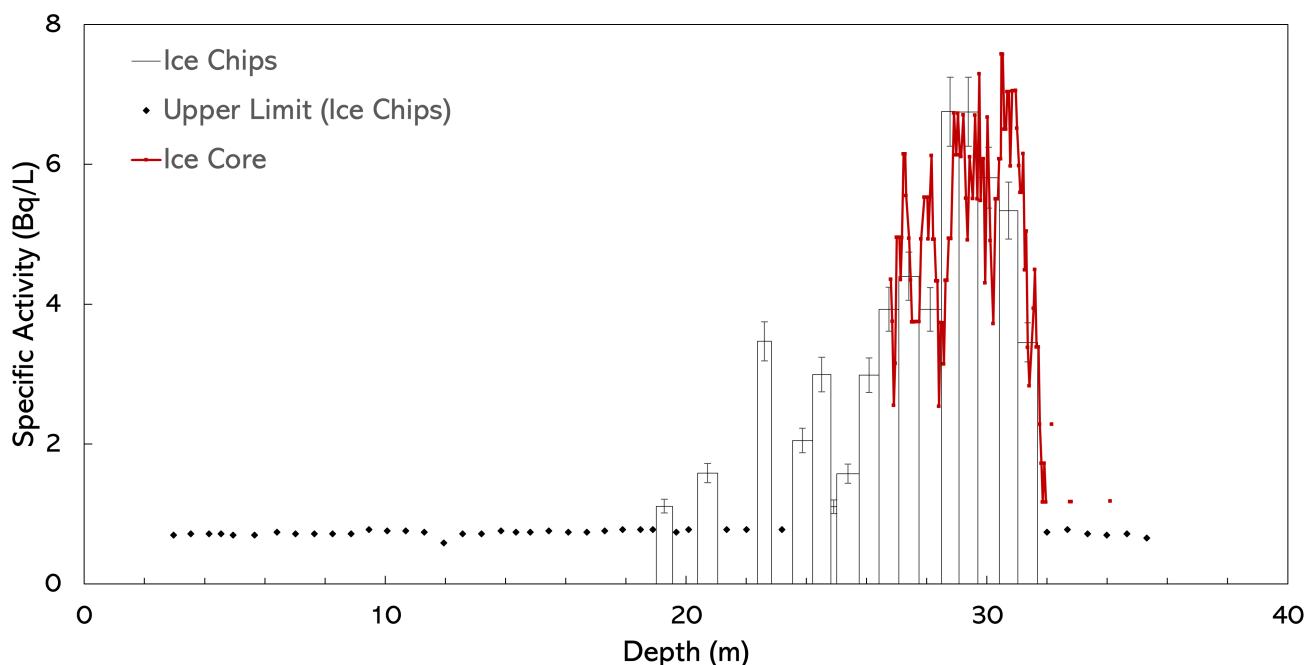


Figure 3. Tritium activity in the Adamello temperate ice core. Data reported in black are related to low resolution measurements (ice chips), diamonds indicate upper limits, bars observed activities. In red results from high-resolution analyses conducted on ice core sections.

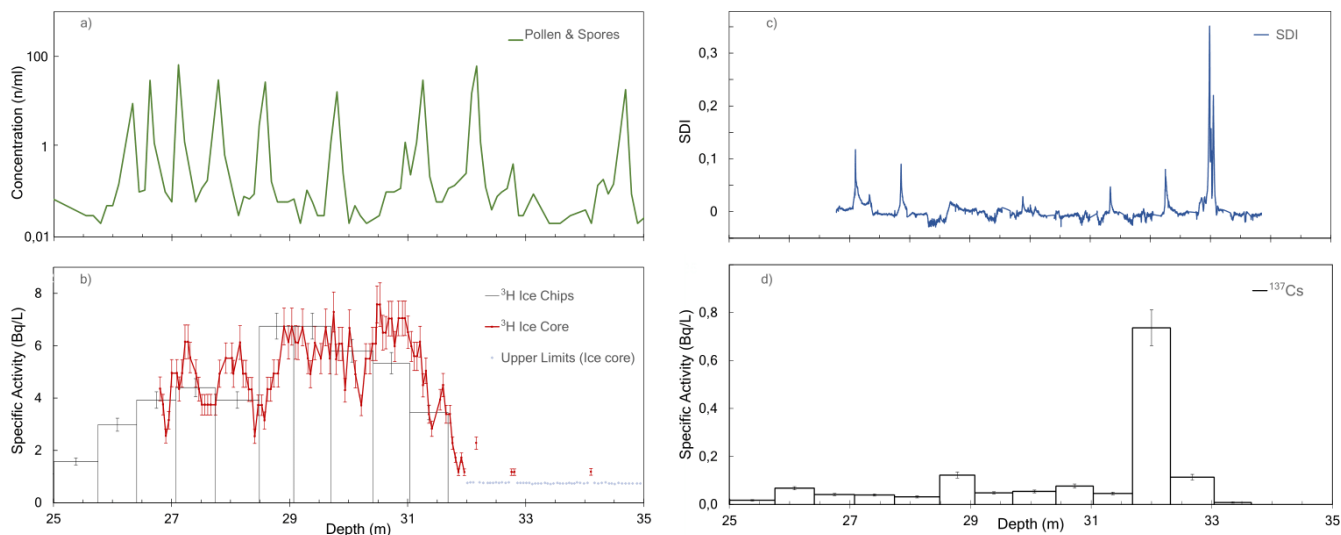


Figure 4. Detailed overview of the ice core in the portion where the the tritium and ^{137}Cs 1963 peaks have been observed (between 25 and 35 m deep). a) Pollen and spores concentration taken from Festi et al. (2021). Each peak of pollen and spores concentration reflects one flowering year (February–September). b) Tritium profiles. Specific activity in Bq L^{-1} of ^3H 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 tritium signal was further investigated by analyzing the high resolution dataset between 27 and 35 m. In general, good
155 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. 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
160 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.

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
165 are almost the same (Pourchet and Pinglot, 1979), it is expected to find the 1963 peak at the same depth for ^{137}Cs and ^3H , as
observed in ice cores from cold glaciers (Pinglot et al., 2003).

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 5. 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. We must note that

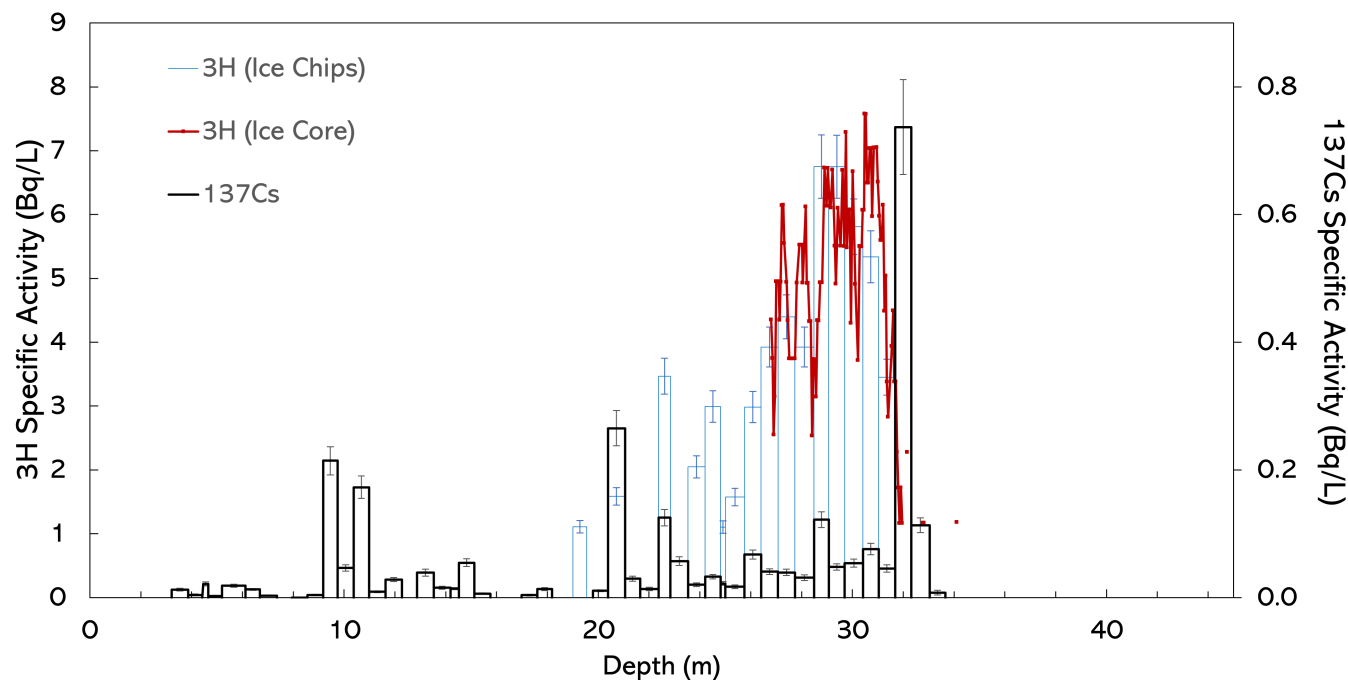


Figure 5. Comparison between ^{137}Cs (data from Di Stefano et al. (2019)) and ^3H

170 Festi et al. (2021) used data from Di Stefano et al. (2019) as a tie point for 1963, so this dating could be susceptible to some degree of error. Because Adamello is a temperate glacier, we cannot in fact rule out the possibility that the ^{137}Cs signal may have been altered.

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
175 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.

The explanation we propose to explain the lag between the ^3H and the ^{137}Cs signals is the presence of meltwater-induced
180 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 ^3H (Pinglot et al., 2003), which is incorporated in the water molecule and can be regarded as a matrix signal. Pinglot et al. (2003) found a similar delay between cesium and tritium in ice cores extracted from low altitude glaciers in Svalbard, although the difference between the ^3H peak and the ^{137}Cs was less marked (<1 m), which can be expected since Adamello glacier has experienced much more melting than Svalbard
185 and much 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).

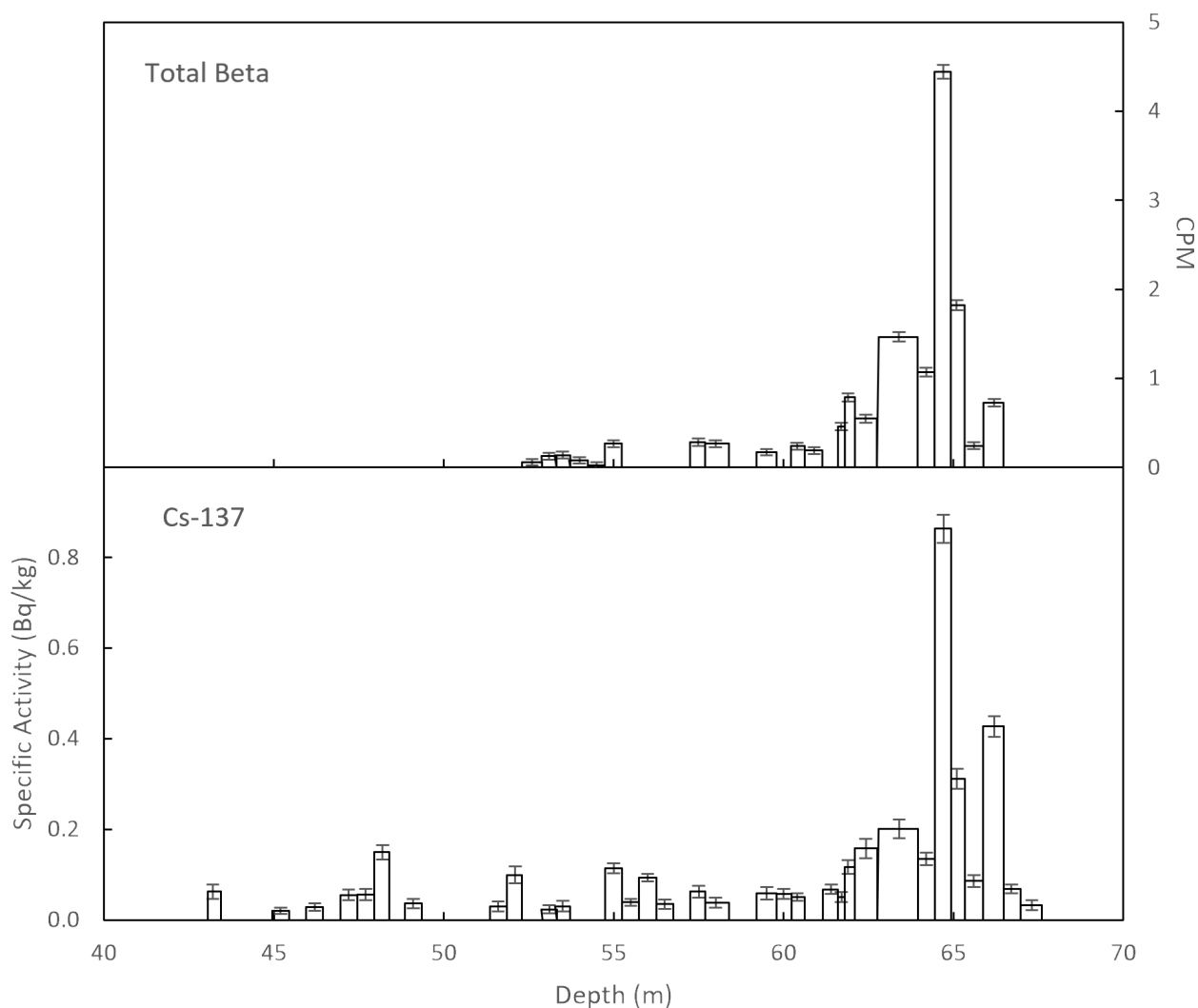


Figure 6. Total beta activity and ^{137}Cs profiles from Lys ice core. ^{137}Cs data taken from Clemenza et al. (2012)

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 6 shows that the main beta (^3H) peak and the main ^{137}Cs coincide. 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 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.

195 We calculated the integrated activity of both ^{137}Cs and ^3H 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 $2597 \pm 400 \text{ Bq m}^{-2}$, where the main contribution is given by
200 a single sample ($1519 \pm 228 \text{ Bq m}^{-2}$). This value is at the lower limit with respect to what observed in other alpine glaciers ($3500 \pm 800 \text{ Bq m}^{-2}$) (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 $71000 \pm 7000 \text{ Bq m}^{-2}$ for the 1963 event (considering from 30,5 to 31 m),
205 with the peak at 30.5 m corresponding to $7300 \pm 700 \text{ Bq m}^{-2}$. As a comparison, Oerter and Rauert (1982) in a study of two ice cores extracted from Vernagtferner Glacier (Austrian 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 $75000 \pm 5500 \text{ Bq m}^{-2}$. 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
210 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*
Grenzgletscher, 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



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 ^3H 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 ^3H 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. On the contrary, ^{137}Cs , being more soluble and mobile in aqueous environments, is more prone to relocation if liquid water is present. 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 ^3H 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.

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 Hyperpectral Data. ED, GB and BD interpreted 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

Acknowledgements. This work is a contribution to the project CALICE – Calibrating Biodiversity in Glacier Ice, a multidisciplinary program between the University of Innsbruck, the Free University of Bozen-Bolzano, and the Fondazione Edmund Mach in San Michele, funded by the EVTZ/Austrian Science Fund (IPN 57- B22). This is CALICE project publication no. 2. We would like to thank all members of the CALICE scientific consortium, especially those who helped during the coring activities and the processing of the ice core. We are grateful to the ENEA drilling team and the Alpine guide Nicola Viotti (Guide Alpine Valsusa) for their excellent work during the coring campaign. Drilling has been possible thanks to a specific grant (POLLice) to FEM (Fondazione Edmund Mach) from the Autonomous Province of



Trento (PAT) and logistic support (helicopter flights) provided by Ernesto Sanutuiliana. EuroCold Lab activities were partially funded by the Italian Regional Affair Ministry.



References

- 245 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.
- 250 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.
- 255 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.
- 260 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.
- 265 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., Oeggel, 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.
- 270 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.
- 275 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.
- 280



- 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.*, 285, 534, 63–72, <https://doi.org/10.1016/j.jhydrol.2015.12.046>, 2016.
- 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.
- 290 International Atomic Energy Agency: Environmental Consequences of the Chernobyl Accident and their Remediation: Twenty years of 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 Continuous Record of Artificial Tritium Fallout at the South Pole (1954–1978), 295 *Earth Planet. Sci. Lett.*, 45, 188–200, 1979.
- 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.
- 300 Lliboutry, L.: Permeability, Brine Content and Temperature of Temperate Ice, *J. Glaciol.*, 10, 15–29, <https://doi.org/10.3189/s002214300001296x>, 1971.
- Lucas, L. L. and Unterwieser, 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.
- 305 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.
- Mikhaleenko, 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.
- 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, 310 <https://doi.org/10.1016/j.scitotenv.2017.01.032>, 2017.
- Oerter, H. and Rauer, W.: Core drilling on Vernagtferner (Oetztal Alps, Austria) in 1979: tritium contents., *Zeitschrift für 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, 14085–14091, 2015.
- 315 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.
- 320 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.
- 325 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.
- Qiao, J., Colgan, W., Jakobs, G., and Nielsen, S.: High-Resolution Tritium Profile 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.
- 330 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.
- 335 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.
- 340 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 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.: Preliminary 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.
- 345 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.
- 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 heterogeneity during the migration of radiocesium from the air into ground and river, *J. Radioanal. Nucl. Chem.*, 295, 1927–1937, <https://doi.org/10.1007/s10967-012-2160-9>, 2013.
- 350 Thatcher, L. L.: The distribution of tritium fallout in precipitation over north america, *Int. Assoc. Sci. Hydrol. Bull.*, 7, 48–58, <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](https://doi.org/10.1016/0921-8181(93)90046-Q), 1993.
- 355



- United Nations Scientific Committee on the Effects of Atomic Radiation: Sources and Effects of Ionizing Radiation, Tech. rep., www.unscear.org/docs/reports/1993/1993c_{_}pages91-120.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.
- 360 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.
- 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.
- 365 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.
- 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.
- 370 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.
- 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.
- Zhang, Q., Kang, S., Gabrielli, P., Loewen, M., and Schwikowski, M.: Vanishing High Mountain Glacial Archives: Challenges and Perspectives, *Environ. Sci. Technol.*, 49, 9499–9500, <https://doi.org/10.1021/acs.est.5b03066>, 2015.
- 375