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Solar Cycle Detected in Natural Tritium of Ice Layers Before the Nuclear Era

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Key Points:

- The link between the solar cycle and the cosmogenic tritium in precipitation has been confirmed in ice layers accumulated before the 1950s
- Our tritium record extends back to 1923, providing the longest pre-bomb tritium record
- Reconstruction of natural tritium levels can validate isotope-enabled atmospheric general circulation models

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract The 11-year cycle of the solar activity affects the production rate of cosmogenic isotopes. For tritium in precipitation, it has been just recently proven that this link exists. Here we show, for the first time, a tritium ice core time series which extends back to 1923, covering a time period that avoids the presence of artificial tritium from the thermonuclear weapon tests of the early 1950s. Accurate analyses of low-level tritium enables us to estimate the natural level of tritium in the study site of Colle Gnifetti, Swiss-Italian Alps, as well as its variation. Statistical analyses using sunspot number and the count rate of cosmic ray secondary neutrons have confirmed that the modulation of the solar activity does affect the tritium concentration of ice layers accumulated earlier than the first hydrogen bomb tests. The tritium level of the ice, as well as in precipitation, is still slightly decreasing during the last three decades. The natural level of tritium obtained in this work fits very well to early tritium analyses of European wine samples, as well as model calculations with the isotope-enabled atmospheric general circulation model MIROC5-iso. Further sensitive tritium analyses of ice cores around the world will provide the opportunity to validate these models.

Plain Language Summary Tritium is an excellent tracer of the water cycle dynamics. It is produced in the upper atmosphere in nuclear reactions of mainly cosmic ray secondary neutrons and atmospheric nuclei. After its production, it enters the water cycle, and reaches the surface in precipitation. It has been just recently proven that the concentration of tritium in precipitation is affected by the 11-year solar cycle. In our study we aimed to confirm that this correlation is visible in ice layers which are not contaminated with anthropogenic tritium emissions from thermonuclear weapon tests nor from industry. The tritium time series of the ice core retrieved in Colle Gnifetti, Swiss-Italian Alps, shows an evident correlation to the solar cycle and provides a good estimation of the natural level of tritium, which fits very well to early tritium analyses of European wine samples and model calculations. Further tritium analyses of ice cores around the world will provide the opportunity to validate isotope-enabled climate models. Our study provides critical insight into the natural variability of cosmogenic tritium, offering a valuable reference for climate models.

1. Introduction

Tritium (^{3}H) is the radioactive isotope of hydrogen with a half-life of 12.32 years (Lucas & Unterweger, 2000). Natural tritium is mainly produced by the interaction of secondary particles from galactic cosmic rays (GCRs, high-energy charged particles) with nitrogen and oxygen atoms of the upper atmosphere at a global average rate of 0.345 atoms s^{-1} m^{-2} (Poluianov et al., 2020). After tritium has been produced, it oxidizes to a water molecule and enters the water cycle, hence becoming an excellent tracer in the atmospheric moisture and the hydrosphere. The tritium concentrations are expressed in tritium units (TU) where 1 TU refers to a ^{3}H/^{1}H isotope ratio of 10^{-18}. 1 TU equals to 0.119 Bq kg^{-1} activity concentration in case of water. Recent annual average tritium concentrations in precipitation vary between 2 and 20 TU (Terzer-Wassmuth et al., 2022), and are supposed to be very close to the natural level.

Previous studies have demonstrated that tritium in precipitation is modulated by the solar cycle (Borković & Krajcar Bronić, 2021; Fourré et al., 2018; László et al., 2020; Palcsu et al., 2018). The magnetic field of the Sun has a strong influence on the entire solar system, providing a huge shield against GCRs. Before reaching the Earth's atmosphere, GCRs are modulated by the magnetic shields of the Sun (heliomagnetic field or solar activity) and the Earth (geomagnetic field). As a result, the higher the solar activity or the geomagnetic shielding, the more

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GCR particles are deflected, and less tritium is produced. However, a significant amount of GCR can reach the planets, including Earth, influencing numerous processes like production of different nuclei, the so-called cosmogenic nuclei or isotopes (Lifton et al., 2005; Poluianov et al., 2016). The magnetic field of the Sun is not stable. It has an 11-year periodicity, which can be observed—among others—in the variation of production rates of cosmogenic isotopes in the atmosphere, such as tritium.

During the 1960s–1990s, elevated levels of anthropogenic tritium obscured the natural level and the influence of the solar cycle on tritium production. Nevertheless, the link between solar activity and cosmogenic tritium has been validated in Antarctic ice cores and precipitation records. Although recent emissions from the nuclear industry could still affect tritium levels in some regions, our study focuses on ice layers that preceded this contamination, offering a unique opportunity to study natural tritium variations. So, if pure natural variations of tritium are to be investigated, an archive has to be found which records the precipitation fallen earlier than the period of nuclear weapons tests.

Annually accumulated ice layers are perfect archives of precipitation in cold regions (Konrad et al., 2013; Shao et al., 2017). Early studies have shown that tritium in Arctic ice caps in Canada and Greenland were natural before 1952 and significantly increased after the 47 megatons (Mt) explosions of hydrogen bombs of the Castle series from March 1954 (Clark & Fritz, 2013; Kotzer et al., 2000). After the 1952–1954 period, tritium produced by thermonuclear weapon tests became the primary source, surpassing cosmic rays in abundance and significance (Begemann, 1959; Begemann & Libby, 1957). Annually layered ice deposits sedimented earlier than 1952, hence, are supposed to be free of artificial tritium, making them an ideal archive to preserve natural variability of cosmogenic tritium in precipitation.

The aim of this study is to quantify the natural variation of tritium in ice layers, extending the record to a period prior to the nuclear era, and assess the relationship between cosmogenic tritium and solar activity. Here we present, for the first time, a novel tritium ice core time series extending back to 1923, covering a crucial pre-bomb period. This data set provides an unprecedented opportunity to investigate natural tritium variability, free from the interference of anthropogenic sources, and offers insights into the modulation of cosmogenic tritium by solar activity. Since tritium has a short half-life, the tritium concentration of precipitation fallen earlier than 1954 has been reduced by a factor higher than 40. This reduction factor in tritium becomes 90 and 160 for the year 1940 and 1930, respectively. If recent tritium concentrations in central Europe are estimated to be around 10 TU, the expected tritium contents are between 0.060 and 0.250 TU. The precise analysis of such a low tritium concentration requires careful sample handling and extreme sensitive tritium analyses (refer to Figure S1 in Supporting Information S1 for comparison of analytical techniques). Here, with an achievement of 0.002 TU precision, we show that the variation in the tritium time series of the ice deposit in the area of Colle Gnifetti (Swiss-Italian Alps), can be linked to the solar cycle. Furthermore, we present the longest natural pre-bomb time series for tritium back to 1923.

2. Site Description

The accumulation zone of the high alpine cold glacier Colle Gnifetti (hereinafter: CG) (top of the Grenzgletscher) in the Swiss-Italian Alps, Europe, forms a small saddle (~0.2 km^2) between the two summits of the Monte Rosa massif (Signalkuppe and Zumsteinspitze) (Figure 1). Geodetic observations suggest that the saddle geometry is near steady state (Lüthi & Funk, 2000) and has remained almost unchanged over the last century (Wagenbach et al., 1996). The englacial temperature distribution is characterized by typically −14°C to −10°C at 20 m depth and −13°C to −12°C at bedrock (Hoelzle et al., 2011), which ensures preservation of the stratigraphy over the entire depth range. The drilling site was located in the middle of the saddle (45.9297°N, 7.8769°E, WGS84), where the ice deposition is vertical and has a depth of 72 m. The site is just a few meters apart from the KCC drilling site described by Bohleber et al. (2018) and Licciulli et al. (2020). The low snow accumulation rate of 0.22 m water equivalent/year is a result of seasonal net snow loss by wind erosion (Bohleber et al., 2013). Since snow consolidation is the most effective during summer (partial melting during the day, freezing at night), precipitation in colder seasons is more likely to be removed from the surface (Wagenbach et al., 1996). This effect has been confirmed by the lack of seasonality in the time series of ^{18}O (Bohleber et al., 2013). The percolation and re-freezing of meltwater enhances the preservation of the water isotope distribution in the firn section of the deposit (Van der Wel et al., 2011). The snow deposition at CG is continuous, since the partial melting and re-freezing of precipitation in the warm months make the ice deposit consolidated enough to resist wind-scouring

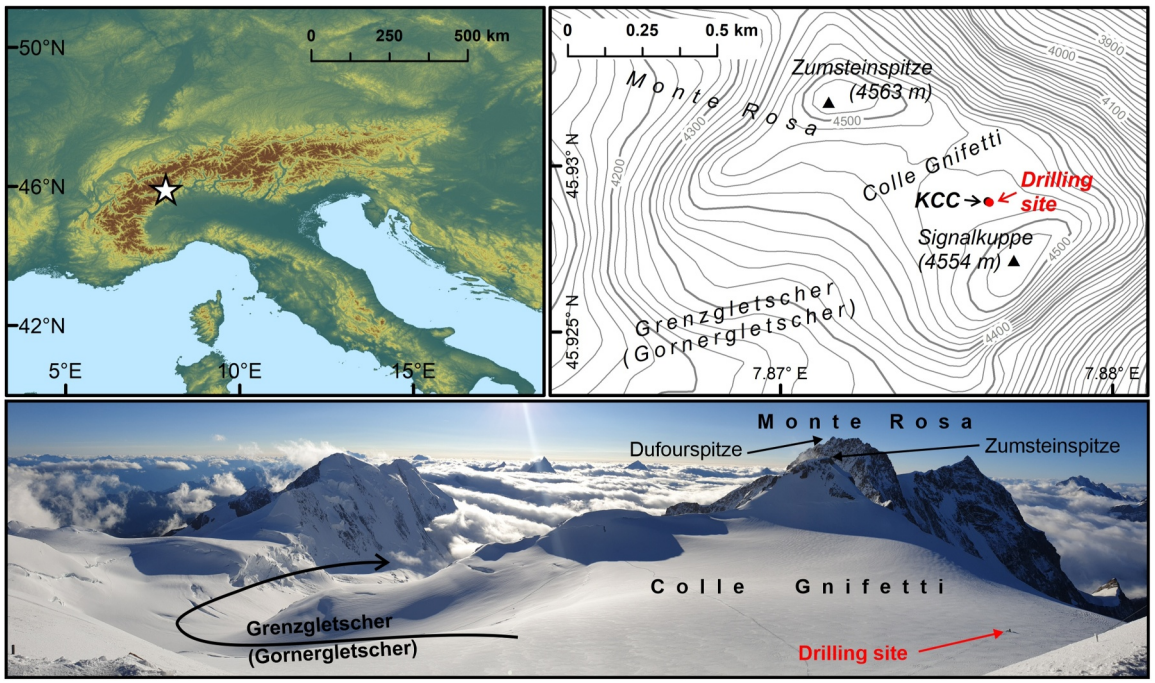

Figure 1. The location of the drilling site on Colle Gnifetti, located nearby the drilling site of KCC ice core (Bohleber et al., 2018).

during colder weather situations. The isotopic composition seems to be preserved in the ice deposition at CG, as shown in a comparison between two ice cores retrieved in 2015 and 2021, respectively (Huber et al., 2024; Sigl et al., 2018). The low accumulation rate makes it possible to retrieve ice cores dating hundreds of years back to the last millennium (Bohleber et al., 2018).

3. Methods

3.1. Ice Coring

After a 5 m shallow coring in 2019, two 34 m long ice cores were drilled with a hand auger (Kovacs Ice Drilling and Coring Equipment) in 2020 in order to obtain enough material of the older samples (below the bomb peak) necessary for sensitive low-level tritium measurements. The distance of the two cores was 1 m. The ice cores were retrieved in 50–70 cm long segments with a diameter of 9 cm, and immediately cut into 10–20 cm long pieces. Afterward, 1–2 mm of outer surface of the ice cylinders was removed, and then the individual samples were stored in plastic containers. The samples were not kept frozen, and melted during the shipment to the laboratory for further isotope analysis. The first core (A-core) was entirely sampled along the full depth, while the second core (B-core) was sampled only below the depth of 27.3 m. The subsampling strategy was based on the known accumulation rates (Bohleber et al., 2013, 2018). Subsamples of 10 cm length from A-core were selected in 1–2 m steps in the depth region of 0–14 m and 16–23 m. To search for the Chernobyl event as a time marker (April/May 1986), the sampling resolution was increased to 10 cm between 14 and 16 m. Similarly, to detect the bomb peak and the onset of the tritium upgrowth, 10 cm long ice samples were cut between depth of 23.0 and 27.3 m. Below the depth of 27.3 m, the whole length of both cores was sampled in 10–20 cm long segments. Altogether, 75 kg of ice was taken in 167 subsamples.

3.2. Isotope Measurements

Tritium, oxygen isotope ratio ($\delta^{18}O$) as well as ^{137}Cs were analyzed. Tritium samples of higher activities around the bomb peak were analyzed with liquid scintillation counting, while for sensitive tritium analyses, the ^{3}He-ingrowth method was used (Clarke et al., 1976). For the ^{3}He-ingrowth method, the melted ice samples were poured into stainless steel vessels of about 6 L equipped with all-metal valves. The headspace and the dissolved gases were completely removed by vacuum pumping. After degassing, the samples were stored for ^{3}He

production from tritium decay for more than a year. The absolute amount of tritiogenic helium was then determined by a Helix SFT helium mass spectrometer. The measurement was calibrated with well-known air aliquots (Palcsu et al., 2010; Papp et al., 2012). Background as well as tailing correction for HD^+ and H_3^+ peaks was also applied. The tritium concentration was then calculated from the tritiogenic ^{3}He, water amount, and storage time (Equation S1 in Supporting Information S1). Contrary to Palcsu et al. (2010), ^{4}He spiking to eliminate systematic errors was not done, since this method raises the detection limit, although it improves the accuracy and precision. Instead, internal standard samples of well-known tritium concentrations were analyzed and used to correct the systematic differences. With this method an accuracy of 0.002 TU or better can be achieved with long storage time (>1 year) and large sample size (>2 kg). A detailed description of the tritium measurement process can be found in Supporting Information S1. Ice samples around the bomb peak (depth at 23–24 m) were analyzed for ^{3}H by liquid scintillation counting (Janovics et al., 2014) without electrolytic enrichment.

The oxygen isotope composition was determined by off-axis integrated cavity output spectroscopy (Los Gatos Research). The results (δ^2H and δ^{18}O) are expressed in ‰ against the Vienna Standard Mean Ocean Water. The uncertainty of the oxygen isotope measurement is 0.15‰. ^{137}Cs activity concentrations were determined by a high-purity germanium detector in a low-level environment (Canberra-Packard BE50307915-30ULB thin-windowed planar HPGe detector).

3.3. Statistical Analysis

Paleoclimate records, including Colle Gnifetti, are typically irregularly spaced. This restricts the use of classical statistical analysis in the time and frequency domain. A frequent approach to bypass the uneven distribution in short and noisy time series is interpolation. Once applied, records become uniformly spaced which permits the application of methods like the Fast Fourier transform or the Continuous Wavelet Transform with their constructed Wavelet Coherence (WTC) and Cross Wavelet Transform (XWT) (Foster, 1996; Grinsted et al., 2004; Mudelsee, 2014). Nevertheless, several studies have shown that interpolation (if not properly carried out) might lead to enhancement of the record's low frequencies (i.e., red noise) and an overestimation of the persistence (i.e., memory) (Rehfeld et al., 2011; Schulz & Stattegger, 1997; Vaughan et al., 2015). Therefore, for the bivariate wavelet analysis (WTC and XWT) of the CG natural tritium record (1923–1955) with sunspots and neutron flux data sets (SILSO, World Data Center, 2024; Usoskin et al., 2002), we used the Weighted wavelet Z-transform (WWZ). This method exposes their common coherence, power, phase, and captures their intrinsic non-stationarity inherent in the solar and atmospheric processes (Foster, 1996; Grinsted et al., 2004). Pre-processing was carried out before the wavelet computation by linear detrending. Mathematically, it involves fitting a linear model to the data and subtracting this trend line from the original data. By removing the trend from the time series data, the cyclical components are better isolated. The tritium and neutron flux time series were linearly detrended, while the raw sunspot data was utilized for the computation. The reason for this is lack of a clear trend during this period for the sunspot data, and a linear detrending only creates a non-existent trend. In case of the univariate spectral analysis, the analysis focused on the Power Spectrum, which shows how the power of a signal is distributed across different frequencies by detection of significant peaks (dominant frequencies).

The application of several spectral methods on a time series is a well established approach for obtaining reliable information. Linear interpolation was utilized to compare the obtained Power Spectral Density (PSD) from evenly and unevenly spaced spectral methods in the full tritium data set (1923–2020), encompassing the deep and shallow cores. The Periodogram, Welch and Multi-Taper Method (MTM) are methods demanding evenly spaced data (i.e., time series interpolation). The first one is based on the Fourier transform, whereas the Welch's periodogram is a variant employing the Welch's method of overlapping segments. Finally the MTM employs small set of tapers to decrease the variance of spectral estimates (Khider et al., 2023, 2025). On the contrary, the calculation of the PSD with the raw tritium data (irregularly spaced) made use of the Lomb-Scargle and WWZ methods (Foster, 1996; VanderPlas, 2018). Unfortunately, due to the shortness of the natural tritium time series (1923–1955, $n = 28$), the calculated PSD gave spectrums tightly close to the AR(1) 90%, 95%, and 99% thresholds (with 2,000 simulated surrogates), making it difficult to determine with clarity the significance of the peaks, hence are not included in this paper. Spectral and wavelet analyses were based on the conceptual framework described by Khider et al. (2025) employing the package pyleoclim in a Python version 3.11.8 (see Jupyter notebook in Supporting Information S1).

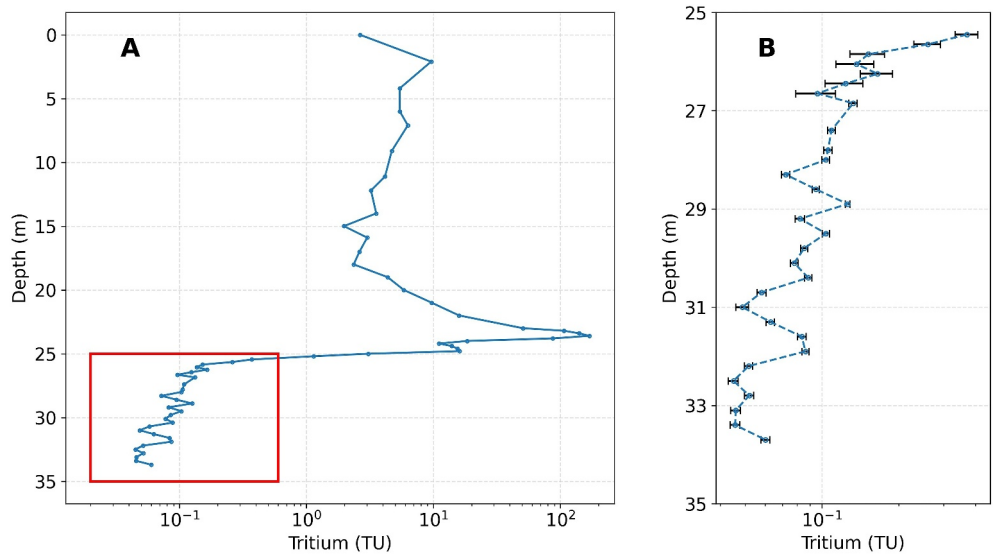

Figure 2. (a) The tritium profile along the entire core showing the bomb peak at 23.65 m. (b) The tritium variation in the lower section (between 25.45 and 33.85 m).

4. Results

4.1. δ^{18}O Profile of the Lower Section of the Two Cores

To perform extremely precise tritium analyses of old ice samples, none of the subsamples of both cores provided enough material. Hence, the subsamples of the lower section (26.8–34.0 m) of the two ice cores were merged. Two and three subsamples of cores A and B from the same depth were merged just prior to tritium analysis. Although the depth of the ice cores was carefully registered during drilling, there was still some uncertainty regarding whether the two cores represent the exact same ice sequence. Subtle variations in snow accumulation, wind patterns, or microtopography could result in differing accumulation rates even within a 1-m distance. We assumed that the samples from the same depth represented the same accumulation period, but this assumption relied on the ice having accumulated horizontally, without any re-accumulation or irregular layering. To make sure that the two cores provided the same ice layering, the variation of hydrogen and oxygen isotope ratios were compared. The Figure S2 in Supporting Information S1 depicts the δ^{2}H and δ^{18}O values along the deep section of the two cores. The stable isotope composition shows that the two parallel cores have similar values, which indicates that the same depth provides the same ice deposit in the two cores. Hence, subsamples from the same depth can be handled as identical, even if some post depositional processes might have slightly affected the stable isotope composition (e.g., *d*-excess; refer to Figure S2 in Supporting Information S1), and therefore, were merged for low level tritium analysis.

4.2. Tritium Profile

The tritium depth profile is shown in Figure 2. The tritium concentrations refer to the date of sampling (1 August 2020). Although, an estimation to the age distribution has been known from Bohleber et al. (2018) and Licciulli et al. (2020), we first looked for time markers to make the time scale more precise. An anchor point of the age profile is the so-called bomb peak that occurred in summer 1963. Since Chernobyl ^{137}Cs could not be detected (see Discussion), the age-depth relation is based only on the detected bomb peak and the previous time scale. Figure 2a clearly shows that the bomb peak can be found at a depth of 23.6–23.7 m. Sensitive tritium analyses have been performed for the lower section of the ice cores (depth between 26.8 and 34.0). The ice samples below 26 m depth do not seem to be affected by bomb tritium depicting values below 0.20 TU at the time of sampling (Figure 2b). Its decrease with depth is probably due to radioactive decay. This indicates that these layers were found to be deposited before the thermonuclear bomb tests.

Note that the lowest tritium value is 0.045 ± 0.002 TU. These values are consistent with expectations for natural cosmogenic tritium production in the atmosphere during this period. Above 26.0 m, tritium values increase with

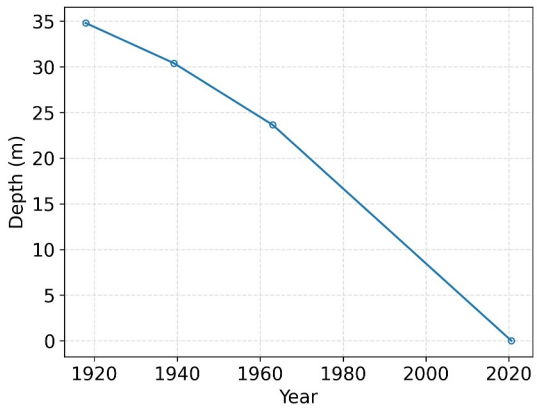

Figure 3. The age-depth profile of the ice cores adopted from Bohleber et al. (2018) modified using the well-known depth of the bomb peak.

decreasing depth, indicating that anthropogenic tritium starts playing a significant role. The tritium increase in this section correlates with known periods of nuclear weapons testing, which significantly elevated atmospheric tritium levels in the mid-20th century. While the bomb peak is a clear marker, additional sources of anthropogenic tritium, such as emissions from nuclear power plants and industrial activities, may also contribute to the elevated tritium levels in this section of the core.

5. Discussion

5.1. Age Profile of the Ice Cores

To convert the depth into calendar years, we have to establish an age-depth profile of the ice deposit. Particular events were planned to be used as fixed anchor points in time, such as the tritium bomb peak (summer 1963) and the Chernobyl accident (April/May 1986). Although there is a seasonal summer bias in snow accumulation at Colle Gnifetti preserving mainly the warm season deposit, significant but a low amount of radioactive fallout was found in July 1986 that could be attributed to the Chernobyl fallout during early May in 1986 (Haeberli et al., 1988). The radioactive signal of 35 Bq kg^{-1} at 20 cm depth was built up mainly by the isotopes of ^{137}Cs, ^{103}Ru, and ^{106}Ru, with a ratio of 1:1.3:0.3 (McAulay & Moran, 1992). Due to radioactive decay and short half-lives of ^{103}Ru and ^{106}Ru (39.25 and 371.8 days, respectively), only ^{137}Cs (half-life: 30.08 years) contributes to the radioactive signal of the Chernobyl event by our time of sampling. The remaining activity concentration was calculated to be ∼6 Bq kg^{-1}. However, the time marker of the Chernobyl event could not be detected in our record. Since the expected depth was estimated by a linear age-depth function between 1963 and 2020, and although the sampling resolution was high enough to observe increased radioactivity, the depth range of high sampling resolution might not have crossed the year of 1986.

Bohleber et al. (2018) determined the chronology of the KCC core (Figure 1) using annual layer counting of impurities such as NH_4^+, NO_3^-, Na^+, Ca^{2+}, and ^{44}Ca, and meltwater conductivity. They combined impurity profiles with absolute age constraints from radiocarbon analysis. This age profile was utilized for our two ice cores using the known depth of the bomb peak (23.6–23.7 m). The constructed age-depth profile is presented in Figure 3. The age-depth relationship for layers deeper than the bomb peak is based on previously established models and the general accumulation rate estimated for the site. However, we acknowledge that this method has inherent uncertainties. While the bomb peak provides a well-established reference point, age estimations for the deeper layers rely on assumptions about the uniformity of deposition and accumulation, both of which may vary over time due to environmental factors.

The annual layer thickness is 41, 28, and 21 cm in the depth sections of 0–23.65 m (years 1963–2020), 23.65–30.4 m (years 1939–1963), and 30.4–34.8 m (years 1918–1939), respectively. This function was used to convert the depth of each of the individual subsamples to calendar age, and then to calculate the initial tritium at the time of deposition. Although, it is assumed that the annual layer thickness is a linear function of depth in each depth section, the thickness of each annual layer might vary, but the layer sequence must be consecutive. Thus, despite the inherent uncertainty of 1–2 years associated with each estimated calendar year, the time series remains continuous and free of time reversal, as the melting and refreezing processes prevent any re-accumulation.

5.2. Reconstruction of the Tritium Profile at the Time of Deposition

The age-depth profile served a dual purpose: first, to convert the depth to the calendar age of deposition, and second, to correct tritium data for radioactive decay since the time of deposition. Figure 4 shows the time series of the tritium values of the ice layers corrected to the date of deposition. Around the bomb peak, the ice tritium is compared to the precipitation tritium time series of Ottawa and Vienna averaged only for the summer months (June, July, and August, noted hereafter as JJA) (Figures 4a and 4b) (IAEA, 2024). As the year 1963 was used for the age-depth profile as an anchor point, the highest tritium values are coeval for the three time series. The shape of the curve of Colle Gnifetti (i.e., width) compares well to that of the Ottawa and Vienna records, indicating that the age-depth profile is robust around the year 1963. In the years 1960 and 1961, Ottawa had a local minimum, so did CG in 1961 (Figures 4a and 4b). From the year 1923–1957, the tritium varied between 4.4 and 14.4 TU

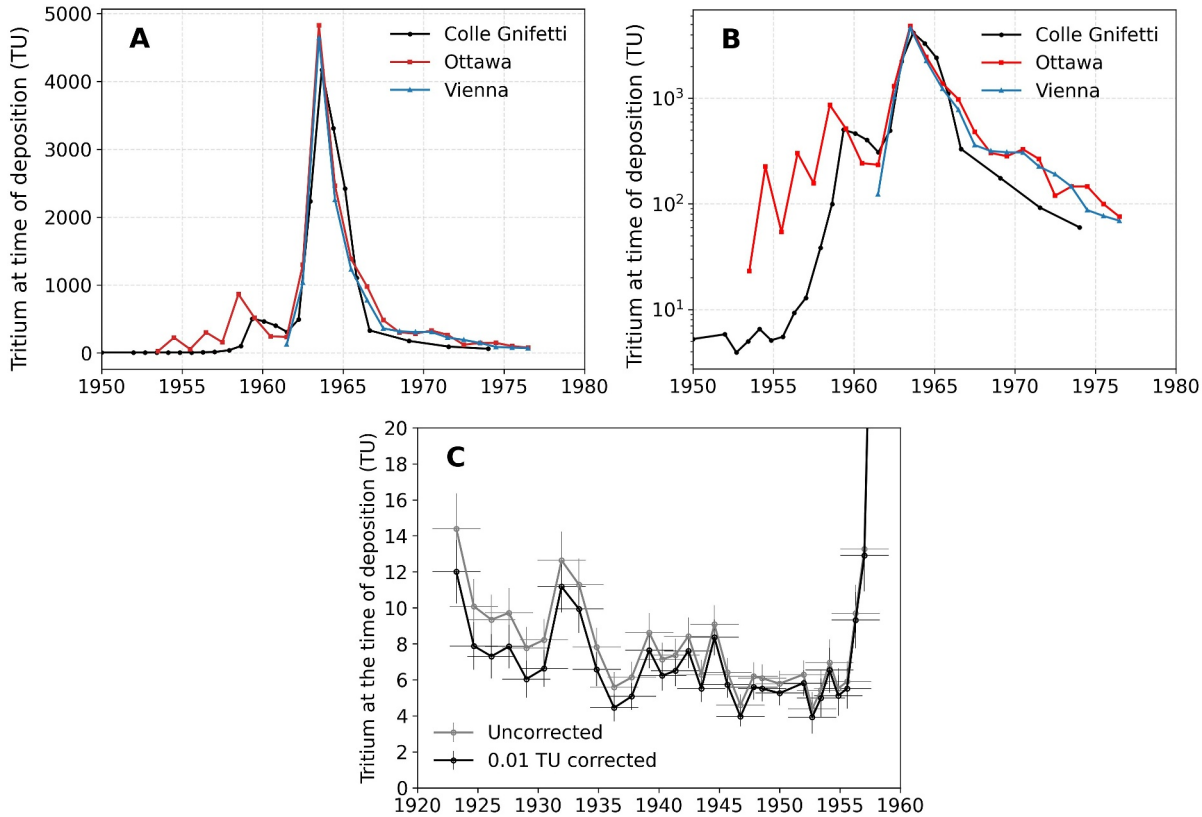

Figure 4. The time series of annual tritium as a function of calendar years of deposition. The Colle Gnifetti tritium profile is compared to that of Ottawa and Vienna ((a, b) linear and logarithmic scale). The uncorrected and corrected tritium profile of the older sections are shown in panel (c).

(Figure 4c). Although there are some local maxima and minima in the time series, a general decreasing trend can be visible from 1926 to 1955. After 1956, a sudden increase appears: in the beginning and in the middle of 1958, tritium is reaching the values of 39 and 100 TU, respectively. These layers must have been contaminated with artificial tritium, mainly due to the thermonuclear weapon tests. We assume that ice layers accumulated earlier than 1958 (according to the time scale of Figure 4) are not affected by anthropogenic tritium. Therefore, we infer that these layers incorporate solely natural tritium. Two questions arise in this regard: (a) how precise is the time scale? (b) what is the reason for the tritium variations between 1923 and 1955? Since the age-depth profile has been composed by combinations of linear functions, identical annual ice thicknesses were assumed between 1918 and 1939, and between 1939 and 1963. However, the annual deposition might have been changing during the period of interest. This means that the curve in Figure 4 has a flexibility to move elastically in a horizontal direction, while no time reversal happens. We can verify the accuracy of the age-depth profile when identifying the date of the rapid increase of tritium due to the first thermonuclear explosions. Although the first hydrogen bomb test explosion of 10 Mt was executed in the Pacific Ocean (Enewetak Atoll) in November 1952, it resulted in an increase of tritium of only 70 TU in the Chicago rain located 11,000 km from the test site (Begemann, 1959; Begemann & Libby, 1957). Operation Castle executed six test detonations (overall 47 Mt) at Bikini Atoll between March and May 1954. The release of tritium load of Operation Castle into the atmosphere increased the tritium concentration to 450 TU in Chicago and to 2,500 TU in Ottawa (Begemann & Libby, 1957). Note that the Chicago and Ottawa values for Begemann and Libby (1957) are event-based and their comparability with the later regular monthly sampling is limited, while Figure 4 shows annual averages of the monthly values. When did the tritium in precipitation rise above the natural level in Europe? Roether (1967) declared that the first distinguishable influence of bomb tritium showed up in German wines in 1953. Indeed, the first sharp increase of tritium recorded in the Fiescherhorn glacier (70 km to the north-northeast of Colle Gnifetti, 3,900 m a.s.l., Figure S3 in Supporting Information S1) can be seen in the year 1954 (Schotterer et al., 1998; Schwikowski et al., 1999). However, high tritium levels appear mainly in spring months with the summer months being less affected. In our record, the tritium concentration suddenly increased from 1956 to 1957. Comparing the shape of our tritium record to the

Ottawa record between 1956 and 1961, a 1-year offset can be observed if we assume that CG was not contaminated in the years before 1957. Although, the age scale might be shifted by 1 year, we arbitrary employ an age uncertainty of 2 years to propagate the uncertainty for the tritium concentrations. Hence, besides the analytical uncertainties of the tritium analyses, the uncertainty of the final value of tritium at the time of deposition includes the age error also (Figure 4c).

5.3. Natural Level of Tritium

After evaluating the age of the tritium time series, we conclude that the ice samples taken before 1956 were not affected by the thermonuclear weapon tests. The oldest ice from 1923 had a tritium concentration of 14.4 ± 2.0 TU, while the lowest tritium concentration value (4.59 ± 0.60 TU) was from 1946. Although these values match that of recent precipitation, an additional correction is needed for two reasons: (a) cosmogenic production of ^{3}H in ice as well as cosmogenic production of ^{3}He in the water during tritium analysis, (b) contamination of the ice core with ambient moisture during sampling. The production rate of cosmogenic tritium in ice layers has been estimated to be less than 1,500 atoms g^{-1} $year^{-1}$ at the latitude and altitude of Colle Gnifetti (Lal et al., 1987). This value corresponds to 0.023 TU, which is far negligible compared to the tritium concentration of fresh snow (5–10 TU). Additionally, the accumulated snow shields the buried layers against secondary neutrons that may produce in situ tritium, hence the production rate of in situ tritium is decreasing with depth. An additional source of systematic errors might be a potential production of ^{3}He in the water samples during the tritium analysis by the ^{3}He ingrowth method (Brown et al., 2000). When tritium is analyzed, the water is degassed, and then stored in a stainless-steel vessel in the laboratory. During the storage, cosmic ray induced secondary particles may produce ^{3}He in the water, in addition to the tritium decay. The ^{3}He from the tritium decay cannot be distinguished from the ^{3}He of in situ production. The production rate depends significantly on the elevation. At the elevation of our laboratory (130 m a.s.l.), the production of ^{3}He does not cause an apparent increase of the measured tritium concentration higher than 0.002 TU (Brown et al., 2000).

The most significant contamination may occur during sampling. As can be seen, ice samples below the depth of 26 m (Figure 2) have tritium concentrations lower than 0.2 TU. At this depth, the ice temperature decreases from −14°C to −10°C, which is below the dew point of the ambient air at the surface during sampling. Hence, atmospheric moisture might condense at the cold surface of the ice cores immediately after pulling them out. Although, the cutting of the ice core into subsamples and cleaning the outer ice surface did not last more than 10 min, contamination due to condensation cannot be excluded. In a laboratory experiment, where the ice and air temperatures were −18°C and +5°C, respectively, we determined that an ice subsample of cylindrical shape (diameter: 9 cm, length 10 cm, and weight: ∼570 g) could adsorb up to 0.3 g of atmospheric water vapor in 10 min. In the area of the glacier, summer precipitation, and hence atmospheric moisture can achieve 15 TU of tritium concentration (Affolter et al., 2020). Taking these (likely overestimated) values into account, the maximum contribution of the ambient moisture to the tritium concentrations of the ice subsamples does not exceed 0.008 TU. If we combine the two sources of contamination (i.e., in situ production of ^{3}He during tritium analysis, and condensation of the ambient vapor during sampling), we can conclude that the measured tritium concentration values have to be corrected by up to 0.010 TU. Note that this correction has been made by overestimating the potential effects of disturbance. It is an additive correction, namely it has to be subtracted from the measured tritium values, thus the older the ice the more significant the correction is. For example, the tritium concentration of the oldest ice became 12.0 ± 1.8 from 14.4 ± 2.0 TU. Moreover, this correction is negligible for the ice layers after 1958, when the bomb tritium is present. In Figure 4c, the black curve depicts the corrected tritium time series, which will be used for further evaluation henceforth. As our tritium record extends back to 1923, to the best of our knowledge, it is the longest pre-bomb tritium record.

One of the main questions is whether the deeper section of the Colle Gnifetti ice core preserves solely tritium of natural origin. Therefore, the tritium profile is compared to other proxies for the tritium concentration of precipitation: European wine samples and nearby ice layers. Figure 5 shows a tritium profile of an ice core retrieved at the Fiescherhorn glacier (hereinafter: FH). The stable isotope record of the FH core indicates that the seasonal variation of the stored precipitation is not remarkably altered by post-depositional processes (Schotterer et al., 1998). Although, the FH tritium profile is monthly resolved, the shape is somewhat different than that of CG, mainly before 1959. Four reasons can be considered to explain the dissimilarity: (a) the age profile of CG record is not absolutely correct, (b) FH tritium record is affected by the bomb tritium, while CG record is not (or is less affected), (c) FH record is contaminated by industrial tritium (while CG signal is not or is less affected), and

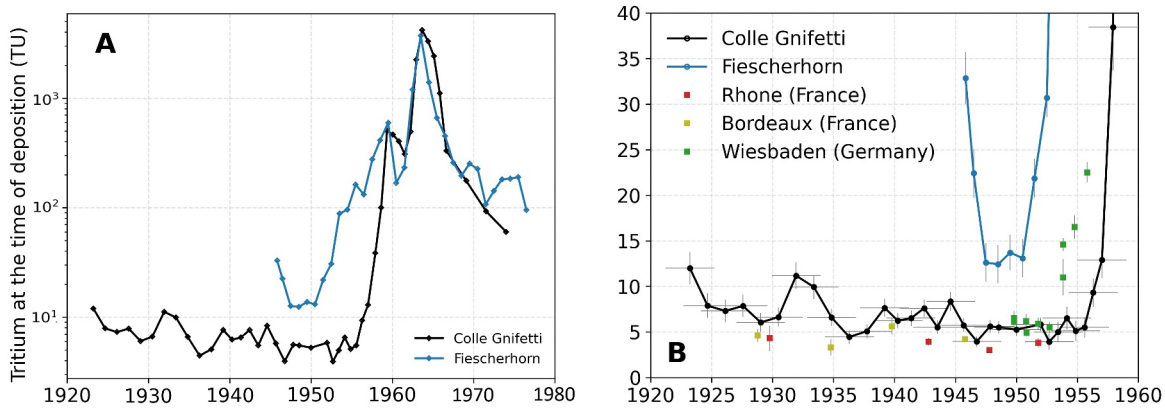

Figure 5. Comparison of the Colle Gnifetti tritium profile to the (a) Fiescherhorn site and (b) European wine samples.

(d) FH record still suffers post-depositional processes. We can accept that the FH profile is well dated using—among other tracers—the seasonality of oxygen and hydrogen stable isotope composition as well as tritium. If the age profile of CG has a large shift in the 1950s, or there is a hiatus in the snow accumulation, that might explain that FH has significantly more tritium than CG does. In the previous section, we argued that the age of the ice layers had been interpreted correctly, and the uncertainty of the age can be considered to be up to 2 years. The shape of the two tritium records in the early 1960s is very similar, so it seems the age profile of CG is appropriate enough in this period, the difference appears only in the 1950s. Another explanation can be that bomb tritium affects only the FH plateau, while CG is not (or less) affected. The most dominant moisture source of the region is the Atlantic, the prevailing wind direction is NW for both sites, hence in principle, although less likely, it is possible that bomb tritium has fallen only at FH. On the other hand, a large difference in tritium can be seen even before 1952, when weapon tritium can be absolutely excluded. The average tritium concentrations of FH and CG for the period from 1945 to 1952 are 20.0 and 5.1 TU, respectively. This difference can be hardly explained by natural processes. Instead, tritium emitted from the Swiss luminous compound industry might have affected the FH site, which is about 64 km from Bern. Recent precipitation of Bern is about 36 TU, while rural sites have an average tritium concentration of 10 TU, so Bern is certainly contaminated with local tritium (Figure S5 in Supporting Information S1). Krejci and Zeller (1978) states that the production of tritium luminous compounds started in 1962, thus no evidence exists that the luminous industry is responsible for the elevated tritium at FH in the 1950s. Since ^{137}Cs from the weapon test before 1952 is clearly measurable above the detection limit in the FH ice, the influence of nuclear weapons cannot be excluded completely (Schotterer et al., 1998).

In order to argue that Colle Gnifetti has only natural tritium, the tritium profile is therefore compared to European wine samples. Since the growing season of grape is mainly late summer/early autumn, the direct comparison to CG, where only summer (JJA) precipitation accumulates, has to be done with the assumption that the tritium concentration of wine may be lower than that of the summer precipitation mean. French and German wines from 1928 to 1952 have tritium concentrations of 3.0–6.1 TU (Roether, 1967), slightly lower than ice layers from the same period (Figure 5b). The average tritium concentrations of Rhone (France), Bordeaux (France) and Wiesbaden (Germany) wines are 3.75, 4.43, and 5.85 TU, respectively, showing that areas closer to the coastal regions (Rhone valley and Bordeaux, France) receive precipitation with lower tritium than inland territories do (Wiesbaden area, Germany). Additionally, the average tritium concentration of CG from 1948 to 1952 is 5.13 ± 0.83 TU, which fits very well to the average tritium of the German wines (5.85 ± 0.57 TU) in the same period. The increase of tritium can also be seen in German wines after 1954, indicating that the wine region in middle Germany received artificial tritium from the early thermonuclear tests.

These comparisons support the conclusion that the tritium in the CG ice layers accumulated before 1954 possesses solely natural tritium. How does the natural tritium level obtained from the ice layers of 1923–1954 compare to that of recent precipitation? Since the early 2000s, the tritium concentration in precipitation is still decreasing, but the trend is much lower than in the earlier years (Anh et al., 2018; Borković & Krajcar Bronić, 2021; Chae & Kim, 2019; Duliński et al., 2019; Duliu et al., 2018; Gusyev et al., 2016; Harms et al., 2016; Palcsu et al., 2018; Schmidt et al., 2020; Terzer-Wassmuth et al., 2022; van Rooyen et al., 2021; Vreča et al., 2024). For instance, the

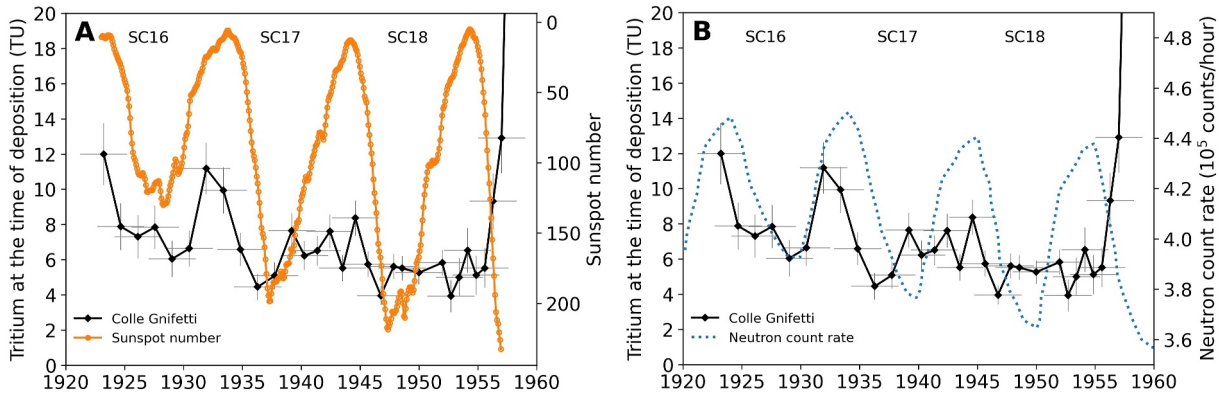

Figure 6. Decreasing and increasing trends in the CG tritium profile over the solar cycles 16–18. (a) The alternation of tritium negatively correlates with the sunspots (note that the scale of the sunspot number is reversed), (b) while it positively correlates with the neutron count rate reconstructed by Usoskin et al. (2002).

Vienna tritium record shows that between 2000 and 2009 (part of solar cycle 23, hereinafter SC23) the average tritium concentration was 10.5 TU, while in SC24 (between the years 2009 and 2019) it was significantly lower, 9.1 TU (Figure S5 in Supporting Information S1). For the summer months, it is 12.9 and 11.7 TU for SC23 and SC24, respectively. Additionally, the CG tritium record between 1985 and 2010 fits well to the tritium time series of the closest monitoring stations (Sion, Grimsel, and Guttannen) (Figure S4a in Supporting Information S1), so CG ice represents very well the local precipitation. However, we cannot refer that the recent level is completely natural, since the presence of artificial tritium cannot be excluded. There is a decreasing trend in tritium at CG from 1923 to 1955, the average tritium concentration is 6.6 ± 2.0 TU, which is remarkably lower than the average tritium content of summer precipitation at nearby monitoring stations between 2000 and 2009, having average summer tritium concentrations of 12.4 ± 1.8 TU (Figure S4b in Supporting Information S1). In south-eastern Switzerland, the tritium concentrations of precipitation was even lower. In Locarno and Pontresina, the summer averages are 9.4 and 10.8 TU (Figure S5 in Supporting Information S1). Since observed data are only available until 2010, the average values relevant to SC24 can be estimated by the decrease of tritium in the Vienna precipitation. After the correction, the summer average for Sion-Grimsel-Guttannen during SC24 is 11.2 TU, while for Locarno and Pontresina it is 8.5 and 9.8 TU. All these values are definitely higher than that of the pre-bomb tritium concentration of Colle Gnifetti.

5.4. Tritium and the Solar Cycle

The main aim of our study is to evaluate how the solar cycle is reflected in the cosmogenic tritium profile of the ice deposit at Colle Gnifetti. In the previous sections we have demonstrated that the ice layers below the depth of 25.9 m (year 1955) contains tritium of solely natural origin, hence the variation of tritium must be attributed only to natural processes. Figure 6 presents the tritium record from the CG glacier showing a quasi-periodic variation. Indeed, from 1923 to 1929, the tritium values decrease from 12.0 to 6.0 TU, and then rise to 11.2 TU by 1932. The periodicity repeats again with its lowest value in 1936 (4.5 TU) and a subsequent peak before 1940 (7.0 TU). From 1940, the periodicity is less clear with small peaks and valleys in a short interval (~5 years) until 1945. The lowest tritium concentration is achieved in 1946 and 1952 (~4.0 TU). Finally, toward the end of the record, from 1948 to 1955, no clear cycle in tritium can be observed. The visual comparison of the tritium record with the sunspot number (SILSO, World Data Center, 2024) (Figure 6a) shows that the lowest tritium concentrations appear at sunspot maxima over the SC16 to SC18, whereas high tritium values can be seen during sunspot minima. Similarly, the tritium record is well correlated with the secondary neutrons produced by the GCRs (Figure 6b) (Lifton et al., 2005). Overall, the strong association between the CG tritium with the sunspots and secondary neutrons strongly suggests an interconnection between the tritium and the solar cycle.

The sunspot number, however, does not have a direct influence on the tritium variation. The main indicator to the tritium production rate, and hence to the tritium amount in precipitation, is the count rate of GCR secondary particles detected in the atmosphere, mainly neutrons and protons (Poluianov et al., 2016). Since regular neutron monitoring started in America and Europe in the early 50s and 60s, respectively, no real neutron data is available for the period of the pre-bomb time series of our ice core. After reconstruction of the solar magnetic flux (Solanki

et al., 2000), Usoskin et al. (2002) reconstructed the neutron count rate for the last four centuries. Figure 6b illustrates the relation between the tritium and the reconstructed neutron count rates for the pre-bomb time period. As a general trend, the neutron count rate decreases by a rate of 0.25% yr^{-1}, while the tritium decreases by 1.8% yr^{-1} compared to the average. Although, the production rate has a larger variability than that of the neutron count rate of a certain energy range (Poluianov et al., 2016), the decreasing trend of tritium seems to be larger than expected. Nevertheless, the alternation of both tritium and neutron indicates a positive correlation. Apart from a time lag of 1–2 years, the tritium variation correlates remarkably with the neutron count rate, indicating a direct link between the solar cycle and the tritium content of the ice. The decreasing trend in the tritium time series is following the decreasing trend in the reconstructed neutron count rate.

Another possible explanation for the CG tritium variability is the climatic influence of the North Atlantic Oscillation (NAO). Indeed, Scherrer et al. (2004) have found differences regarding the influence of the NAO and snow day variability in the northern and southern parts of Switzerland. We evaluated the effect of the NAO in the CG tritium record employing back trajectories and spectral analysis comparing with two NAO indexes (See Figures S6 and S7 in Supporting Information S1). We found that, in case of moisture uptake, there is an Atlantic signal imprinted along the year but reduced during summertime (June–August), consistent with the literature indicating NAO major role during winter time (Figure S6 in Supporting Information S1). During summer, the NAO signal weakens significantly as the pressure systems (Icelandic Low and Azores High) diminish in intensity leading to weaker westerly winds (Affolter et al., 2020; Beniston & Jungo, 2002; Scherrer & Appenzeller, 2006). Our analysis (Figures S6a and S6b in Supporting Information S1) further supports this idea, as uptake of moisture comes chiefly from a reduced and more localized Atlantic area (contrary to the extensive flux during the rest of the seasons), along with neighboring continental and Mediterranean contributions. This suggests that precipitation during summer is more influenced by local processes rather than large-scale atmospheric circulation at the CG site. The spectral analysis with the mean annual NAO indexes suggests (at least for the natural period 1923–1955) a more important 5–10 years influence than the interannual variability (not significant $n = 2{,}000$ surrogates) (Figure S7 in Supporting Information S1). Similar periodicity has been observed by Massei et al. (2007), which leaves the 11-year period signal from the solar cycle free.

Further statistical analysis in the frequency domain reveals that this link with the solar cycle is indeed robust. Figure 7 shows the wavelet analysis between the Colle Gnifetti tritium record and the sunspot number. Both records are visually compared as in Figure 6a with the only difference that the tritium time series has been linearly detrended and focused on the natural period 1923–1955 (Figure S8 in Supporting Information S1). The WTC (Figure 7b) indicates a strong relationship at the 10–11 year period for the tritium and sunspot data. Similarly, XWT analysis (Figure 7c) shows a high common power for both records in the 9–11 year period region. Here, white contour lines indicate significant coherence ($\alpha < 0.05$), whereas arrows pointing left indicate an anti-phase angle. It is worth noting the clear anti-phase across all timescales observed in the WTC and XWT, whose WTC mean phase angle over the significant regions at 11-year is −157.42°, confirming that the tritium and the sunspots are in anti-phase. In a similar manner, wavelet analysis between the tritium record and neutron flux depicts a strong signal at the 10–11-year XWT periodicity band (Figure S9 in Supporting Information S1). What stands out in this figure is the direction of the arrows (up-right) in high coherence regions. The WTC phase angle for the 11-year period is 63° indicative of the tritium record leading to the neutron flux data but with a phase difference less than 90°, which is an artifact, as tritium cannot be produced until there is neutron population. Furthermore, there is a broader periodicity of 10–17 years in the WTC (warmer orange colors). These apparent counterintuitive results in the WTC might be due to the use of the modeled neutron flux data set for the natural period, with an intrinsic time uncertainty and short period, that inflates the coherence toward higher periods.

Nevertheless, despite this limitation, their correlation at 11-year is not overridden, but less localized. Turning now to the spectral analysis of the individual tritium record, the Figure S10 in Supporting Information S1 shows the PSD evaluated using the evenly and unevenly spaced tritium. The power spectrum in Figure S10a in Supporting Information S1 shows a prominent peak at a frequency of approximately 0.09 cycles per year (~11 years), along with other small peaks around 0.15, 0.25 cycles per year (~4–7 years). Except for the PSB analysis using WWZ method, the PSD graphs (Figures S10b–S10d in Supporting Information S1) also indicate 5 to 11-year period peaks, which are significant at 90%, 95%, and 99% threshold using an AR(1) red noise model benchmark and 2,000 simulated surrogates of the tritium record. In summary, the wavelet and spectral results indicate that the 11-year cycle is statistically significant and is present in all three data sets: sunspots, neutron flux and tritium. The *p*-values are all below the threshold of 0.05, strongly suggesting that the 11-year periodicity observed in these time

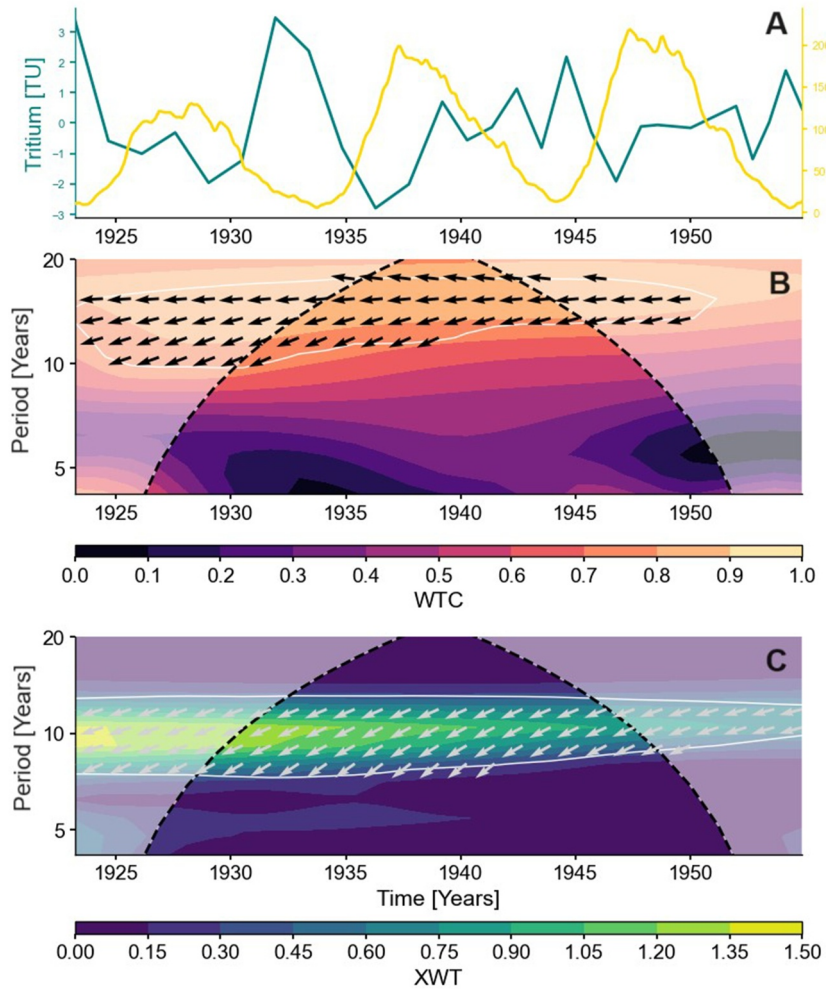

Figure 7. Wavelet analysis of the Colle Gnifetti glacier tritium record compared with the sunspot number for the period 1923–1955. (a) Linear detrended tritium and sunspots time series. (b) Wavelet coherence (WTC) and (c) Cross Wavelet Transform (XWT) of the same records.

series is not due to random chance but is a genuine feature. The significance of these cycles highlights the presence of underlying periodic phenomena that could be linked to solar activity, particularly the well-known 11-year solar cycle.

5.5. Comparison of Colle Gnifetti Tritium Record to Model Results

Poluianov et al. (2020) have provided a simulation called CRAC:3H that calculates tritium production rates taking into account continental and latitudinal effects due to Earth Magnetic field as well as temporal changes in solar activity. As the mean residence time of water in the troposphere is just 9 days (van der Ent & Tuinenburg, 2017), much shorter than in the stratosphere (Cauquoin et al., 2016; Ehhalt et al., 2002), variation of the tropospheric production rate of tritium might be directly reflected in the tritium concentration of precipitation. Cauquoin et al. (2024) showed that significant changes in tritium production occur not only in the stratosphere but also in the mid-to high-troposphere, which could explain the rapid responses of tritium in precipitation. However, solar cycle-related variations are also present in their modeled stratospheric tritiated water vapor. In addition to production effects, tritiated water is influenced by internal climate variations. Cauquoin et al. (2024) provided an isotope-enabled atmospheric general circulation model MIROC5-iso by considering cosmogenic tritium production changes due to solar activity variations. Using the global tritium production from CRAC_3H_solar simulation, the MIROC5-iso shows that tritium in precipitation at different locations is not similarly influenced by changes in tritium production due to solar activity variations. Figure 8a shows the correlation of the linear

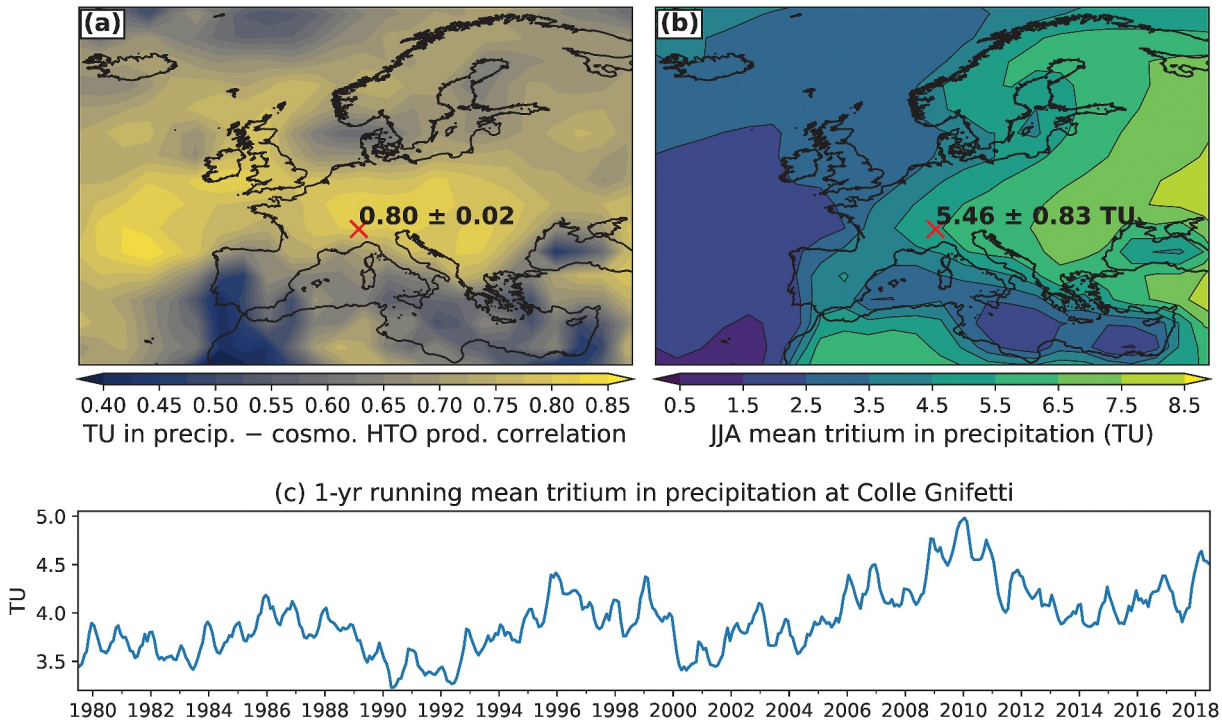

Figure 8. Tritium in precipitation in Europe according to CRAC_3H_solar MIROC5-iso simulation for the period of 1979–2018 (Cauquoin et al., 2024). (a) Correlation coefficients of the linear regressions for Europe between 1-year running mean tritium in precipitation and global tritium production, (b) spatial distribution of JJA tritium in precipitation, and (c) modeled 1-year running mean tritium in precipitation variations at CG are shown.

regression between 1-year running mean tritium in precipitation and global tritium production from the CRAC_3H_solar simulation over Europe. The calculation predicts that the influence of the solar cycle to the tritium variation in precipitation is visible in middle Europe ($r = 0.80 \pm 0.02$ at CG), and it is not overwhelmed by other atmospheric processes (Cauquoin et al., 2024). Indeed, our tritium record and modeled tritium variations (Figure 8c) exhibit a significant 11-year cycle that aligns with solar activity, suggesting a potential influence of solar activity on tritium levels. The wavelet analysis reveals a significant relationship between sun spots, neutron count rate and tritium concentrations. The significant correlation values suggest that solar activity influences tritium levels, providing valuable insights into the interaction between solar cycles and environmental variables. Figure 8b shows the spatial distribution of modeled mean tritium in summer (JJA) precipitation for the period of 1979–2018. The modeled tritium in JJA precipitation value at nearest grid cell of CG site is equal to 5.46 ± 0.83 TU (Figures 8b and 8c), which agrees very well, within uncertainty, to the average tritium level obtained from the CG ice (6.6 ± 2.0 TU).

6. Conclusion and Outlook

A complete tritium time series is presented, covering the period of 1923–2020. The study provides the longest pre-bomb tritium record so far. Evaluation of low-level tritium concentrations of ice layers retrieved in Colle Gnifetti (Swiss-Italian Alps) provides a good estimation to the natural level of cosmogenic tritium in summer precipitation. The average tritium concentration of ice layers between 1923 and 1954 is 6.6 ± 2.0 TU, which is lower than that of the estimations for the closest monitoring stations during SC24 (8.5 TU for Locarno and 11.2 TU for Sion-Grimsel-Guttannen, Figures S3, S4, and S6 in Supporting Information S1). It is in a good agreement with MIROC5-iso, which simulates 5.46 ± 0.83 TU for the average tritium level of summer precipitation at Colle Gnifetti. The tritium time series has variation which correlates to the secondary neutron count rate, hence the link between the atmospheric cosmogenic tritium and the solar cycle has been confirmed once again. This finding agrees well with the MIROC5-iso results. A viable way to validate AGCMs is to study tritium in ice layers. Polar and continental ice records have already provided tritium time series, however mainly back to the bomb peak. Sensitive tritium analyses will contribute to the knowledge of natural level and natural variation of tritium of

precipitation over the world. Our work fills a gap in understanding variation of natural tritium of the pre-nuclear period, particularly in its relationship with solar activity. By providing a high-resolution data set, this study enhances the validation of global atmospheric models, which rely on accurate isotope data to simulate past climate conditions and validate predictions. Studying ice cores in Antarctica (Fourré et al., 2018), Greenland (Du et al., 2016; Nakazawa et al., 2021), Svalbard (Isaksson et al., 2001; Van der Wel et al., 2011), Severnaya Zemlja (Opel et al., 2009), Alaska (Tsushima et al., 2015), Tibetan Plateau (Shao et al., 2017), Caucasus (Mikhalenko et al., 2015), Altai (Henderson et al., 2006; Herren et al., 2013), or the Andes (Schotterer et al., 2003) will have a strong impact to advancing tritium time series to verify AGCMs. Additionaly, our work highlights the important role of current precipitation isotope monitoring networks, since only ongoing monitoring activities can provide recent and up to date data that can be compared to the natural isotope fingerprint of ice cores.

One of the key challenges for future research is overcoming the analytical limitations of tritium measurement, especially given the isotope's short half-life of 12.32 years. As tritium decays, the remaining amount after many decades becomes increasingly difficult to detect using current techniques. To extend the temporal range of tritium studies, it will be essential to develop more advanced methodologies. Although tritiogenic ^{3}He has been proposed as a proxy, the fast diffusion of helium in ice impedes this approach. Nevertheless, pre-bomb tritium in ice is still visible due to sensitive analytical techniques, but it will completely disintegrate below the best detections limits in 10–20 years. To perform such a work, the following requirements have to be fulfilled: (a) continuous, vertical ice deposition without post depositional processes and re-accumulation, (b) well dated profile using multiple proxies, like layer counting, seasonality of isotopes or dissolved ions, time markers like volcanic eruptions, tritium bomb peak, etc., (c) large sample size for sensitive tritium analyses, (d) careful ice coring to avoid contamination from the ambient tritium-rich moisture, and (e) sensitive tritium analysis using counting techniques with high efficiency electrolytic enrichment or ^{3}He ingrowth with mass spectrometry. The research of the coming years will have to find the possibility to study this excellent natural tracer in ice.

Data Availability Statement

The age-depth profile of the KCC site at Colle Gnifetti is available in Bohleber et al. (2018). The tritium profile of Fiescherhorn has been retrieved from Schotterer et al. (1998) and Schwikowski et al. (1999). The neutron count rate has been obtained from Usoskin et al. (2002). Sunspot data are from the World Data Center SILSO, Royal Observatory of Belgium, Brussels (SILSO, World Data Center, 2024). The tritium time series of Ottawa and Vienna are available in the Global Network of Isotopes in Precipitation (GINP) database (IAEA, 2024). The script used to support the spectral analysis in this study is available at Zenodo (Version 1.1.0) and is preserved at https://doi.org/10.5281/zenodo.14947829 (Palcsu et al., 2025).

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