No signature of extreme solar energetic particle events in high-precision $^{14}$C data from the Alaskan tree for 1844–1876 CE

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Abstract – Cosmogenic nuclides – $^{14}$C from tree rings and $^{10}$Be and $^{36}$Cl from ice cores serve as an effective proxy for past extreme solar energetic particle (SEP) events. After identifying the first signature of an extreme SEP event in 774 CE, several candidates have been found in these proxy archives, such as 993 CE, 660 BCE, and 7176 BCE. Their magnitudes have been estimated to be tens of times larger than that of the largest SEP event ever observed since the 1950s. Although a detailed survey of such extreme SEP events is ongoing, the detection of intermediate-sized SEP events that bridge the gap between modern observations and extreme events detected in cosmogenic nuclides has not progressed sufficiently, primarily because of the uncertainties in cosmogenic nuclide data. In this study, we measured $^{14}$C concentrations in tree rings in the 19th century (1844–1876 CE) to search for any increases in $^{14}$C concentrations corresponding to intermediate-size extreme SEP events. We utilized Alaskan tree-ring samples cut into early and late woods to suppress the potential seasonal variations in intra-annual $^{14}$C data. Notably, no significant $^{14}$C variations were observed between early and late woods (0.0 ± 0.3 ‰), and the annual resolution $^{14}$C data series displayed an error of ~ 0.8 ‰. Over the entire study period, no significant increase in $^{14}$C concentrations characterized by other candidates of extreme SEP events such as the 774 CE event was detected in the annual $^{14}$C data. The present result imposes a constraint on the SEP fluence when the largest class of recorded solar storms occurred (especially those in 1859 CE and 1872 CE).

Keywords: Radiocarbon / Solar energetic particle / Extreme solar event / Tree ring / Carrington event

1 Introduction

Solar eruptions cause various phenomena such as geomagnetic storms and solar energetic particle (SEP) events (solar radiation storms) in the solar-terrestrial environments (Riley et al., 2018; Miyake et al., 2019; Cliver et al., 2022; Kusano, 2023). Some of these phenomena are referred to as extreme events, on the basis of their large magnitude and infrequently. In certain instances, events with once-in-a-century to once-in-a-millennium occurrences are also regarded as extreme events (Gopalswamy, 2018; Cliver et al., 2022). As these extreme events could pose enormous space-weather effects, it is essential to understand the characteristics of these extreme events, especially regarding the occurrence rate and upper magnitude limit of these events (Riley et al., 2018; Hapgood et al., 2021; Kusano, 2023). Signatures of extreme solar events that occurred beyond modern observations have been investigated using cosmogenic nuclides in the Earth's natural archives (e.g., Miyake et al., 2019; Usoskin, 2023).

Thus far, cosmogenic nuclides, namely, $^{14}$C from tree rings and $^{10}$Be and $^{36}$Cl from ice cores, have served as proxies for past extreme SEP events (e.g., Miyake et al., 2019; Usoskin, 2023). These cosmogenic nuclides are produced by energetic particles, primarily in the stratosphere and upper troposphere (Golubenko et al., 2022), and accumulate in tree rings and ice sheets through atmospheric transportation. Despite the effects of transportation, such as the attenuation of the original signal of the energetic particles and background variation (e.g., seasonal effect on the deposition of cosmogenic nuclides), extreme SEP events that significantly exceed the background production

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of cosmogenic nuclides are recorded as spikes in cosmogenic nuclide datasets (Miyake et al., 2019). To date, several candidates of extreme SEP events have been discovered, e.g., those in 774 CE, 993 CE, 660 BC, and 7176 BCE as reported in previous studies (Miyake et al., 2012, 2013, 2015; Mekhaldi et al., 2015; Park et al., 2017; O’Hare et al., 2019; Brehm et al., 2022; Paleari et al., 2022). Based on the information derived from the \(^{10}\text{Be}/^{36}\text{Cl}\) ratio of ice cores, these candidates of extreme SEP events exhibit a hard energy spectrum, corresponding to events such as ground level enhancement (GLE) in February 1956 (GLE #5) and that in 2001 (GLE#69) (Mekhaldi et al., 2015; O’Hare et al., 2019; Koldobskiy et al., 2022; Paleari et al., 2022), which display the hardest energy spectra in the observed GLEs (Usoskin et al., 2020a). The SEP event in 774 CE has been estimated to be 40–100 times larger than GLE#5, which recorded the greatest flux and hardest spectra in the observational history (Cliver et al., 2022). In addition, other possible candidates of extreme SEP events have been reported in 1279 CE, 1054 CE, 5410 BCE, and 5259 BCE; whereas, these events have only been investigated through tree-ring \(^{14}\text{C}\) records (Brehm et al., 2021; Miyake et al., 2021; Miyahara et al., 2022; Brehm et al., 2022) and await further confirmations of the origin on the basis of analyses of ice core records. Compared with the largest events (e.g., 774 CE and 7176 BCE), those at 1279 CE, 1054 CE, and 5410 BCE display small \(^{14}\text{C}\) enhancements, i.e., \(\approx 5\)‰ increase in \(^{14}\text{C}\) corresponding to \(\approx 1/3\)rd of the largest event (Brehm et al., 2022).

In contrast, geomagnetic storms have been measured with magnetograms since the early 19th century (e.g., Chapman & Bartels, 1940; Beggen et al., 2023). Especially from the International Geophysical Year (IGY: 1957–1958), their intensity has been quantified with the Dst index (WDC Kyoto et al., 2015). Recently, our understanding of past extreme geomagnetic storms has been considerably improved, due to the numerous studies on historical geomagnetic storms using geomagnetic measurements. So far, the greatest geomagnetic storms have been located in September 1859, February 1872, and May 1921 (Cliver et al., 2022; Hayakawa et al., 2023a,b). Further beyond, auroral records allow us to further extend our chronology back to the 10th century BCE (Hattori et al., 2019; Hayakawa et al., 2019b; Van der Sluijs & Hayakawa, 2022).

Magnitudes of extreme SEP event candidates detected so far using cosmogenic nuclides are estimated to be tens of times larger than those of the largest GLE recorded in modern observations (Mekhaldi et al., 2015; Cliver et al., 2022). As such, these events occur only once every several centuries to 24 centuries (Brehm et al., 2022), thereby rendering their classification as “extreme” extreme SEP events. According to Usoskin et al. (2023), both the occurrence distributions of SEP events, i.e., the occurrence frequencies of extreme events observed by cosmogenic nuclides data and SEP events observed in space era, can be fitted using the same Weibul distribution function. This finding signifies that the extreme events are an extension of the events detected in modern observations. However, extreme SEP events that have a magnitude in the gap between modern observations and “extreme” extreme SEP events, i.e., once in every several centuries, have not been extensively investigated owing to several uncertainties involved in the cosmogenic nuclides data – statistical and systematic errors in measurements and insufficient separation from background fluctuations (Usoskin et al., 2020b, 2023; Usoskin & Kovaltsov, 2021). Moreover, none of the extreme events detected thus far in cosmogenic nuclides have been definitively determined to originate from extreme solar eruptions by means other than cosmogenic nuclides information (although current knowledge indicates that non-solar origins are unlikely; see Usoskin, 2023). Therefore, it is crucial to detect small enhancements of cosmogenic nuclides (corresponding to extreme SEP events) during the period that has been shown to be occurrences of extreme solar eruptions by alternative methods.

In this study, we investigated \(^{14}\text{C}\) concentrations in tree rings from 1844 to 1876 CE to search for potential minor \(^{14}\text{C}\) enhancements. The target period was deemed suitable for this study because of the following reasons:

1. This period accommodates the Carrington event. This event has been associated with the greatest solar flare in observational history (Cliver & Dietrich, 2013; Hudson, 2021; Hayakawa et al., 2023a);
2. This period includes at least three extreme geomagnetic storms (minimum Dst index < −500 nT; Cliver et al., 2022) on 28 August 1859 CE, 2 September 1859 CE (Carrington event; Cliver & Dietrich, 2013; Hayakawa et al., 2022a), and 4 February 1872 CE (Chapman–Silverman event; Silverman, 2008; Hayakawa et al., 2023b), as summarized in Table 1. These geomagnetic superstorms indicate relatively high solar activities and potential occurrences of extreme SEP events;
3. The background variations of the Schwabe cycle can be estimated from sunspot number (e.g., Clette et al., 2023); and
4. The \(^{14}\text{C}\) dilution “Suess effect” caused by anthropogenic fossil fuels, is minimal (Suess 1955; Stuiver & Quay 1981).

To suppress the influence of seasonal changes of \(^{14}\text{C}\) concentrations caused by atmospheric transportation, the \(^{14}\text{C}\) analysis was conducted with seasonal resolution using earlywoods and latewoods.

## 2 Sample and methods

For this study, wood samples were obtained from *Picea sitchensis* (Bong.) Carrière (sitka spruce) from Alaska, USA (Hakozaki & Nakamura, 2013, Figs. S1 and S2). As the tree-ring width series of the sample exhibits the highest correlation with the master chronology of the Prince of Wales Island (Bailie–Pilcher \(t\)-value \(t_{BP} = 9.36\), Baillie & Pilcher, 1973) among several Alaskan master chronologies (ITRDB, https://www.ncdc.noaa.gov/paleosearch/study/15215), we can presume that the sample was produced from the Prince of Wales Island or the surrounding region of the southeastern Alaska Pacific coastal area. We utilized two wood specimens, with sample IDs (Hakozaki & Nakamura, 2013) denoted as AKNC10 and AKNC16 for the measurement periods of 1876–1849 CE and 1851–1844 CE, respectively.

Each tree ring was segmented into earlywood and latewood using a utility knife and a graver (only the ring sample of 1849 CE from AKNC16 remained whole because of the difficulty in separating early and latewoods). Holocellulose was obtained by
chemically processing the sample as follows: (1) 3 M HCl treatment at 68 °C for overnight, (2) 1.2 M NaOH treatment at 68 °C for overnight, (3) 3 M HCl at 70 °C for 3 h, (4) a bleaching treatment (HCl/NaClO2) at 77 °C for overnight and an additional bleaching treatment for a few hours until the sample color became white, (5) a neutralization with Milli-Q water, and (6) drying the sample in a drying oven. Between each procedure from (1) to (4), a Milli-Q water step for ~1 h was interposed. The wood separation and chemical treatment were performed at Nagoya University.

The holocellulose samples were graphitized using AGE at ETH Zürich (Wacker et al., 2010) and their 14C concentrations were measured using the Micadas at ETH Zürich (Sookdeo et al., 2020). Each earlywood and latewood sample was measured two times to reduce uncertainties and confirm reproducibility (only the latewood 1846 CE was measured once). Brown coal (Reichwalde, Germany) was used as a process blank.

3 Results and discussion

3.1 14C data

The weighted mean of the Δ14C measurements of early and latewoods across the study period from 1844–1876 CE is depicted in Figure 1a. As observed, the two measurements from the same layers were consistent with each other (Table S1). Although the layer of earlywood in 1858 CE yielded a chi-square value of 4.4 (square of differences in Δ14C/sum of squared errors) and was rejected at the 95% confidence level as the layer with the weakest correspondence, it can be considered to be statistically acceptable among the total entries of ~70 layers. Overall, the radiocarbon ages (1849–1851 CE) of AKNC10 and AKNC16 were consistent.

3.2 14C Difference between early and late woods

As depicted in Figure 1a, the distribution between earlywood and latewood resided within the margin of error. The average disparity between latewood and earlywood throughout the measurement period was 0.0 ± 0.3‰ (the error was calculated through an error propagation), and the values between the early and latewoods were not discernible even with the high precision of the measurements. Conversely, recent atmospheric 14C data reveal a pronounced seasonal variation in 14C concentrations, signifying that the principal stratosphere–troposphere exchange.
occurs in the northern hemisphere during spring (e.g., Kitagawa et al., 2004; Levin et al., 2010; Leuenberger et al., 2018). According to the atmospheric $^{14}$C data presented by Levin et al. (2010), there exists $a \approx 5\%$ $\Delta^{14}$C increase from early spring to autumn at mid-high latitude observational points in the northern hemisphere (e.g., at Alert and Jungfraujoch stations), which corresponds to locations at similar latitudes to our Alaskan tree sample. Considering the general season of ring formation – spring to summer for earlywoods and, summer to autumn for latewoods, the present data should exhibit a difference between early and late woods in case of a similar seasonal variation in the atmosphere.

In addition to the $^{14}$C data of the contemporary atmosphere, analyses of actual tree rings from the pre-industrial period have reported a difference of approximately $3\%e$ between earlywoods and latewoods (e.g., McDonald et al. (2019). McDonald et al. (2019) explored the possibility that the disparities between early and latewood reflect seasonal changes in the atmosphere, as well as the utilization of stored carbon in early spring (earlywood), as shown by the $^{14}$C analyses of their deciduous tree samples. Since the Sitka spruce used in this study is an evergreen coniferous tree, and it is generally understood that the use of stored carbon during tree-ring formation is minimal, our results suggest the potential that early and late wood ring formation occurred within a shorter period than anticipated (for example, most of a ring may have formed intensively within a ~5-month period). If a comparable ~5-month period is present, the seasonal change existed in the 19th century in the region where our tree sample grew. However, there is a report on the utilization of photosynthetic products from the previous year for Siberian Larix gmelinii (deciduous coniferous tree) based on the $^{13}$CO$_2$ pulse-labeling (Kagawa et al., 2006), and the utilization of longer-term stored carbon may be possible for our samples. Considering that research on seasonal changes in atmospheric $^{14}$C in premodern periods, the timing of tree-ring formation, and the utilization of stored carbon across various tree species and regions is limited, additional investigations will be necessary.

Specifically, to examine marginal alterations in $^{14}$C ($< ~5\%e$), understanding the characteristics of seasonal $^{14}$C variations in the specific tree sample used becomes vital for eliminating uncertainties related to the seasonal variations.

### 3.3 $^{14}$C variation from 1844–1876 CE

As we were unable to discern the difference between early and latewood in our data, we calculated the weighted average $\Delta^{14}$C data for the same year. Figure 1b presents time series data with annual resolution, spanning from 1844 to 1876 CE, in conjunction with a recently acquired annual-resolution $^{14}$C dataset (Brehm et al., 2021). Within the current dataset, the increase in $^{14}$C was not significant compared to the errors (average error: 0.8$\%e$ in $\Delta^{14}$C). Figure S3 displays a histogram of the yearly $^{14}$C difference, which is fitted by a normal distribution. Here, although we did not include any additional uncertainty caused by the sample preparations (e.g., Sookdeo et al., 2020), the value of the average error is considered reasonable compared to the normal distribution parameter of yearly $^{14}$C difference (1-sigma: 1.1$\%e$, Fig. S3). Therefore, no extreme SEP event could be detected during the survey period, similar to the lower-resolution $^{14}$C data from previous studies (e.g., Stuiver et al., 1998; Brehm et al., 2021) (the SEP-driven $^{14}$C variation is typically characterized by a single-year elevation, followed by a sustained period of several years with high concentration data). Although a relatively steep decrease was noted after 1870 CE, this decrease may be influenced by the Suess effect. According to Stuiver and Quay (1981), the influence of fossil fuel on $^{14}$C data after 1860 CE is detectable, and its impact is $\approx 2\%e$ from 1860 to 1880 CE, which aligns with the diminishing pattern observed in the present data post-1860 CE.

Figure 2 illustrates a comparison between the sunspot records and the 3-year moving average of the present dataset. The phase of an 11-year cycle is shifted by approximately 3 years in the tree-ring $^{14}$C data, owing to the global carbon cycle (Siegenthaler et al., 1980; Scifo et al., 2019). Accounting for this delay in $^{14}$C data, a similar cyclic fluctuation can be observed in the $^{14}$C data according to sunspot variations. Consequently, the decadal variability in our $^{14}$C data is deemed to reflect the Schwabe cycle (with an amplitude of 1–2 $\%e$), thereby indicating the absence of extreme SEP events of a magnitude that would disrupt this Schwabe cycle variation.

### 3.4 Relationship between extreme flare and SEP events

The three geomagnetic storm events that transpired in August and September 1859 CE and February 1872 CE, were categorized as the largest class of geomagnetic superstorms (min Dst $\leq -500$ nT; Cliver et al., 2022). Specifically, the magnitudes of the geomagnetic superstorms in September 1859 and 1872 CE surpass any geomagnetic storms observed in modern times, except for the superstorm in May 1921 (Table 1 of Hayakawa et al., 2019a). The frequency of such extreme geomagnetic storm events can be estimated to be less frequent than once in every century based on the calculated Dst indices (Gopalswamy, 2018). Regarding the Carrington event in 1859 CE, a recent investigation re-estimated the flare magnitude as X80 (X46–X126) based on Carrington’s original drawings for
white-light flare regions and the source active regions as well as Carrington and Hodgson’s reports for the flare duration and flare brightness (Hayakawa et al., 2023a). This estimation is greater than previous estimations (X64.4 ± 7.2) based on the amplitude of the resultant magnetic crochets (Owens et al., 2022). In addition to these three geomagnetic storms occurred in August and September 1859 CE and February 1872 CE, geomagnetic data acquired by several geomagnetic observatories denote presence of extreme geomagnetic storms in October 1847 CE, October 1870, and October 1859 CE (Vaquero et al., 2008; Lakhina et al., 2012; Cliver & Dietrich 2013); which requires further future investigations.

Consequently, we deduced that intense solar activity caused extremely large-scale solar flares and geomagnetic storms during the survey period of this study, and a significant volume of SEP accordingly reached the Earth. Owens et al. (2022) highlighted that the “event-by-event correspondence” between geomagnetic storms andGLEs is low when utilizing datasets of the modern era. However, higher solar activities are inclined to generate large-scale solar flares, and therefore, extensive magnetic storms and GLEs.

The magnitudes of extreme solar flares of ≥X14.3 (≥X10: unrecalibrated classification in GOES data prior to GOES16, https://www.sws.bom.gov.au/Educational/2/3/9; see also Cliver et al., 2022; Machol et al., 2022) occurring after 1976 CE (considered the largest flare if several extreme flares occurred in the same year) and cumulative F200 [MeV] values from GLEs (Kovaltsov et al., 2014) occurring in the same year as the extreme solar flares are comparatively presented in Figure 3. Herein, we assumed that a large-scale event occurred in the same year, even in the case of a weak one-to-one relationship between flares and SEP events. In addition to the modern data, the upper limit of the cumulative F200 in 1859 CE derived herein is exhibited in Figure 3. Here, the upper limit is assumed to be two times the 14C average error and scaled to the total 14C increment of the 774 CE event, i.e., 17.6‰ (Brehm et al., 2022), using an estimated F200 value for the 774 CE event (Usoskin et al., 2023; Table 1). We observed that the upper limit does not deviate from a fitted power function of the modern data. Although the present samples did not display any significant increase in 14C concentrations after 1859 CE, a significant yet marginal 14C enhancement has been confirmed in a tree sample collected from a substantially high latitude in Finland, which may be correlated to SEPs (Usitalo et al., private communications). To confirm this possibility, further investigation may yield the SEP fluence of the Carrington event, whereas we need ice core datasets (10Be and 36Cl) and higher precision tree-ring 14C datasets.

4 Conclusions

This study conducted high-precision 14C measurements to investigate the increases in 14C concentrations caused by intermediate SEP events between GLEs and “extreme” extreme SEP events detected by modern observations and cosmogenic nuclides, respectively. The analysis utilized Sitka spruce tree-ring samples from Alaska for the period spanning from 1844 to 1876 CE, during which several extreme geomagnetic storms and the Carrington solar flare (1859 and 1872 CE) occurred. As conventional 14C analyses in annual tree rings (viz. time resolution of one year) can be affected by variations in the scale of a few permil of 14C induced by the seasonal atmospheric alteration in 14C concentrations owing to stratosphere–troposphere transportation, we carefully segmented the early and latewoods to minimize this influence. In the 14C data of the Sitka spruce sample between early and latewoods, no significant difference was observed with high accuracy (Δ14C difference: 0.0 ± 0.3‰), alluding to the possibility that tree-ring formation transpired in a briefer period than anticipated. Further investigations concerning the seasonal change in various species across expansive regions are required to deem the seasonal variations in 14C concentrations as a background fluctuation of marginal variations in 14C concentrations.

In the obtained annual 14C series, no significant increase in 14C concentrations was detected with respect to the errors (average error: 0.8‰). The error of the 14C data obtained herein was approximately 1/20th of the total increase in 14C concentrations for the 774 CE event (17.6‰; Brehm et al., 2022), which represents the largest class in the Holocene. We distinguished fine structures in the 14C data, which were consistent with the Schwabe cycle estimated from the sunspot data, implying that no SEP event occurred during the survey period that would disrupt the 14C fluctuations caused by the Schwabe cycle with an amplitude of ~1–2‰. Concerning the Carrington event in September 1859 CE, which is associated with one of the largest flares and geomagnetic storms on record, the upper limit (detection limit) of the SEP event obtained herein is in accord with the magnitude of the SEP event projected from contemporary data. Thus, it is not unexpected that SEP events exist slightly beneath the detection threshold. Although we did not detect any indicators of an extreme SEP event in the three

Figure 3. Relationship between SXR peak intensity of extreme solar flare (largest intensity within a year, >X10 in the previous category, https://www.sws.bom.gov.au/Educational/2/3/9) and integrated SEP fluence above 200 MeV (F200 [cm−2], Kovaltsov et al., 2014) for the same year of the extreme solar flare, for the period from 1976 to 2020 CE (circles). The flare scale for the period 1976–2017 CE is recalibrated (i.e., the previous SXR peak intensity × 1.43) (Cliver et al., 2022). Estimations of SXR peak intensity for the Carrington events are reported in Hayakawa et al. (2023a). The arrow indicates the upper limit of F200 for the Carrington event (see text).
pronounced solar events in the 19th century, previous research has documented more signatures of extreme solar storms in other periods (e.g., 1770, 1903, and 1909 CE: Hayakawa et al., 2017b, 2019c, 2020). Therefore, we might uncover intermediate-size extreme SEP events that are detectable with the current measurement precision in these ages.

Supplementary material

The supplementary material of this article is available at https://www.swsc-journal.org/10.1051/swsc/2023030/olm.

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