

## SEARCH FOR ANNUAL $^{14}\text{C}$ EXCURSIONS IN THE PAST

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**ABSTRACT.** Two radiocarbon excursions (AD 774–775 and AD 993–994) occurred due to an increase of incoming cosmic rays on a short timescale. The most plausible cause of these events is considered to be extreme solar proton events (SPE). It is possible that there are other annual  $^{14}\text{C}$  excursions in the past that have yet to be confirmed. In order to detect more of these events, we measured the  $^{14}\text{C}$  contents in bristlecone pine tree-ring samples during the periods when the rate of  $^{14}\text{C}$  increase in the IntCal data is large. We analyzed four periods every other year (2479–2455 BC, 4055–4031 BC, 4465–4441 BC, and 4689–4681 BC), and found no anomalous  $^{14}\text{C}$  excursions during these periods. This study confirms that it is important to do continuous measurements to find annual cosmic-ray events at other locations in the tree-ring record.

**KEYWORDS:** cosmogenic nuclides, annual cosmic-ray event.

## INTRODUCTION

Annual large increases and subsequent decay in the radiocarbon content of tree rings were originally found in Japanese tree-ring samples. These increases were reported in the periods from AD 774–775 and AD 993–994 (Miyake et al. 2012, 2013). The AD 775 event has been confirmed by independent measurements using different trees from all over the world (Usoskin et al. 2013; Jull et al. 2014; Güttler et al. 2015). On the other hand, although the AD 994 event was confirmed by  $^{14}\text{C}$  measurements of several tree-ring samples, it is disputed whether the event occurred between AD 992–993 or AD 993–994 (Miyake et al. 2014; Lukas Wacker, personal communication, 2016).

The best explanation is that these events reflect rapid increases of incoming cosmic-ray intensity within 1 yr. Possible causes have been proposed by several studies, including a nearby supernova, a cometary impact on the Earth, a gamma-ray burst, and an extreme solar proton event (SPE) (Eichler and Mordecai 2012; Miyake et al. 2012, Hambaryan and Neuhäuser 2013; Pavlov et al. 2013; Thomas et al. 2013; Usoskin et al. 2013; Cliver et al. 2013; Liu et al. 2014; Mekhaldi et al. 2015). Recent studies regarding a quasi-annual measurement of  $^{10}\text{Be}$  concentrations in ice cores from Antarctica and Greenland reported corresponding  $^{10}\text{Be}$  increases around AD 775 and AD 994 (Mekhaldi et al. 2015; Miyake et al. 2015; Sigl et al. 2015). Considering the existence of the  $^{10}\text{Be}$  peaks in both hemispheres around two cosmic-ray events, it is highly likely that the sources of the two  $^{14}\text{C}$  increase events are extreme SPEs (Usoskin et al. 2013; Mekhaldi et al. 2015; Miyake et al. 2015).

The scale of the AD 775 event has been estimated as ~50 times larger than the extreme SPE that occurred in AD 1956 (Usoskin and Kovaltsov 2012; Usoskin et al. 2013), or more than 5 times larger than the largest historical SPE (Mekhaldi et al. 2015). Although annual  $^{14}\text{C}$  data exist around AD 1856 when the historical largest Carrington flare occurred, the data during this period show no increase (Miyake et al. 2013; Jull et al. 2014). If we assume the  $^{14}\text{C}$  increase of the Carrington flare is within the measurement error (~2%) around AD 1856, the AD 775 event should be at least 10 times larger than the Carrington event. If such a large SPE occurred today,

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heavy damage would result on our modern electronic society. It is very important to investigate an occurrence rate of these events to contribute to our understanding of space weather and to understand the frequency of solar activities. Also, such a  $^{14}\text{C}$  excursion can be useful to give an age determination with 1-yr precision where it occurs for historical or geological samples. For example, Wacker et al. (2014) were able to date a wooden beam in a church to 1 yr based on this approach. A  $^{14}\text{C}$  event also gives a new possibility to date ice cores with 1-yr resolution (Sigl et al. 2015).

The findings of the two  $^{14}\text{C}$  increase events in the last 2 millennia indicate that more yet undetected events are available in tree-ring records going back to 12,000 BP. However, without the  $^{14}\text{C}$  measurements with annual or at least biannual time resolution, we cannot detect such  $^{14}\text{C}$  excursions. Although smaller annual variations are not observable in the IntCal (Reimer et al. 2013) data with 5-yr resolution due to the averaging IntCal employs, it is possible that a large annual  $^{14}\text{C}$  increase event would appear in the IntCal data. Actually, the increase rate ( $\%/\text{yr}$ ) of the AD 775 event is one of the largest ( $0.4\%/\text{yr}$ ) in the IntCal13 data (Reimer et al. 2013) during the past 12,000 yr. There are 15 events where increase rates are larger than  $0.3\%/\text{yr}$  in the IntCal13 data for these 12,000 yr (Figure 1), and it is possible that annual  $^{14}\text{C}$  increase events are hidden in these periods. We report  $^{14}\text{C}$  results for four time intervals (4680, 4440, 4030, and 2455 BC), which show rates of increase larger than  $0.3\%/\text{yr}$  with 2-yr time resolution.

## SAMPLES AND METHODS

We analyzed four bristlecone pine (*Pinus longaeva*) samples (Figure 2). All of the samples were dated by the dendrochronological method and are now archived at the Laboratory of Tree-Ring Research (LTRR) at the University of Arizona in Tucson. The samples were collected in the White Mountains of California ( $37.3794^\circ\text{N}$ ,  $118.1654^\circ\text{W}$ ) as part of a decades-long effort by multiple researchers at the LTRR (see, for example, Ferguson 1969; LaMarche and Harlan 1973; Salzer et al. 2014). We separated annual rings carefully using a knife under a dissecting microscope. Our study focused on four intervals in the BC time period: 2479–2455,

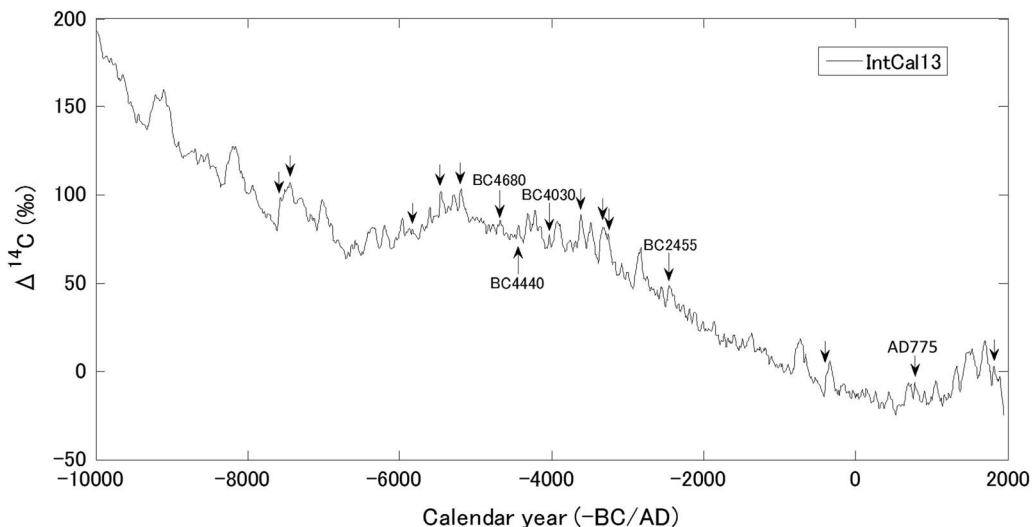


Figure 1 Carbon-14 content ( $\Delta^{14}\text{C}$ ) for the last 12,000 yr (IntCal13; Reimer et al. 2013). The arrows show the periods when the increase rates of the  $\Delta^{14}\text{C}$  data are more than  $0.3\%/\text{yr}$ . We analyzed the 4680, 4440, 4030, and 2455 BC time intervals in this paper.

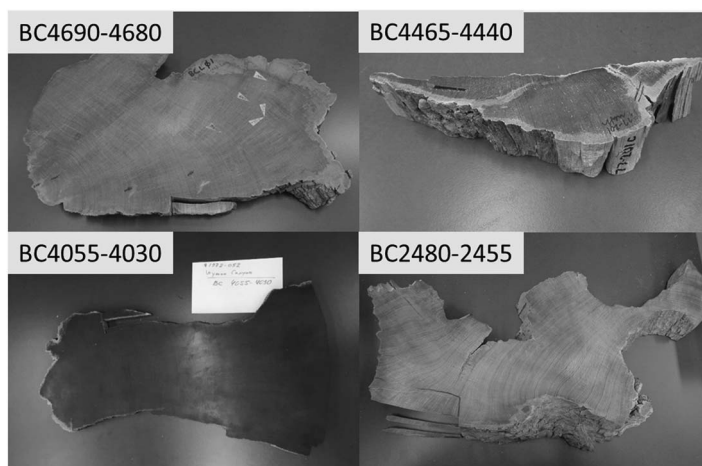


Figure 2 Bristlecone pine samples for this study. These samples came from the White Mountains of California, USA (37.3794°N, 118.1654°W).

4055–4031, 4465–4441, and 4689–4681 BC. The time resolution of our measurement is 2 yr (i.e. we measured every other annual ring). There were no missing rings in the intervals examined.

We extracted hemi-cellulose from sliced wood samples by a standard cellulose extraction method. Chemical cleaning consisted of an AAA treatment and a sodium chlorite treatment, and the cellulose samples were combusted and converted to graphite in the chemistry laboratory of the AMS laboratory in the University of Arizona. The  $^{14}\text{C}$  contents were measured using the 2.5MV National Electrostatics Corporation AMS at the University of Arizona lab.

## RESULTS AND DISCUSSION

We obtained  $\Delta^{14}\text{C}$  data for four intervals by using the calculation method of Stuiver and Polach (1977). Figure 3 shows the measured results in  $\Delta^{14}\text{C}$  values (‰), which are compared with the IntCal13 curve and the original  $\Delta^{14}\text{C}$  data of IntCal13. The measured data are listed in Table S1 in the online Supplementary Material.

Although we expected to see the annual increase and subsequent decay in the  $\Delta^{14}\text{C}$  due to a cosmic-ray event, the data show no such variation. The 4680, 4440, and 2455 BC time intervals basically show a good agreement with the IntCal series within measurement errors. However, there are some offsets between our results, and the interpolated IntCal13 data were about 4.9‰ lower for the 4440 BC interval and 3.9‰ lower for the 2455 BC interval on average. On the other hand, some data of the 4030 BC time interval are significantly different from that of IntCal13. The data of 4045 and 4035 BC are more than  $3\sigma$  different ( $3\times$  measurement error) from the IntCal line. Although these two points (4045 and 4035 BC) increase rapidly, the following variation is not continuous. If there is only a cosmic-ray input with a short timescale ( $<1$  yr), the variation should be a rapid increase followed by decay like the AD 775 event (Miyake et al. 2012; Usoskin et al. 2013; Gütler et al. 2015). Since there is no possible natural origin to explain the two points of 4045 and 4035 BC, we hypothesize that these data deviate due to some experimental problems. It will be necessary to remeasure these points to establish the accurate  $^{14}\text{C}$  variation. Nevertheless, even if the two points are valid, the  $^{14}\text{C}$  pattern does not reflect an annual cosmic-ray event.

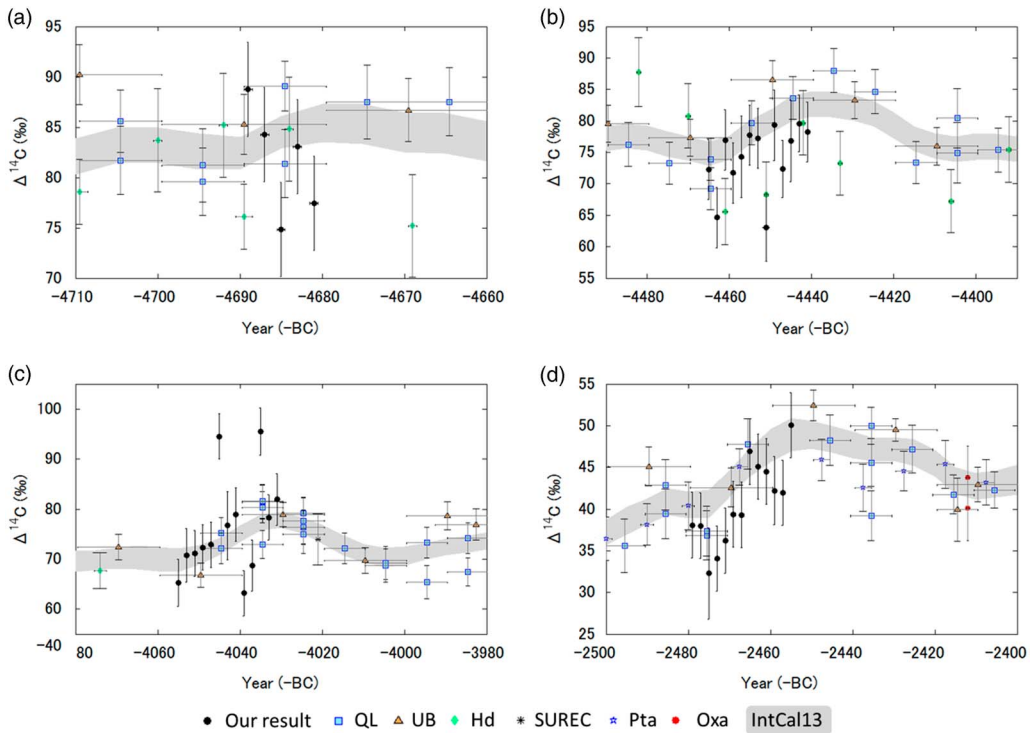


Figure 3 Comparison of measured results from this study (black circles), the original data of the IntCal13: QL: blue squares (Stuiver and Braziunas 1993), UB: orange triangles (Pearson et al. 1986), Hd: green diamonds (Kromer et al. 1986), SUREC: black star (Bronk Ramsey et al. 2012), Pta: blue stars (Vogel and van der Plicht 1993), and Oxa: red circles (Bronk Ramsey et al. 2012), and the IntCal13 data (gray line) (Reimer et al. 2013). Please see online version for color.

From the present measurements during the periods when the IntCal13 data show a large change, we determined the following: (1) the 4680 BC event does not show any increase; (2) the 4440 and 2455 BC events increase continuously, which is consistent with the IntCal data; (3) the 4030 BC event is almost consistent with the IntCal data but two points (4045 and 4035 BC) are significantly different from IntCal; and (4) we could not detect any annual cosmic-ray event like the AD 775 event for the four intervals.

In the case of the AD 994 event, the annual increase is not visible in the IntCal data due to the averaging of the IntCal data. Therefore, there may be other smaller annual cosmic-ray events that are not shown in the IntCal data. However, it seems unlikely that an event as strong as (or stronger than) the AD 775 event may be found in the Holocene, given that of the 15 best candidates for such  $^{14}\text{C}$  excursions (Figure 1), already five intervals (four intervals of this study plus the 19th century increase; Stuiver et al. 1998) have been demonstrated to lack the spike and decay pattern characteristic of cosmic-ray events.

## CONCLUSIONS

We measured the  $^{14}\text{C}$  content for the periods of 2479–2455, 4055–4031, 4465–4441, and 4689–5681 BC to investigate possible rapid  $^{14}\text{C}$  excursion events at annual resolution. The results obtained did not show any such annual events. In order to detect more annual  $^{14}\text{C}$  increase events, it is important to conduct a detailed survey of continuous annual  $^{14}\text{C}$  measurements.

We plan to survey continuous  $^{14}\text{C}$  data with 1-yr or 2-yr resolution over the last 12,000 yr in the future.

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## SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit <http://dx.doi.org/10.1017/RDC.2016.54>

## REFERENCES

- Bronk Ramsey C, Staff RA, Bryant CL, Brock F, Kitagawa H, van der Plicht J, Schlolaut G, Marshall MH, Brauer A, Lamb HF, Payne RL, Tarasov PE, Haraguchi T, Gotanda K, Yonenobu H, Yokoyama Y, Tada R, Nakagawa T. 2012. A complete terrestrial radiocarbon record for 11.2 to 52.8 kyr B.P. *Science* 338(6105):370–4.
- Clover EW, Tylka AJ, Dietrich WF, Ling AG. 2013. On a solar origin for the cosmogenic nuclide event of 775 A.D. *The Astrophysical Journal* 781:32.
- Eichler D, Mordecai D. 2012. Comet encounters and carbon 14. *The Astrophysical Journal Letters* 761:L27.
- Ferguson CW. 1969. A 7104-year annual tree-ring chronology for bristlecone pine, *Pinus aristata*, from the White Mountains of California. *Tree-Ring Bulletin* 29(3–4):3–29.
- Güttler D, Adolphi F, Beer J, Bleicher N, Boswijk G, Christl M, Hogg A, Palmer J, Vockenhuber C, Wacker L, Wunder J. 2015. Rapid increase in cosmogenic  $^{14}\text{C}$  in AD 775 measured in New Zealand kauri trees indicates short-lived increase in  $^{14}\text{C}$  production spanning both hemispheres. *Earth and Planetary Science Letters* 411:290–7.
- Hambaryan VV, Neuhäuser R. 2013. A galactic short gamma-ray burst as cause for the  $^{14}\text{C}$  peak in AD 774/5. *Monthly Notices of the Royal Astronomical Society* 430(1):32–6.
- Jull AJT, Panyushkina IP, Lange TE, Kukarskih VV, Myglan VS, Clark KJ, Salzer MW, Burr GS, Leavitt SW. 2014. Excursions in the  $^{14}\text{C}$  record at A.D. 774–775 in tree rings from Russia and America. *Geophysical Research Letters* 41(8):3004–10.
- Kromer B, Rhein M, Bruns M, Schoch-Fischer H, Münnich KO, Stuiver M, Becker B. 1986. Radiocarbon calibration data for the 6th to the 8th millennia BC. *Radiocarbon* 28(2B):954–60.
- LaMarche VC Jr, Harlan TP. 1973. Accuracy of tree ring dating of bristlecone pine for calibration of the radiocarbon time scale. *Journal of Geophysical Research* 78(36):8849–58.
- Liu Y, Zhang Z, Peng Z, Ling M, Shen CC, Liu W, Sun X, Shen C, Liu K, Sun W. 2014. Mysterious abrupt carbon-14 increase in coral contributed by a comet. *Nature Communications* 4:3728.
- Mekhaldi F, Muscheler R, Adolphi F, Aldahan A, Beer J, McConnell JR, Possnert G, Sigl M, Svensson A, Synal H-A, Welten KC, Woodruff TE. 2015. Multiradionuclide evidence for the solar origin of the cosmic-ray events of AD 774/5 and 993/4. *Nature Communications* 6:8611.
- Miyake F, Nagaya K, Masuda K, Nakamura T. 2012. A signature of cosmic-ray increase in AD 774–775 from tree rings in Japan. *Nature* 486(7402):240–2.
- Miyake F, Masuda K, Nakamura T. 2013. Another rapid event in the carbon-14 content of tree rings. *Nature Communications* 4:1748.
- Miyake F, Masuda K, Hakozaiki M, Nakamura T, Tokanai F, Kato K, Kimura K, Mitsutani T. 2014. Verification of the cosmic-ray event in AD 993–994 by using a Japanese Hinoki tree. *Radiocarbon* 56(4):1189–94.
- Miyake F, Suzuki A, Masuda K, Horiuchi K, Motoyama H, Matsuzaki H, Motizuki Y, Takahashi K, Nakai Y. 2015. Cosmic ray event of A.D. 774–775 shown in quasi-annual  $^{10}\text{Be}$  data from the Antarctic Dome Fuji ice core. *Geophysical Research Letters* 42(1):84–9.
- Pavlov AK, Blinov AV, Konstantinov AN, Ostryakov VM, Vasilyev GI, Vdovina MA, Volkov PA. 2013. AD 775 pulse of cosmogenic radionuclides production as imprint of a galactic gamma-ray burst. *Monthly Notices of the Royal Astronomical Society* 435(4):2878–84.
- Pearson GW, Pilcher JR, Baillie MGL, Corbett DM, Qua F. 1986. High-precision  $^{14}\text{C}$  measurement of Irish oaks to show the natural  $^{14}\text{C}$  variations from AD 1840 to 5210 BC. *Radiocarbon* 28(2B):911–34.
- Reimer PJ, Bard E, Bayliss A, Beck JW, Blackwell PG, Bronk Ramsey C, Buck CE, Cheng H, Edwards RL, Friedrich M, Grootes PM, Guilderson TP, Haffidason H, Hajdas I, Hatté C, Heaton TJ, Hoffman DL, Hogg AG, Hughen KA, Kaiser KF,

- Kromer B, Manning SW, Niu M, Reimer RW, Richards DA, Scott EM, Southon JR, Staff RA, Turney CSM, van der Plicht J. 2013. IntCal13 and Marine13 radiocarbon age calibration curves, 0–50,000 years cal BP. *Radiocarbon* 55(4):1869–87.
- Salzer MW, Bunn AG, Graham NE, Hughes MK. 2014. Five millennia of paleotemperature from tree-rings in the Great Basin, USA. *Climate Dynamics* 42(5):1517–26.
- Sigl M, Winstrup M, McConnell JR, Welten KC, Plunkett G, Ludlow F, Büntgen U, Caffee M, Chellman N, Jensen DD, Fischer H, Kipfstuhl S, Kostick C, Maselli OJ, Mekhaldi F, Mulvaney R, Muscheler R, Pasteris DR, Pilcher JR, Salzer M, Schüpbach S, Steffensen JP, Vinther BM, Woodruff RE. 2015. Timing and climate forcing of volcanic eruptions for the past 2,500 years. *Nature* 523(7562):543–9.
- Stuiver M, Braziunas TF. 1993. Sun, ocean, climate and atmospheric  $^{14}\text{C}$ : an evaluation of causal and spectral relationships. *The Holocene* 3(4): 289–305.
- Stuiver M, Polach HA. 1977. Discussion: reporting of  $^{14}\text{C}$  data. *Radiocarbon* 19(3):355–63.
- Stuiver M, Reiver PJ, Braziunas TF. 1998. High-precision radiocarbon age calibration for terrestrial and marine samples. *Radiocarbon* 40(3):1127–51.
- Thomas BC, Melott AL, Arkenberg KR, Snyder BR II. 2013. Terrestrial effects of possible astrophysical sources of an AD 774–775 increase in  $^{14}\text{C}$  production. *Geophysical Research Letters* 40(6):1237–40.
- Usoskin IG, Kovaltsov GA. 2012. Occurrence of extreme solar particle events: assessment from historical proxy data. *The Astrophysical Journal* 757:92.
- Usoskin IG, Kromer B, Ludlow F, Beer J, Friedrich M, Kovaltsov GA, Solanki SK, Wacker L. 2013. The AD775 cosmic event revisited: the Sun is to blame. *Astronomy & Astrophysics* 552:L3.
- Vogel JC, van der Plicht J. 1993. Calibration curve for short-lived samples, 1900–3900 BC. *Radiocarbon* 35(1):87–91.
- Wacker L, Güttler D, Goll J, Hurni JP, Synal H-A, Walti N. 2014. Radiocarbon dating to a single year by means of rapid atmospheric  $^{14}\text{C}$  changes. *Radiocarbon* 56(2):573–9.