Variation of $^{14}$C concentrations of single-yr tree rings at the rapid change in 2600-yrBP

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Abstract: We have measured the $^{14}$C concentrations of old tree rings with the measurement accuracy 0.2% to search an ancient cosmic ray intensity fluctuation. The $^{14}$C concentrations of single-yr tree rings using the Choukai Jindai cedar in 2650 to 2600 yr BP showed a rapid change with the decreasing rate of 11‰ for 7 years.

Introduction

The flux of cosmic rays reaching the earth are modulated by time variations of geomagnetic and heliomagnetic fields. Solar wind associated solar activity affects the magnetic field in the heliosphere and hence causes a variation of the galactic cosmic rays invaded in the heliosphere. At present the solar activity indicates a typical periodic behavior, comprising the 11-year variation of sunspot number and hence the magnetic field. Moreover, the polarity of the solar magnetic fields changes with a period of 22-year.

While we have no available observed data of the sunspot number for years earlier than A.D. 1610, cosmogenic nuclide $^{14}$C is available to investigate the variations of cosmic rays in past time [1]. Since $^{14}$C is produced by galactic cosmic rays in the atmosphere of the earth, the production yield is anti-correlated with the solar activity. The $^{14}$C forms carbon dioxide, which exchanges among the various reservoirs in the global carbon cycle. Some of the carbon dioxide produced in the atmosphere is built into plants. Therefore, $^{14}$C concentrations of single-yr tree rings indicate solar modulations of cosmic rays such as 11-yr solar cycle. Thus, old tree rings are a powerful tool to detect the variations of cosmic rays in past time.

Figure 1: Locations of the Choukai Jindai cedar (photograph) and Maunder era in the time profile of $\Delta^{14}$C by Stuiver [3].

From $^{14}$C dating, the calendar age of the Choukai Jindai cedar in Japan (39°N) was ranged in from 2757 to 2437 yr BP (807 to 487 yr BC) [2]. As shown in Figure 1, this wood sample is located at a very interesting era because the variation of $^{14}$C concentrations shows a peak area as well as Maunder Minimum [3]. The Maunder Minimum (1645-1715 AD) is considered as a period of very weak solar activity, because the sunspot number was absent at the duration. This period seems to coincide with the so-called Little Ice Age, that is,
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a period of cold climate in around the 17th century.

According to IntCal04 which is the standard internationally recommended radiocarbon calibration data set, a rapid change of the $^{14}$C concentrations is seen in 50 years in 2650 to 2600 yr BP by calendar year. In order to investigate the structure in detail, we have measured the $^{14}$C concentrations of single-yr tree rings using the Choukai Jindai cedar during the 50 years, because the data set of IntCal04 was for 5-year span. The $^{14}$C concentrations have been measured with the highly accurate liquid scintillation counting system with 0.2% accuracy. We report the structure of the $^{14}$C variation in the 50 years.

**Experiment**

The Choukai Jindai cedar was buried by the eruption of Mt. Choukai in Japan ca. 2500 years ago. The old tree rings as a photograph shown in Fig.1 have been used in this study. It had about 320 tree rings, each 0.5 to 3.0 mm wide.

The tree rings were separated with single-year intervals to measure the concentrations of $^{14}$C.

As the $\alpha$-cellulose in the cell walls of the tree rings is the most reliable chemical component of the wood for measuring the annual concentration of $^{14}$C, it was chemically extracted.

From the extracted-cellulose, benzene was synthesized, because benzene has many carbon atoms in its molecule and is a suitable solvent for liquid scintillation counting (LSC). The amount of synthesized benzene was typically 5.3 g and it was produced from the single-year wood sample of 70 g. The benzene was poured to a high purity Teflon/copper counting vial (7 ml). The material of the vial is selected to reduce the background counting rate.

Measurements of $^{14}$C in the synthesized benzene were carried out with a statistical accuracy of 0.2% using liquid scintillation counting system the Quantulus with an ultra-low background level. The counting rate of the old tree ring sample is approximately 40 cpm for 5.3 g of benzene and the background rate is typically 0.08 cpm for 1 g dead benzene sample without $^{14}$C. The systematic error was less than 0.1% in total.

![Image](image-url)

Figure 2: Comparison between $\Delta^{14}$C of the tree rings and of the IntCal04 data. The symbols indicate: black open circles for IntCal04 data set, red solid circles for Choukai single-yr data set, blue solid squares for Choukai 10-yr data set, cyan solid diamonds for Choukai 5-yr data set, green solid triangles for Choukai 5-yr data set using AMS.

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The $^{14}$C dating of the tree rings was carried out by a wiggle matching for the IntCal04 calibration data using OxCal (ver.3.10) [4]. The calendar age of the outer edge of tree rings is 2427.5 yr BP ± 12.5 with the confidence level of 95.4%. Therefore, the Choukai tree rings are ranging from 2757 yr BP to 2437 yr BP.

Figure 2 plots the $^{14}$C concentrations of the Choukai samples compared with the IntCal04 calibration curve. In the figure, four kinds of data sets are shown for the IntCal04 data set of black open circles. Choukai data sets are green solid triangles for a 5-yr data set measured by the accelerator mass spectrometry (AMS), the blue solid squares for a 10-yr data set measured by the LSC and cyan solid triangles for 5-yr data set measured by the LSC. The red solid circles are a single-yr data set measured by the LSC. In the dataset of IntCal04 between 2650 yr BP and 2625 yr BP the $^{14}$C concentrations decreased with approximately 13% for 25 years and they increased with 6.7‰ for 20 years. However, in the Choukai data set for the duration, the $^{14}$C concentrations decreased with 11‰ for 7 years, and then they increased with 4.7‰ for 30 years.

The decreasing rate of the Choukai data set from 2650 yr BP is greater than that of the IntCal04. For this discrepancy, we consider two possibilities. One is a kind of smoothing effect for the IntCal04 data set, because the data set is constructed using a random walk model for the data sets of 10-yr and/or 20-yr span [5]. The other one is an intimate difference between both the data sets due to regional effects, because the IntCal04 data set is based on the measurements using Irish oak (55°N) and German oak (50°N) and the Choukai data set is from Japan cedar (39°N). However, as the rapid change of the $^{14}$C concentrations occurs for both the data sets, it is no doubt that the change is global phenomena. Therefore, it is important to measure the variations with high time resolution such as single-year, because it is essential to that an 11-yr variation when we judge that the change is caused by a solar effect or by a variation of the earth environment.

**Conclusion**

We have been measuring the concentration of $^{14}$C in a Choukai Jindai cedar ca. 2500 years ago in Japan to investigate a time variability of the solar activity at ancient time. The $^{14}$C concentrations of single-yr tree rings have been measured with the highly accurate liquid scintillation counting system with 0.2% accuracy focusing on 11-yr periodic cycle. The measured $^{14}$C concentrations showed the decrease rate is 11‰ for 7 years from 2650 yr BP, although the rate of the IntCal04 data is 13‰ for 25 years.

**References**


