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High Accurate C-14 Measurement of an Old Tree Rings in the Past 2500 years

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Abstract

We attempted to investigate prceisely the time variations such as the periodicities of solar activity in the past older than 2000 years by ^{14}C measurements of old tree rings. The sample wood is a cedar buried by a volcanic eruption at Mt. Choukai in Japan about 2500 years ago. Two large amounts of about 10 g benzen were synthesized for two tree rings with the time difference of 170 years. From the simple extraporation by the life time of ^{14}C, the radiocarbon ages of the tree were calculated as 2563BP and 2510BP. On the other hands, using the decadal calibration data the duration of 900-600BC were estimated as the calendar dates of the tree ring, which is the duration between the Maunder- and the Spöler type.

1 Introduction:

Measurement of ^{14}C concentration in annual rings of a tree is a powerful method to study solar activity, variations of geomagnetic field, and to explore explosions of supernovae in the past. Because a cosmogenic isotope ^{14}C produced by cosmic rays in atmosphere is respired as carbondioxcide and is fixed as chemical material of the carbon in a tree, the tree rings annually record information of the cosmic ray flux corresponding to the year. Also, the record is memorized in the tree rings for a long time because the half life of ^{14}C is 5730 yr. In fact, using the bi-decadal Δ^{14}C records in wood over 9600-year, Stuiver and Braziunas (1989) presented the modulation of cosmic ray flux due to Sun with the periodicities of 420, 200, and 140 year and that the interval 880-200BC is the Maunder- and the Spörer type. Also, Kocharov et al. (1995) reported that the variations of the ^{14}C content in the tree rings over the last several years is related with the 11-year periodicity of solar activity. In particular, data of 22- year periodicity at Maunder Minimum, if it is comfirned, is useful to study dynamo models during deep minima of solar activity. Therefore, we can expect that the ^{14}C content in an old tree ring buried ca. 2500 years ago recorded the solar activities in the duration of the Maunder- and the Spöler type. Under the consideration, we planed to investigate preceisely the time variations of the ^{14}C concentration in atmosphere at the past time focussed on periodicities of solar activity such as 11-year cycle, measuring the concentration of ^{14}C in the old tree rings by a single year.

Because the amplitude of Δ^{14}C with 11-year cycle in 18-19 century were 0.14% and 0.48% for Pacific trees and Russian trees, respectively (Stuiver and Braziunas 1993, Kocharov et al, 1995), the variation of the ^{14}C content in the old tree rings due to 11-year solar cycle would be also small. Hence the ^{14}C measurement with high accuracy $<$0.2% is necessary for the single-year tree rings. To find the small variations in the measurements, careful pretreatments for a large amount of sampleare are important (Olsson and Possnert 1992) such as the separation of tree rings, the extraction of α-cellulose and the synthesis of benzene. For the benzene sample the ^{14}C concentration is measured with ultra-low background level, employing a liquid scintillation counting system Quantulus 1220^{TM}

First of all, we have measured the ^{14}C concentrations for two distant rings of the old tree ring to ensure the accuracy for single-year measurement of the ^{14}C measuring system. One is the sample R100 for the ring number of100 counted from the center of the tree ring. The other is the sample R270 for the outside part of the tree ring. For the two samples there was the difference of 170 years from the calculation of the ring number. Moreover, the estimation of the year when this sample tree was alive, is important to confirm that the age of the sample wood was during the second Maunder and Spörer year. Therefore, from the two radiocarbon ages

and the time difference between two rings we have invesigated the calendar age of the wood, compared with a published decadal calibration data (Stuiver and Becker, 1993). We describe the results obtained by the high accurate ^{14}C measurements.

2 Methods:

The wood sample is a cedar tree ring buried before ca. 2500 years by a volcanic eruption of Mt. Choukai (140° 03'E, 39° 05'N) which is ∼10 km distant from the Japanese Sea. The cedar tree dug out at spring 1996 is ≈2 m in diameter and ≈1.5 m in length. As the cedar tree was in the clay mud flow, it was in good preservation. It had about 300 tree-rings with the widths of ≈0.5-3mm, and many clear barks were left in the outside.

To obtain wood samples for each single-year tree ring, the wood block was boiled in a pressurised pot at a pressure of 2.0 atm, and then the samples were peeled for each tree ring using a thin cutting-knife and a fine tweezers. After drying the peeled pieces, they were grinded to < 0.5 mm with a Wiley Cutting Mill WT-50^{TM} to carry out a chemical pretreatment.

To remove oils, saps, and resins in the wood powder sample, they were distillated in hexane and then in ethanol with use of Soxhlet for ca. 2 days. For α-cellulose extraction, we used a chemical cooking method employed in Quaternary Isotope Laboratory, University of Washington. It consists of two steps by lignin removal and α-cellulose separation. ≈33 g of the cellulose was extracted from ≈110 g of the wood powder sample to obtain a large amount of benzene 10 g.

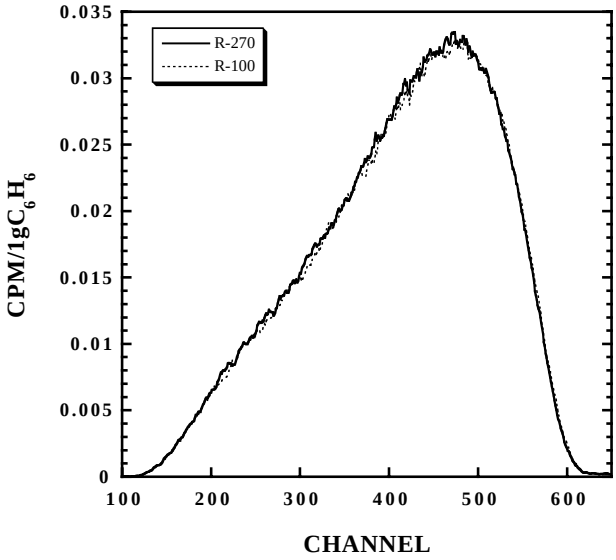

Figure 1: The pulse height distributions for two samples of R100 and R270. They clearly represent the β rays spectrum from the ^{14}C.

In benzene synthesis, the α-cellulose is at first burnt in oxygen at pressure of 20 atm with a Parr 1121^{TM} Oxygen Conbustion Bomb to produce CO_2. Then, from the CO_2, benzene is synthesized via Li_2C_2 and C_2H_2 with a benzene synthesizer composed of glass parts by greaseless joints. The synthesis efficiency from the CO_2 to the benzene was typically 83%. The amounts of each benzene were 10.5225 g, 10.5843 g for R100, R270, respectively. A modern standard benzene called STD was synthesized from Oxalic Acid (SRM 4990-C) with the standard concentration of ^{14}C in A.D.1950 certified by NIST. For the measurement of background, a dead benzene without ^{14}C called DB, which is commercially manufactured, was used. The amounts of benzene were 10.5222 g and 10.5025 g for STD and DB, respectively. We carefully adjusted the amounts of benzene for four kinds of samples within 80 mg.

The synthesized benzene was put in a high purity teflon/copper counting vial (20 ml) and butyle-PBD was added as the scintillator. The vial was mounted inside the counter Quantulus to measure the β rays spectrum from the ^{14}C in the benzene. For one run of the measurement, we measured R100, R270, STD, and DB by turns with a cycle of 45 minutes for $\sim$ 3 days to realize the statistical error less than 0.3%. The cyclic measurements are useful to ensure uniformity of the conditions for time variations of the background rate.

3 Results and Discussion:

Figure 1 shows the pulse height distributions for R100 and R270. Note that the counting rate for R270

is slightly greater than that for R100. Both the distributions clearly represent the β rays spectrum from the ^{14}C indicating high purity of the synthesized benzene samples (Suzuki, Sakurai et al, 1999). Moreover, the spectrum are very similar in the shape and the difference of the maximum channels between the two samples was less than approximately 1 channel. It indicates that the scintillation efficiencies in benzene are very similar between two benzene samples. Hence we confirmed the sample treatment and the benzene synthesis were conducted stably in the operations.

To investigate the stability of the scintillation counting system Quantulus, four runs for the same samples were carried out by the same measuring conditions except the date of the measurements. The duration between the first run and the fourth run was approximately 1 month. As shown in figure 2, the counting rates for R100 and R270 normalized per 1 g benzene were constant for four runs. The standard deviations for the average counting rates of four runs were 0.13% and 0.09% for R100 and R270, respectively. It indicates the Quantulus is very stable in the ^{14}C measurement.

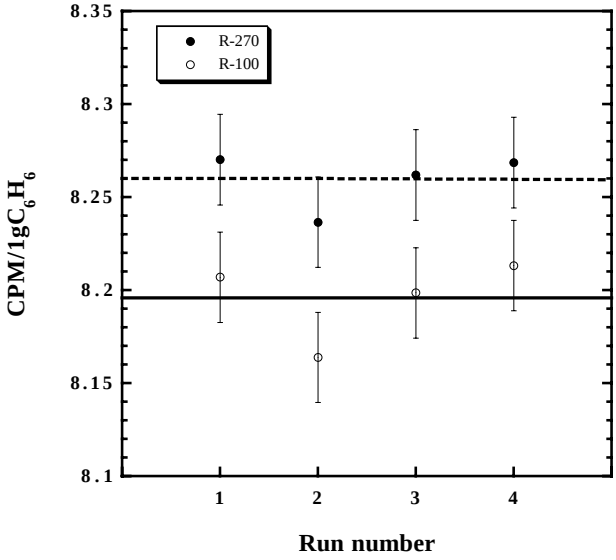

Figure 2: The counting rates of R100 and R270 as a function of run number of the measurement. They indicate good stability of the the ^{14}C measuring system Quantulus.

From four measurements, the counting rates for R100 and R270 were obtained with high accuracy of 0.15% as the statistical errors taking account of background rate of 0.095 cpm/g benzene. As shown in figure 3 the counting rate of R270 was 0.77% greater than that of R100. When the concentrations of ^{14}C in the atmosphere were same for R100 and R270, the counting rate of R270 would be 2.05% greater than that of R100 taking account of the half life of ^{14}C and the time difference of 170 year between R100 and R270. These imply that the ^{14}C concentration in the atmosphere at R270 would be about 1.3% less than that at R100.

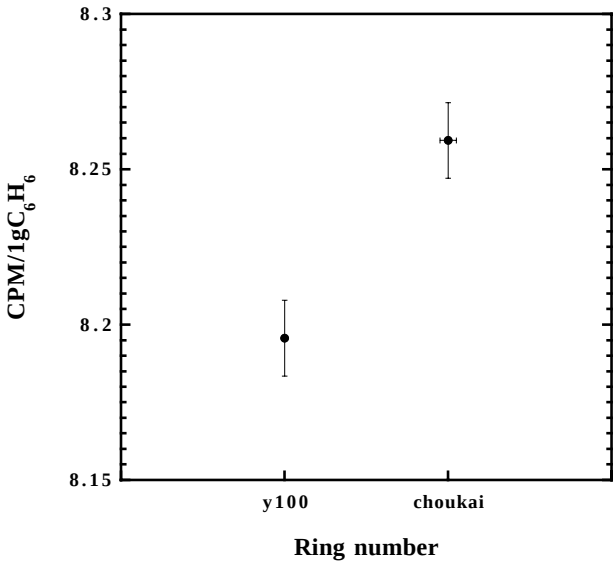

Figure 3: High accurate counting rates of R100 and R270 with the statistical error of 0.15%. The difference between the counting rates was 0.77% for the time difference of 170 years.

We have calculated radiocarbon ages for R100 and R270, respectively, using the counting rate of 15 cpm/1 g benzene for STD. They were 2563$\pm$17year BP and 2510$\pm$15 year BP for R100 and R270, respectively. The radiocarbon ages and the decadal calibration data (Stuiber and Becker, 1993) are shown in figure 4. The calendar year corresponding to R100 is during 805 - 770BC Corresponding to R270 there are two candidates around 765BC and 600BC. Considering the time difference of 170 year between both the rings the first candidate is around 600BC. Hence we roughly estimate the ages of this old tree ring during 900-500BC.

4 Conclusion:

By measuring the time variations of the ^{14}C content in old tree rings by a single year, we attempted to inves-

tigate preceisely the time variations of the ^{14}C concentration in atmosphere at the past time of 2500 years, focussed on periodicities of solar activity such as 11-year cycle. The sample tree ring is a cedar tree buried before 2500 years by a volcanic eruption of Mt. Choukai in Japan. The cedar tree dug out is ca. 2 m in diameter and ca. 1.5 m in length with about 300 tree-rings.
Two large amounts of benzen about 10 g were synthesized for two tree rings with the time difference of 170 years. The concentration of the ^{14}C for the samples were measured with the high accuracies of 0.15% by the liquid scintillation counter Quantulus. The difference of the counting rates was about 0.8% between two samples. It indicates that the concentration of the ^{14}C in the atmosphere was not constant at those dates, because the radiocarbon ages were calculated as 2563BP and 2510BP. Using decadal calibration data we estimated that the calendar dates of the tree ring were during 900-600BC similar to the Maunder- and the Spöler type.

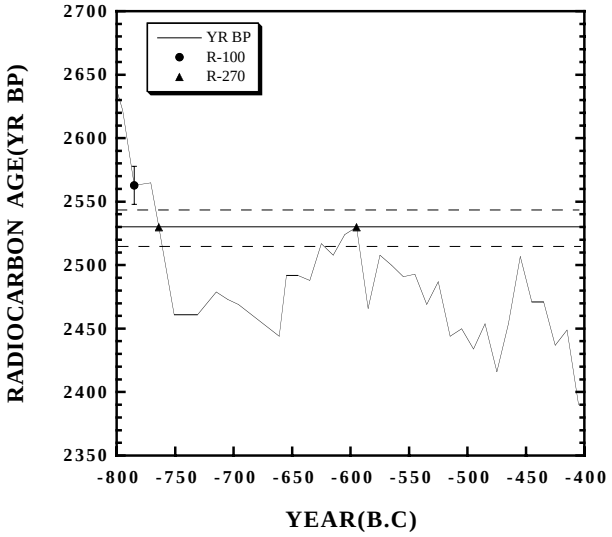

Figure 4: Calculated radiocarbon ages of R100 and R270 shown on the decadal calibration data as a function of the calender dates. The ages of the tree ring was estimated the duration 900-600BC.

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