

# **Radiocarbon Dating with Tandetron Accelerator Mass Spectrometer at Nagoya University**

## **-An Introduction to Mechanical and Electrical Details-**

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A Tandetron accelerator mass spectrometer (AMS), a machine dedicated to radiocarbon ( $^{14}\text{C}$ ) measurements, constructed by General Ionex Corporation, USA, has been used since 1983 to measure  $^{14}\text{C}$  concentrations of environmental samples, as well as  $^{14}\text{C}$  dates of geological and archeological materials at the Dating and Materials Research Center, Nagoya University.

The natural abundance of radioactive  $^{14}\text{C}$  in the ratio to stable carbon  $^{12}\text{C}$  ( $^{13}\text{C}$ ) is  $1.2 \times 10^{-12}$  ( $1.2 \times 10^{-10}$ ). Details of how the low-abundant  $^{14}\text{C}$  atoms are separated from  $^{12}\text{C}$  and  $^{13}\text{C}$  atoms with AMS are discussed here. Mainly described are the mechanical and electrical designs of (1) production of negative carbon ions with a Cs sputter ion source, (2) ion-mass selection by magnetic field, (3) charge exchange at the accelerator terminal, (4) ion-energy selection with electrostatic deflector, (5)  $^{14}\text{C}$  identification from background particles by a heavy ion detector.

**Radiocarbon dates of total organic carbon of  
the BDP 100-meter-long cores (BDP-93 Hole 1 and 2) from  
Lake Baikal measured with a Tandetron AMS  
(Preliminary Results)**

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## **1. Introduction**

Lake Baikal, locating in the eastern Siberia (104-110 E, 51-56 N), is one of the largest lakes in the world, being in the shape of crescent moon with west-east width of about 50 km and a north-south length of 639 km. The lake is formed on a rift valley system similar to the huge rift valley in Africa, and is considered to be expanding its width by several cm or several-ten cm in the west-east direction. Lake Baikal, being the deepest lake in the world with a maximum depth of 1643 m, has accumulated more than 5,000 meter thick sediments, which preserve many kinds of information concerning local as well as global changes of paleo environment. The period of sedimentation covered by the lake sediments is considered as old as about three billion years before present. A project collecting the full-length sediment of Lake Baikal has been planned to investigate the paleoclimatic history and tectonic evolution of the Lake Baikal sedimentary basin in the Late Neogene, as the Baikal Drilling Project (BDP) started a few years ago.

As a first step of the big BDP plan, two boring cores of 100-meter length each were collected from the Buguldeika saddle in the

southern basin of Baikal in March 1993. Eleven and thirteen radiocarbon ( $^{14}\text{C}$ ) dates are measured for organic carbon separated from the upper portion of the cored sediments hole 1 and hole 2, respectively.

## **2. Location of boring site**

The Lake Baikal is normally divided into three basins; southern, central and northern ones.

The maximum water depth of the southern basin is 1410 m. The southern basin has Angara River which is the only one flowing out from Lake Baikal. Selenga River, the biggest river flows in Lake Baikal, carries a big amount of sedimentary particles into the lake. Owing to rather high deposition rate of such particles, a shallow bump is produced in the lake and this separates southern and central basins. The maximum water depth of 1643 m locates in the central basin. An Olhon Island and Academic Ridge of a shallow bump with several hundred meter deep running straight from north-east to south-west separates the central and northern basin. The northern basin is rather shallow (600-700m) with the maximum depth of 900m), compared to other two basins (1200-1600m).

The sediments were cored over a topographic high called the Buguldeika saddle in the southern basin of Lake Baikal, locating at the opposite side from Selenga River mouth (Fig.1). Water depth of the site is 354 m. Two cores of 98 m (BDP93 hole 1) and 102 m (BDP93 hole 2) long, neighbouring about several meters, were collected continuously in 78 mm diameter plastic liners (BDP93, 1993). The BDP93 cores as well as other shorter sediment cores collected previously (Ogura *et al.*, 1992; Peck *et al.*, 1994) are now under geological, chemical and physical analyses for many respects.

## **3. Experimental procedures**

Radiocarbon dating was conducted for sediment samples from 11 and 13 horizons at the depths from 5.5 to 1,400 cm of the cored sediment hole 1 and hole 2. A few grams of each sediment sample was treated with 1.2 N HCl for 2 hours at 80 C, rinsed with distilled water and dried. Organic carbon and nitrogen contents, given in weight ratio to the dried sediment, are measured for a several

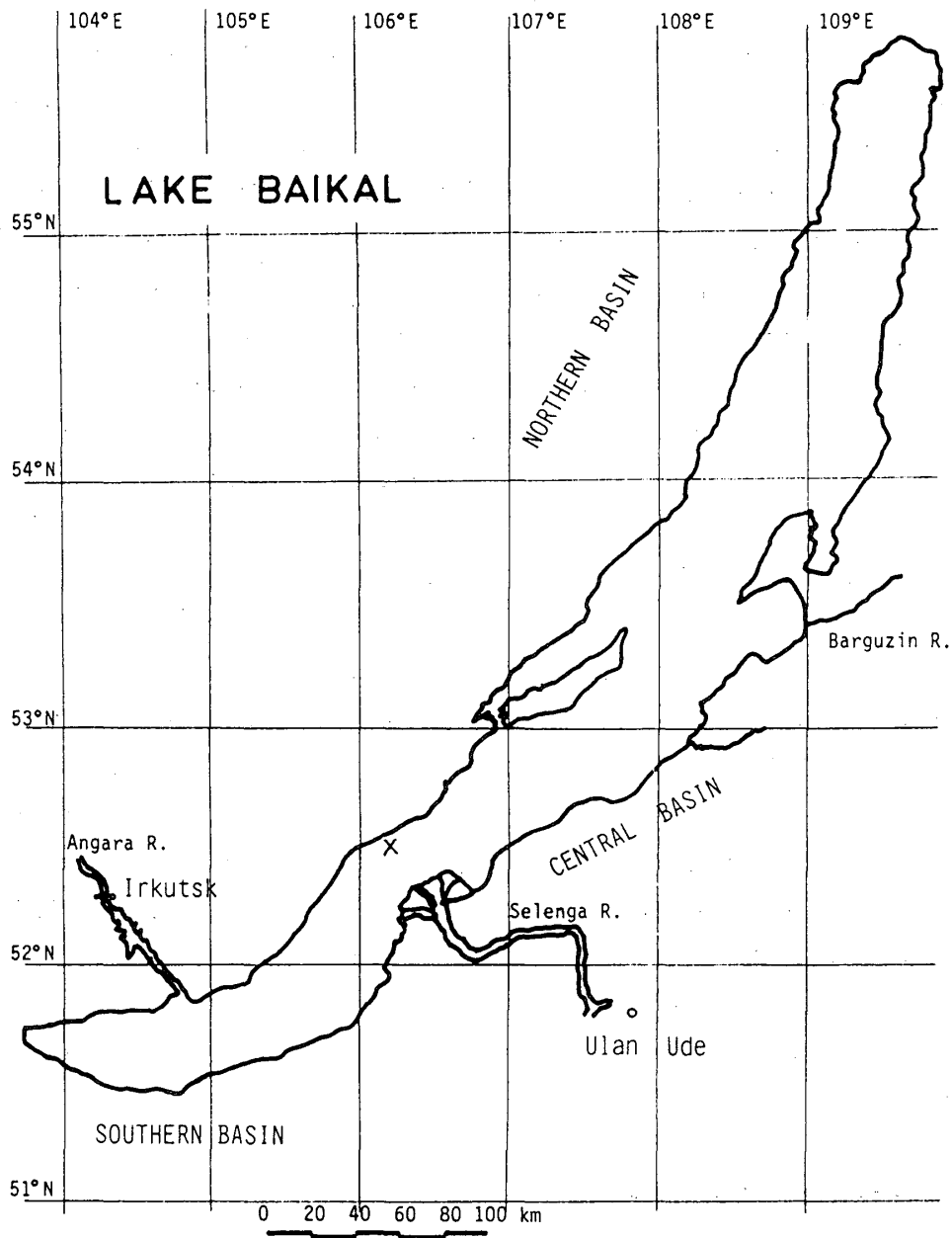


Fig. 1 Schematic map of BDP93 drilling site(X).

hundred mg of the acid-pretreated sediment samples, using a C-N coder( MT-700, Yanaco Ltd., Japan). Carbon dioxide of about 2 mg carbon was produced from total organic carbon in sediment samples, by heating a relevant amount of the sediment samples at 950 C in a sealed Vycor tube with about 500 mg of CuO as an oxidizer, and purified cryogenically using liquid nitrogen and an ethanol and liquid-nitrogen mixture in a vacuum line. About half of the carbon dioxide thus produced was then converted to graphite on iron powder (1.5mg) catalyst, by reducing CO<sub>2</sub> with hydrogen in a Vycor tube at 650 C for 4-6 hours. Details of the graphitization procedures are described by Kitagawa, *et al* (1993).

The <sup>14</sup>C/<sup>13</sup>C ratio of sample graphite was measured with a Tandetron accelerator mass spectrometer (AMS) at Nagoya University (Nakamura *et al.*, 1985). As a <sup>14</sup>C standard, NBS oxalic acid (SRM-4990) was used. The obtained <sup>14</sup>C/<sup>13</sup>C ratio was converted to <sup>14</sup>C age by using the Libby's half life of <sup>14</sup>C of 5,570 years. The <sup>14</sup>C age is given in years BP, and 0 years BP is defined as AD 1950.

Residual of the carbon dioxide extracted from sediment samples was analyzed for stable carbon isotope ratio, <sup>13</sup>C/<sup>12</sup>C, using a Finnigan Mat 252 triple collector mass spectrometer, which was described as δ<sup>13</sup>C<sub>PDB</sub>.

#### 4. Results and discussion

Values of organic C and N contents, C/N ratio, δ<sup>13</sup>C<sub>PDB</sub>, <sup>14</sup>C ages are given in Table 1 and 2 for only upper portion of the cored sediment hole 1 (from surface to 1200 cm) and hole 2 (from surface to 665 cm), respectively. Errors quoted for <sup>14</sup>C ages are one sigma ones.

##### 4.1. δ<sup>13</sup>C<sub>PDB</sub> values and <sup>14</sup>C ages of organic fractions

<sup>14</sup>C ages of organic fractions extracted from cored sediments of BDP93 hole 1 and 2 are shown in Fig.2. δ<sup>13</sup>C<sub>PDB</sub> values were measured only for the BDP93 hole 2 sediments, as shown in Fig.3. The δ<sup>13</sup>C<sub>PDB</sub> values, ranged from -25‰ to -30‰, were used for the correction of carbon isotopic fractionation to get the more precise <sup>14</sup>C ages (Nakamura, *et al.*, 1994). The correction factors were, however, as small as 30 years at maximum and almost negligible compared with the error of <sup>14</sup>C ages in the present study. The ages

Table 1  
 Summary of carbon isotopic analysis for organic materials in the sediment core BDP93 hole 1

| Sample #   | Depth<br>(cm) | C cont.<br>(%) | N cont.<br>(%) | C/N ratio | $\delta^{13}\text{C}$<br>(‰) | $^{14}\text{C}$ dates<br>(y.BP) | Error<br>( $\pm 1 \sigma$ ) | Code #<br>(NUTA-) |
|------------|---------------|----------------|----------------|-----------|------------------------------|---------------------------------|-----------------------------|-------------------|
| BDP93-1-1  | 5-6           | 2.13           | 0.23           | 9.20      | -                            | 1940                            | 90                          | 2775              |
| BDP93-1-2  | 44-45         | 2.06           | 0.23           | 8.94      | -                            | 2420                            | 190                         | 2664              |
| BDP93-1-3  | 119-120       | 3.47           | 0.31           | 11.10     | -                            | 5710                            | 180                         | 2665              |
| BDP93-1-4  | 224-225       | 1.61           | 0.17           | 9.38      | -                            | 11510                           | 130                         | 2666              |
| BDP93-1-5  | 295-296       | 0.45           | 0.06           | 7.48      | -                            | 17170                           | 140                         | 2673              |
| BDP93-1-6  | 417           | 0.98           | 0.12           | 7.97      | -                            | 21280                           | 140                         | 2776              |
| BDP93-1-7  | 483           | 0.86           | 0.10           | 8.96      | -                            | 22320                           | 190                         | 2674              |
| BDP93-1-8  | 532           | 0.77           | 0.09           | 8.20      | -                            | 24680                           | 200                         | 2791              |
| BDP93-1-9  | 779-780       | 0.88           | 0.10           | 8.80      | -                            | 25710                           | 240                         | 2675              |
| BDP93-1-10 | 1065          | 0.56           | 0.07           | 7.73      | -                            | 27310                           | 240                         | 2790              |
| BDP93-1-11 | 1203          | 0.95           | 0.10           | 9.60      | -                            | 30610                           | 240                         | 2777              |

Table 2 Summary of carbon isotopic analysis for organic materials in the sediment core BDP93 hole 2

| Sample #    | Depth (cm)  | C cont. (%) | N cont. (%) | C/N Ratio | $\delta^{13}\text{C}$ (‰) | 14C dates (Y.BP) | Error ( $\pm 1\sigma$ ) | Code # (NUTA-) |
|-------------|-------------|-------------|-------------|-----------|---------------------------|------------------|-------------------------|----------------|
| BDP93-2-2   | 15.5-16.5   | 1.36        | 0.15        | 8.82      | -26.17                    | -                | -                       | -              |
| BDP93-2-4   | 45.0-46.7   | 1.94        | 0.20        | 9.60      | -26.10                    | 3800             | 70                      | 3297           |
| BDP93-2-4'  | 45.0-46.7   |             |             |           | -26.11                    |                  |                         |                |
| BDP93-2-6   | 74.5-76.5   | 3.06        | 0.29        | 10.48     | -27.84                    | 4370             | 70                      | 3298           |
| BDP93-2-8   | 101.0-103.2 | 3.06        | 0.28        | 11.04     | -28.51                    | -                | -                       | -              |
| BDP93-2-8'  | 101.0-103.2 | 3.07        | 0.28        | 11.13     |                           |                  |                         |                |
| BDP93-2-10  | 189.8-191.0 | 2.29        | 0.21        | 11.16     | -29.38                    | 10900            | 80                      | 3307           |
| BDP93-2-12  | 221.8-223.5 | 1.27        | 0.13        | 9.47      | -29.61                    | 13670            | 100                     | 3308           |
| BDP93-2-14  | 254.1-256.1 | 0.73        | 0.09        | 8.24      | -27.93                    | 15500            | 100                     | 3309           |
| BDP93-2-14' | 254.1-256.1 |             |             |           | -27.92                    |                  |                         |                |
| BDP93-2-16  | 284.9-286.6 | 0.35        | 0.05        | 6.69      | -25.24                    | 18950            | 100                     | 3319           |
| BDP93-2-18  | 316.4-318.4 | 0.45        | 0.06        | 6.96      | -25.04                    | 19780            | 190                     | 3302           |
| BDP93-2-18' | 316.4-318.4 |             |             |           |                           | 20550            | 130                     | 3303           |
| BDP93-2-20  | 362.3-363.8 | 0.59        | 0.08        | 7.28      | -27.12                    | 21440            | 130                     | 3320           |
| BDP93-2-22  | 391.7-393.6 |             |             |           |                           |                  |                         |                |
| BDP93-2-24  | 424.5-426.2 | 0.59        | 0.08        | 7.70      | -26.06                    | 25780            | 170                     | 3304           |
| BDP93-2-24' | 424.5-426.2 | 0.61        | 0.08        | 7.59      |                           |                  |                         |                |
| BDP93-2-26  | 456.0-457.9 | 0.70        | 0.09        | 8.21      | -26.79                    | 26600            | 170                     | 3321           |
| BDP93-2-28  | 489.4-491.0 | 0.79        | 0.10        | 8.35      | -26.28                    | 28200            | 210                     | 3305           |
| BDP93-2-30  | 545.7-547.3 | 1.01        | 0.11        | 9.29      | -26.21                    | -                | -                       | -              |
| BDP93-2-32  | 603.0-605.0 | 0.77        | 0.10        | 8.12      | -26.26                    | 36360            | 360                     | 3669           |
| BDP93-2-32' | 603.0-605.0 | 0.82        | 0.10        | 8.19      |                           |                  |                         |                |
| BDP93-2-34  | 663.3-665.4 | 0.49        | 0.07        | 6.63      | -25.53                    | 38710            | 350                     | 3670           |

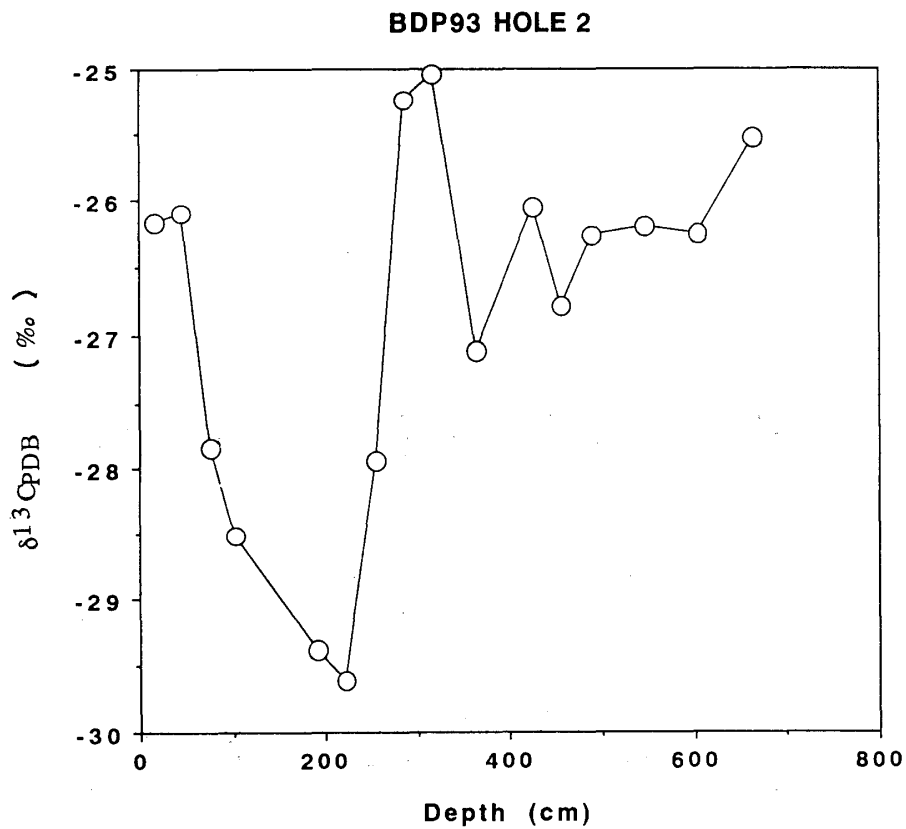


Fig.3.  $\delta^{13}C_{pDB}$  values vs. depths for organic fractions from sediments of BDP93 hole 2.

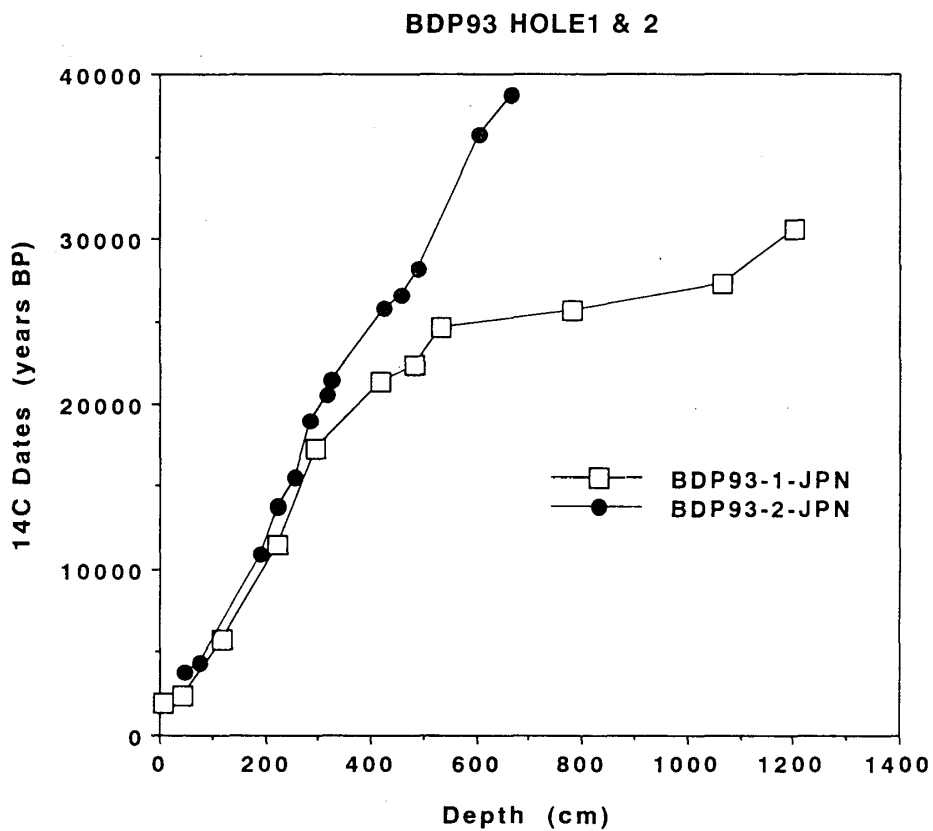


Fig. 2.  $^{14}C$  Ages vs. depths for organic fractions from sediments of BDP93 hole 1 and 2.



of the two cores are almost consistent for the upper portion of the sediments from surface to about 300 cm deep. They show different trends, however, at depths greater than 300 cm. The age-depth relation is expressed very nicely with a straight line for the hole 2 core. A least squares fit provided the age-depth relation represented by

$$Y = 57.7 X + 813, \quad (1)$$

where, Y is the  $^{14}\text{C}$  age in years BP and X is the depth of the sediment in cm. The correlation coefficient R was calculated to be 0.997. From the age-depth relation of Eq.(1), the sedimentation rate can be calculated as 0.017 cm/y for the sediments from surface to 670 cm deep for the hole 2 core. On the other hand, the  $^{14}\text{C}$  ages of the hole 1 core increased lesser extent than those of the hole 2 core, as the depth increased. The age-depth relation for the hole 1 core can be expressed by two straight lines, breaking at the sediment depth of around 530 cm. The sedimentation rates are obtained as 0.022 cm/y for the upper sediments, and as 0.125 cm/y (6 times larger than the upper value) for the lower sediment from 530 to 1200 cm. The sedimentation rate of 0.022 cm/y for the upper sediments of the hole 1 core is almost equal to 0.017 cm/y for the sediments of the hole 2 core.

The age-depth relation intercepts the sediment surface at an apparent age of 1270 and 810 years BP for the hole 1 and 2 cores, respectively. The constant shift of the  $^{14}\text{C}$  age by rather small values of 1270 and 810 years will not affect the discussion on the sedimentation rate discussed above. The problem of apparent non-zero  $^{14}\text{C}$  age at the sediment surface is, however, an important issue, for analyzing the carbon sources of the sediment, which will be discussed elsewhere.

It is not clear yet why the sedimentation rate is different as described above between the two cores drilled within several meters distance. The  $^{14}\text{C}$  dates for the hole 2 core may be generally accepted, because the sediments with older  $^{14}\text{C}$  ages tend to show younger  $^{14}\text{C}$  ages, owing to contamination by carbon with younger ages. The hole 1 sediments may be contaminated by dust particles or organic chemicals in the process of sediment

division or by bacterial growth during storage. The sedimentation rate of 0.017 cm/y for the hole 2 core is almost consistent with the rate of 0.019 cm/y over the past 30,000 years for the BDP hole 1 core, obtained by the USA research group of the BDP-93 Baikal drilling project (BDP, 1993).

#### 4.2. C/N ratios of organic fractions

Contents of total organic carbon (TOC) and nitrogen and C/N ratio for the hole 1 and 2 cores are shown in Figs. 4-1 and 4-2, respectively.

For the hole 2 core, organic carbon and nitrogen contents as well as C/N ratio show a clear bump-dip-bump structure, varying values of the C/N ratio from 6.7 to 11.2, depending on the depth. The upper bump with C/N ratios from 9 to 11 initiated in early Holocene, according to the  $^{14}\text{C}$  ages measured for organic materials in the sediments (Fig. 2). The dip of C/N ratio with the smallest value of 6.7 appeared at around 18,000 years BP, the date known as one of the coldest time in the last glacial period. The second bump with C/N ratio being smaller values than the upper one and ranging from 7 to 9.3, lasted from 35,000 to 25,000 years BP, that was considered as a relatively warm period.

Ishiwatari *et al.* (1992) used C/N ratios to differentiate source materials of organic fractions in the sediment of Lake Baikal. They summarized that C/N ratios of organic materials derived from lacustrine biomass (autochthonous and lacustrine origin) tend to be 6-9 by the contribution of lower organisms as algae, phyto- and zoo-plankton with rather high protein content, while those of organic materials, being supplied by higher plant (allochthonous and terrestrial origin) with low protein contents, are much higher. Ishiwatari *et al.* (1992) analyzed the core sample 323-PC1 collected from northern basin of Lake Baikal, and estimated the contributions to organic materials in the sediment from autochthonous and allochthonous fractions, based on the assumptions that organic materials whose C/N ratios ranged from 6.7 to 8.8 are predominantly autochthonous.

The lowest C/N ratio of 6.7 which appeared in the last glacial maximum period, as shown in Fig. 4-2, implies that the autochthonous fractions predominated to supply organic materials in the sediments at that time. In the warmer periods, on the

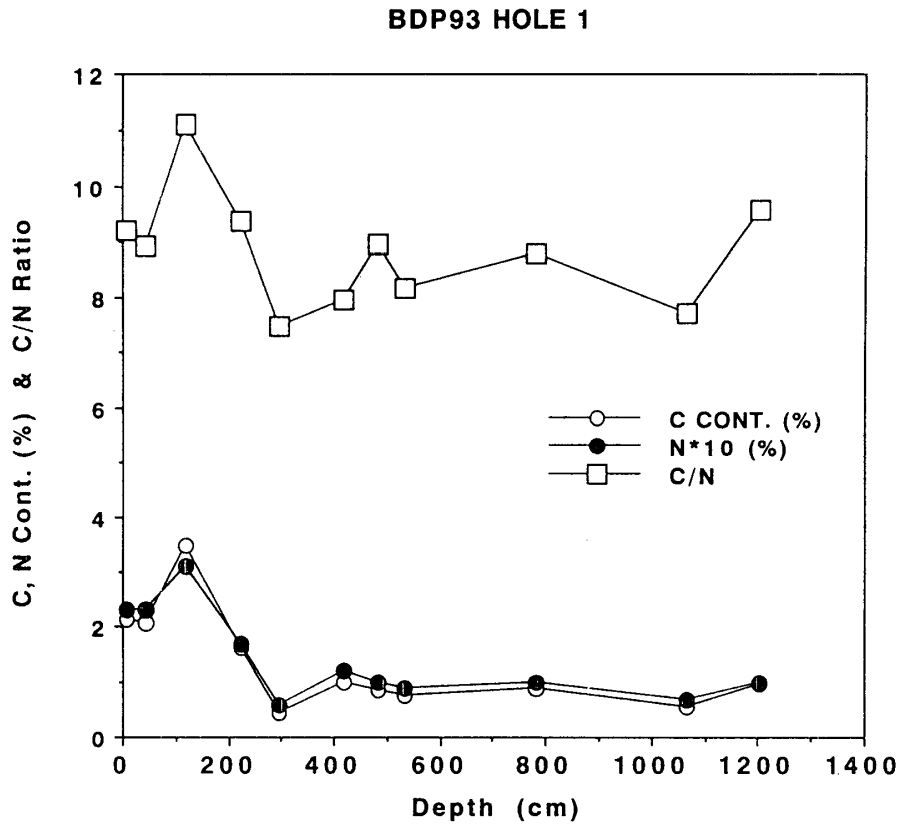


Fig.4-1. Contents of TOC and N, C/N ratios vs. depths for BDP93 hole 1 core.

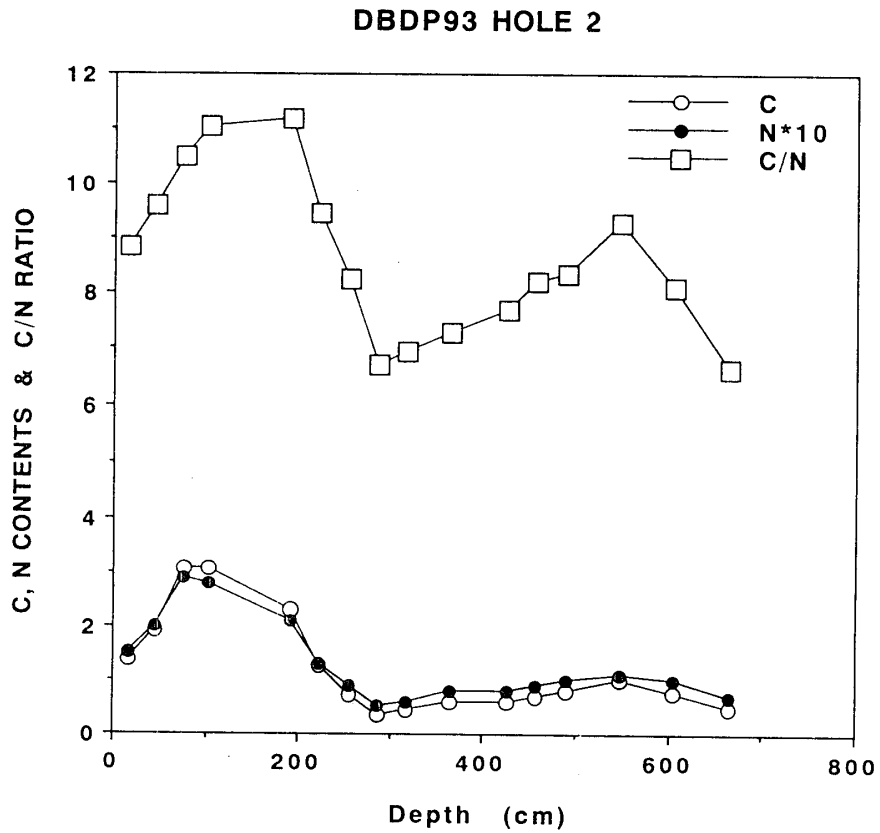


Fig.4-2. Contents of TOC and N, C/N ratios vs. depths for BDP93 hole 2 core.

contrary, allochthonous fractions increased and resulted in rather larger values of the C/N ratio. Vertical variations of the C/N ratio in the sediments assigned with accurate  $^{14}\text{C}$  ages are very important to evaluate the paleo-climatic change around Baikal after late Pleistocene. The  $^{14}\text{C}$  ages obtained for the BDP93 hole 2 core sediments can be used to build up the accurate chronology of the paleo-climatic change which is deduced by the C/N ratio of sedimentary organic materials.

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## 研究成果

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