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# Response of Biogenic Silica Production in Lake Baikal and Uranium Weathering Intensity in the Catchment Area to Global Climate Changes

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## 1. Introduction

Lake Baikal, in southeast Siberia, is a structural basin in the Baikal rift valley (Fig. 1a) and is the largest lake on earth in terms of fresh water volume (23,000 km$^3$). With a surface 454 m above sea level (asl), it covers an area of 31,500 km$^2$ (length, 636 km; maximum width, 80 km) and has a maximum depth of 1,620 m. The vegetation around the lake is characteristic of the steppe and taiga. And the annual mean temperature and rainfall around the lake are -2.2 ºC and 400–500 mm per year, respectively. The catchment area of the lake is 540,000 km$^2$, extending from northwest Mongolia to southeast Siberia (Fig. 1b), of which 83% constitutes the drainage basin of the Selenga River (Fig. 1b and c). This is the largest river flowing into the lake and its water inflow makes up 50% of the total riverine input. The climate in the Lake Baikal region is influenced by westerly wind weather systems (Mackay, 2007). Therefore, most of the atmospheric moisture in southeast Siberia is from the North Atlantic Ocean and the Arctic Ocean.

The bottom sediment of Lake Baikal documents the long-term history of environmental changes in the Asian continental interior (southeast Siberia), showing the shift in climate on various time-scales. The main proxy records obtained from the sediment are based on the concentration of diatom frustules and biogenic silica (bioSi) (Colman et al., 1995; Kashiwaya et al., 2001; Mackay, 2007; Prokopenko et al., 1999, 2001, 2002; Williams et al., 1997) and the amount of pollen fossils (Shich et al., 2007; Tarasov et al., 2005) in the sediment indicating the bioproductivity in the lake and its surrounding watersheds. The main source of the bioSi in the sediment is diatom frustules (Karabanov et al., 1998). Studies to date using the biological records revealed that the diatom and vegetation changes were correlated with the Milankovitich periods (Kashiwaya et al., 2001; Williams et al., 1997) and were in phase with the glacial-interglacial cycles (Colman et al., 1995; Prokopenko et al., 2001a, 2002; Shichi et al., 2007). Moreover, the variations are found to follow the centennial-to-millennial-scale climate changes that occurred in the North Atlantic region: Bond cooling cycles during the last glacial periods (Prokopenko et al., 2001b); a Younger Dryas cold period for the last glacial/Holocene transition (Prokopenko et al., 1999); and IRD (ice-rafted debris) cooling (Bond) events in the Holocene (Mackay, 2007; Tarasov et al., 2005).

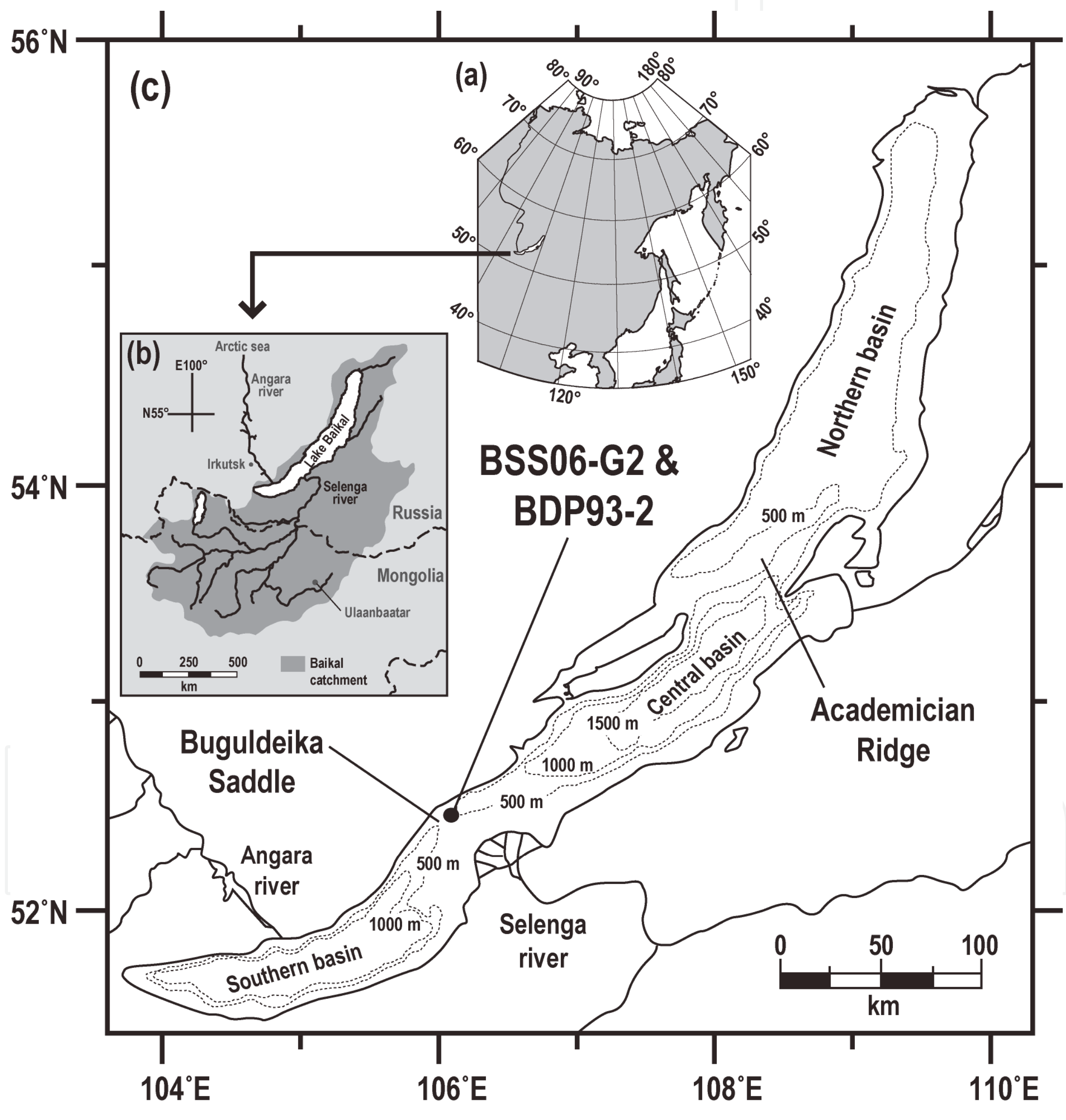


Fig. 1. Maps of (a) northeastern part of continental interior Asia and (b) catchment basin of (c) Lake Baikal. Bathymetric map of the lake showing the collection sites of cores BSS06-G2 and BDP93-2 at Buguldeika saddle.

On the other hand, the uranium record in Lake Baikal sediments as well as the biological records provides information on the environmental changes in the region. The variation in the uranium concentration is thought to be due to the weathering intensity in the Selenga drainage basin associated with changes in rainfall/moistures levels (Goldberg et al., 2010; Murakami et al., under review). The U-Th isotope study of Edgington et al. (1996, 1997) revealed that the uranium in Lake Baikal sediment is composed mainly of authigenic components, which originated in uranium-bearing rocks distributed in Mongolia and southeast Siberia, and that the uranium is transported from the source rock into the lake via the Selenga River. Based on geochemical evidence, Edgington et al. also concluded that the variation of uranium concentration in the sediment resulted from changes in the input from the Selenga River and its tributary. The uranium variations are reported to have corresponded to the Pleistocene glacial-interglacial cycles (Chebykin et al., 2007; Edgington et al., 1996; Goldberg et al., 2010) and the Holocene Bond events (Goldberg et al., 2005).

The purpose of the present study is to investigate the degree of similarity in the variations in the Lake Baikal records of bioSi and uranium and the paleoproxy records of global climate changes on a centennial-to-millennial-scale as well as a glacial-interglacial time scale. An examination of the correlation among these paleoclimate proxy datasets has been conducted for lake sediment from the underwater Academician ridge (Fig. 1) in Lake Baikal (Edgington et al., 1996), focusing on the variations on a glacial-interglacial scale. However, we analyzed the geochemical data of sediment from the Buguldeika saddle in Lake Baikal (Fig. 1). This site is located at the opposite side of the Selenga Delta, where uranium is directly transported to from the watershed of the Selenga River.

In the present study, we used the SPECMAP $\delta^{18}O$ record (Imbrie et al., 1984) and the North Atlantic IRD index (Bond et al., 1997, 2001) as a proxy of global climate change. The SPECMAP was acquired by stacking $\delta^{18}O$ data of planktonic foraminifera collected from five deep-sea cores in low- and mid-latitudes, generally reflecting the continental ice sheet volume. The IRD index represents cooling events in the North Atlantic, which occurred nine times during the Holocene period. The IRD cooling events are referred to as Bond events labeled 8-0 from the past to the present day.

## 2. Materials and methods

### 2.1 Sediment cores

In the present study, we used two cores, BDP93-2 and BSS06-G2 taken at Buguldeika saddle in Lake Baikal, southeast Siberia (Fig. 1). Buguldeika saddle is a local elevation that developed at the opposite side of the Selenga Delta, separated from the mouth of the river by a deep trough. Because of these topographic features, the sedimentation of the saddle is controlled by the supply of a fine suspended load from the Selenga River. Seismic surveys and the lithologic features of the drilled cores indicate that the upper 50 m of sediment at the drill sites consists of continuous and sub-parallel layers (BDP-93 Baikal Drilling Project Members, 1997).

Core BDP93-2 was collected in March 1993 at a water depth of 354 m (52º 31′ 3.0″N and 106º 09′ 6.01′′E) using a piston corer (BDP-93 Baikal Drilling Project Members, 1997). This core was 102 m long. We measured the chemical components of 228 sediment samples from the core. The samples were collected at intervals of 30 to 80 cm corresponding to the temporal resolution of 2–7 kyr. In the present study, we used the upper 37.2 m of the core, equivalent to the last 250 kyr for data analyses because the concentrations of the chemical components in this section show remarkable patterns on the glacial-interglacial cycles. On the other hand, core BSS06-G2 as well as -G1 was collected in August 2006 at a water depth of 360 m (52°27′27.1″N, 106°07′46.1″E) using a gravity corer. This core was 39 cm long, sliced into 39 sediment samples every 1.0 cm. Each sample from cores BDP93-2 and BSS06-G2 was freeze-dried, then powdered and homogenized with an agate mortar for chemical analyses.

### 2.2 Analyses

The concentrations of bioSi in the sediment of core BSS06-G2 were determined following the protocol of DeMaster (1981). Each 50-mg dry sediment sample was mixed in a 50-ml polypropylene centrifuge tube with 50-ml of 5% $Na_2CO_3$ solution. The mixture was then heated at 85ºC in a thermostatic bath and the 2-ml samples were collected after 3, 4, 5, and 6 hours. Each digested sample was diluted to 20-ml with distilled water for the analyses. Concentrations of the dissolved Si were analyzed using an ICP-AES (Ultima 2, HORIBA Jobin Yvon) at Gifu Univ. and a UV-Vis spectrometer (Metertik SP-830, Metertech Inc.) at Nagoya Univ. Their analytical precisions were <2% and <3%, respectively. Finally, the concentrations of the bioSi were calculated by determining the intercept of the line through the timed aliquots. With respect to core BDP93-2, we used Figure 3 of Colman et al. (1999), and read the values of concentrations of the bioSi corresponding to the core depth of our samples used in the uranium analysis.

The concentrations of uranium in the bulk-sediment of Lake Baikal were analyzed by two methods. In core BDP93-2, the uranium concentrations were determined neutron-activation instrument analysis (INAA; Koyama and Matsushita, 1980). This experiment was performed at the Nuclear Reactor, Kyoto Univ., Kyoto, Japan. Each 50-mg dry sample was packed in a double polyethylene film bag. It was then placed together with 30-µg of Co as a standard in a capsule. Neutron irradiation of the sample was performed in a pneumatic transfer tube for 50 min. After 1-week of cooling, the gamma-ray spectra were measured using a diode detector system of Ge (Li) coupled with a 4096-channel pulse-height analyzer. In core BSS06-G2, the bulk-U was quantified using a microwave-digestion-ICP-MS (HP4500, Hewlett Packard) at Nagoya Univ. based on the method of Murakami et al. (2010). The analytical precisions of the INAA and ICP-MS were <17% and <3%, respectively.

The radiocarbon ($^{14}C$) analyses of core BSS06-G2 were conducted using a Tandetron accelerator mass spectrometer (AMS, Model-4130, HVEE) at the Center for Chronological Research, Nagoya Univ, according to the procedures of Watanabe et al. (2009).

### 2.3 Sediment chronologies

For core BDP93-2, we used the chronology established in Colman et al. (1999). This is based on a combination of the $^{14}C$ ages and the paleomagnetic relative intensity correlations. The chronology of the depth interval 0–3.68 m in the core was determined by using the $^{14}C$ dates on the total organic carbon (TOC). Generally, in Lake Baikal, the topmost layer of the bottom sediment is known to give a "non-zero" TOC date. This is mainly caused by contamination by terrigenous organic matter. The apparent surface ages near the Buguldeika saddle range from 1000 to 1600 ka (Colman et al., 1996; Karabanov et al., 2004; Watanabe et al., 2009) and are evaluated to average 1.16 ka for two BDP93 cores (Colman et al., 1996). Colman et al. (1999) applied a 1.16-kyr correction to each date in core BDP93-2. Consequently, the age of the core at the depth interval of 0–3.68 m ranged from 0 to 21.0 ka. The date for the depth interval 3.68–50 m was determined from the correlation of the paleomagnetic relative intensities of the cores between BDP93 and dated marine sediment. Consequently, it ranged from 21.0 to 338 ka.

The chronology of core BSS06-G2 was determined in the present study using five AMS $^{14}C$ dates on the TOC (Fig. 2). As in core BDP93-2, the topmost layer has a $^{14}C$ age of 1.418 ka. We therefore applied a 1,418-year correction to the five $^{14}C$ dates over the entire sequence prior to calibration. The five dates were calibrated using an IntCal04 (Reimer et al. 2004). The age model for core BSS06-G2 was established assuming that the sedimentation rate was constant between the dated levels.

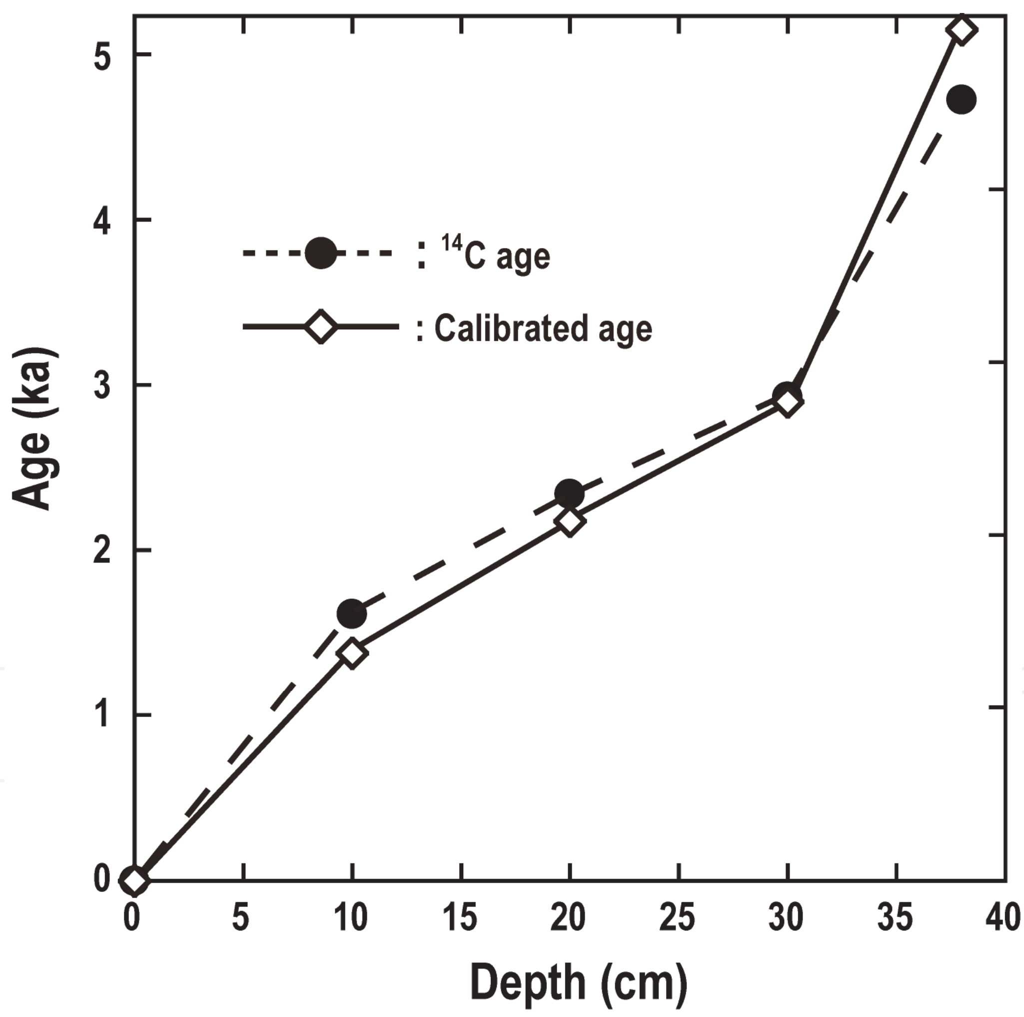


Fig. 2. Plot of age against core depth, based on 5 AMS $^{14}C$ dates for TOC in core BSS06-G2. The dates were calibrated with CALIB 5.0.1 and IntCal04 (Stuiver et al., 1998; Reimer et al., 2004).

### 2.4 Cross correlation analysis

Cross correlation analysis was used to examine the time lags between Lake Baikal core BDP93-2 and the SPECMAP $\delta^{18}O$ records. A cross-correlation analysis of such two time-series identifies the time delay that makes the two time-series most similar. The correlation coefficient quantifies the similarity of the two time-series at that time offset. The cross-correlation function for two time-series *f(t)* and *g(t)* is defined as follows:

$$\varnothing fg(\tau) = \frac{1}{T-\tau}\int_0^{T-\tau} f(t)g(t+\tau)dt$$

where $\tau$ and $T$ are the time lag and length or period of the time-series, respectively. The SPECMAP $\delta^{18}O$ data is given as a function of age in steps of equally spaced 2 kyr. However, the BDP93-2 datasets are unevenly spaced time-series with an interval of 1.9–6.8 kyr. We therefore used the average interval 2.76 kyr (standard deviation, 1.30 kyr) as a time lag for the BDP93-2 core. The correlation coefficients of cross-correlation between these two time-series were computed after linearly interpolating the neighboring two values in the SPECMAP $\delta^{18}O$, in order to adjust the SPECMAP data points to the BDP93-2 ones.

## 3. Results and discussion

### 3.1 Long- and short-term variability in the Lake Baikal records of bioSi and uranium

Figure 3 shows successive 250 kyr-profiles of bioSi and uranium concentration in core BDP93-2, SPECMAP $\delta^{18}O$, and the 65ºN June insolation. The concentration of bioSi and uranium in core BDP93-2 increased during the interglacial periods (bioSi, 5.0–45.0%; uranium, 2.5–20.0 ppm), and declined during the glacial cold periods (bioSi, 3.0–10.0%; uranium, 2.5–7.0 ppm). With respect to the variations during MIS 5 and 7, the uranium record has peaks corresponding to the all the substages (Fig. 3b), whereas the bioSi record does not show peaks corresponding to MIS 7a and b (Fig. 3a). A comparison of the BDP93-2 (Fig. 3a and b) and SPECMAP (Fig. 3c) records indicates that the uranium record shows a pattern similar to the SPECMAP $\delta^{18}O$ curve. Especially, the uranium variation during MIS 5 displays a gradual decrease (that is, sawtooth pattern) representative of the late Pleistocene glacial/interglacial cycle observed in the $\delta D$ record from the Antarctic ice core (EPICA, 2004; Petit et al., 1999) and the SPECMAP $\delta^{18}O$ record.

The relationships between the SPECMAP $\delta^{18}O$ and BDP93-2 records (Fig. 4) show a lower correlation coefficient for the bioSi ($R$ = 0.37) than for the uranium ($R$ = 0.46). These results are consistent with the visual inspections of the variations in the records (Fig. 3). On the other hand, the cross-correlation coefficients of the bioSi are 0.37~0.50 and those of the uranium are 0.46~0.52 at a time-lag range of 0 to 5.52 kyr (Fig. 5). In addition, the bioSi and uranium have a time-lag of 2.76 kyr and 5.52 kyr, respectively, but this difference is not significant because the standard deviation of the time-lag is 1.30 kyr. The results of these cross-correlation analyses indicate that there are no statistically significant differences between the Baikal bioSi and uranium records (Fig. 5); therefore we may regard the bioSi and uranium variations as paleoproxy records having the same degree of response to global climate changes.

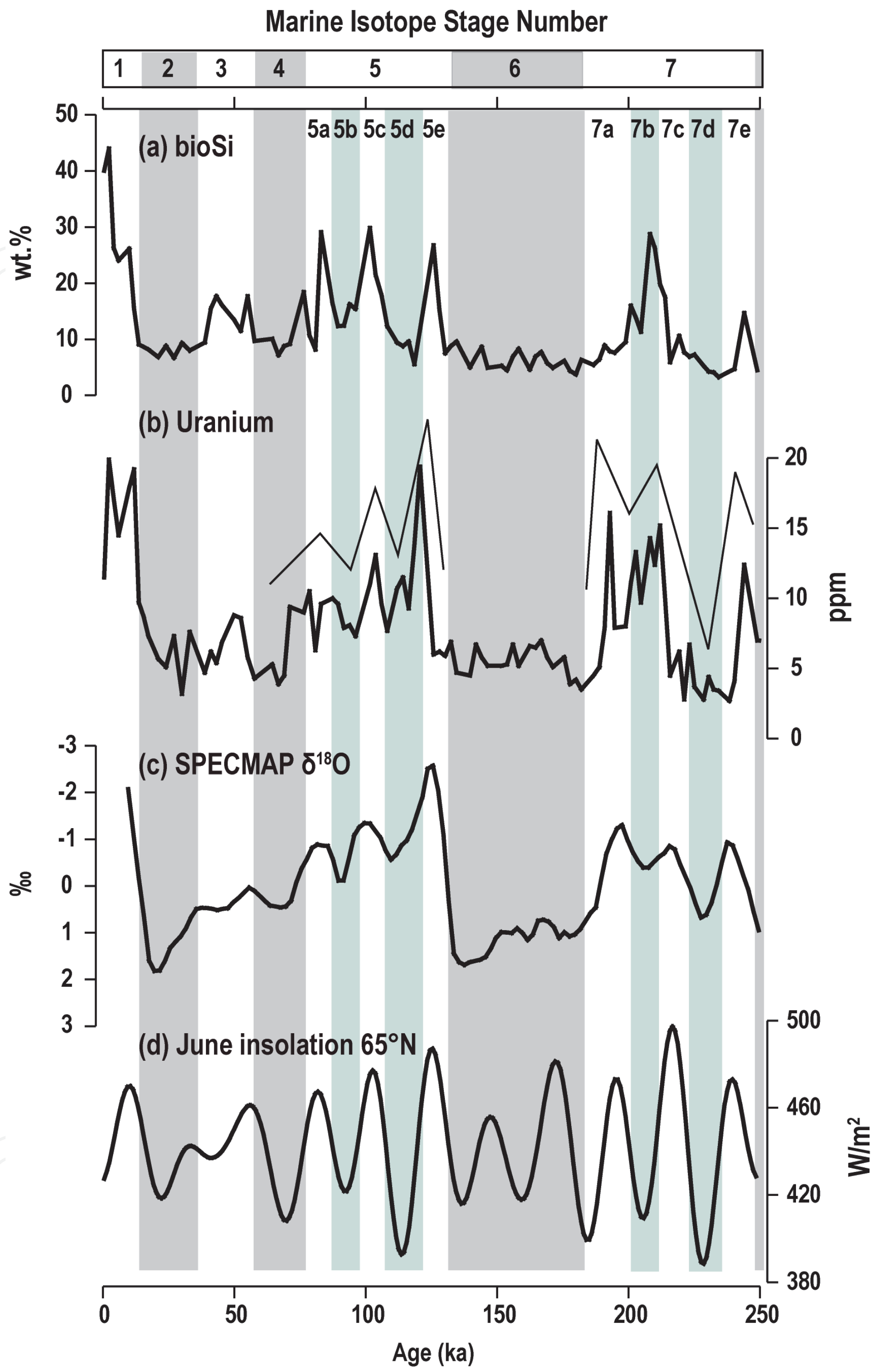


Fig. 3. Comparison of (a) bioSi (Colman et al. 1999) and (b) U in core BDP93-2 with (c) SPECMAP $\delta^{18}O$ data (Imbrie et al. 1984) and (d) 65º N June insolation (Laskar et al., 1993). Shaded and blue bands represent glacial periods and cooling substages in the interglacial periods, respectively. Boundaries between the glacial and interglacial periods were identified by Colman et al. (1999) based on correlations between the Lake Baikal bioSi data and SPECMAP curves.

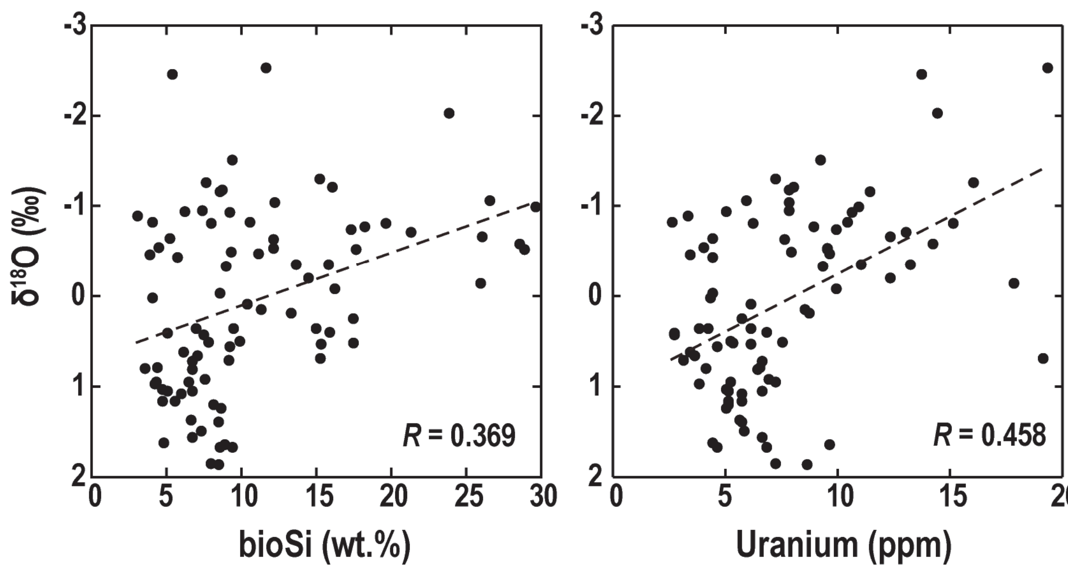


Fig. 4. Diagrams showing (a) relationship between Lake Baikal bioSi and SPECMAP $\delta^{18}O$ and (b) relationship between Lake Baikal uranium and SPECMAP $\delta^{18}O$.

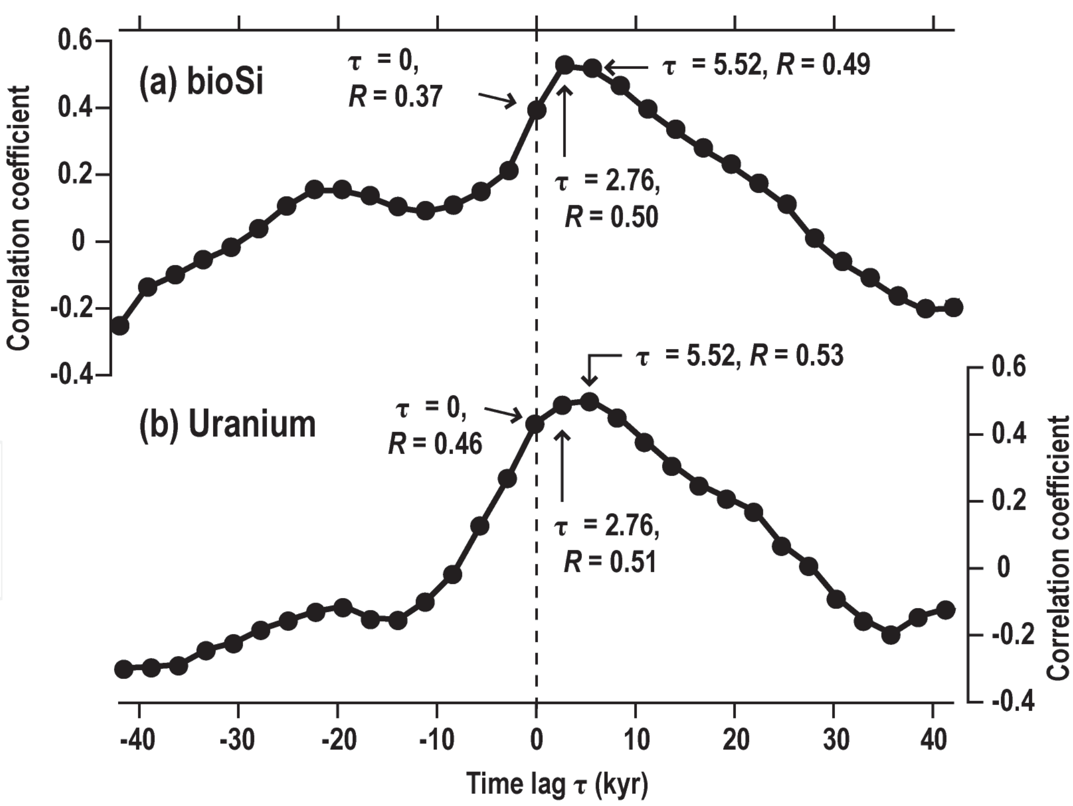


Fig. 5. Correlation coefficients versus time lag in thousand years (kyr) for individual cross correlations between Baikal paleoenvironmental proxies ((a) bioSi and (b) uranium) and SPECMAP $\delta^{18}O$ data. The highest peak represents the time lag at which the two time series were most similar. P-values of correlation coefficient in (a) and (b) are $<0.01$. This P-value represents >99% confidence level of the obtained correlation coefficient.

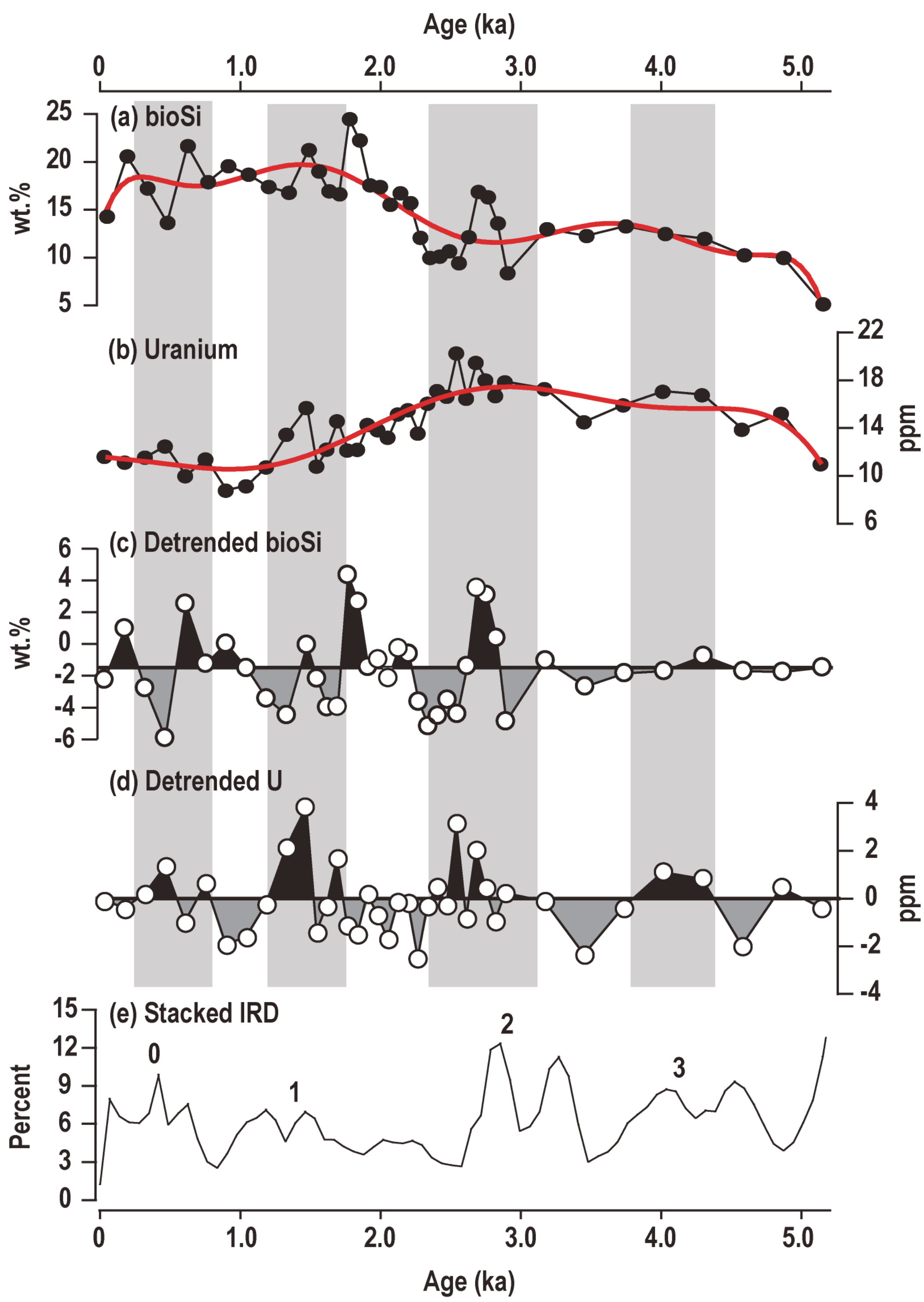


Fig. 6. Comparison of (a-d) Lake Baikal sediment records with (e) stacked IRD index in the North Atlantic (Bond et al., 1997, 2001): concentrations of (a) bioSi, (b) U, (c) the detrended bioSi, and (d) the detrended U in the sediment of core BSS06-G2. Variability in (c) and (d) were extracted by removal of the long-term trend in (a) and (b) (an 8th-order polynomial, red line) from the original curves, respectively. Shaded bands represent intervals of relative high U concentration in (d). Numbers in (e) indicate the Holocene IRD events labeled by Bond et al. (2001).

These tendencies observed in the long-term Lake Baikal records are also identified in the short-term variations in core BSS06-G2. Figure 6 shows the successive profiles of the Baikal bioSi and uranium and the North Atlantic stacked IRD records. A slight decline in the bioSi record, indicating the need to detrend (Fig. 6c), are identified for IRD events 2–0 (Fig. 6e). On the other hand, the uranium records show small peaks corresponding to events 3–0 during the last 5.2 kyr (Fig. 6b and d).

### 3.2 Relationship between the Lake Baikal records and climates in the Asian continental interior

In core BDP93-2 (Fig. 3), the bioSi and uranium increased in the interglacial period and decreased in the cold glacial periods. These fluctuation patterns can be associated with the bioSi production in the lake and the uranium weathering intensity in the watershed, reflecting the climate changes in the region. Specifically, the bioSi and uranium variations are dependent on changes in temperature and moisture in the region, respectively. This tendency is consistent with the previously reported climate in the Asian continental interior: dry conditions developed during the glacial periods and wet conditions during the interglacial periods.

Vegetation changes reflecting such climatic conditions were observed in the pollen records of sediment from Lake Baikal. Shichi et al. (2007) conducted pollen analyses on two cores from the northern and southern basins in Lake Baikal, and then reconstructed the geographical distribution of vegetation-types in the watershed between the glacial and interglacial periods. According to the analytical results, during the interglacial periods (MIS 1, 5, 7, and 9), the forest, mainly composed of *Pinus* and *Picea*, spread widely in the northern and southern watershed, whereas during glacial periods (MIS 2, 4, 6, and 8), open forest of *Artemisia*, *Betula*, etc., was distributed mainly around the southern region, while open forest and desert greatly expanded in the northern region. Rainfall reconstructed from pollen analyses of Lake Baikal sediment represents about 250 mm at the early stages of the last glaciation (117.5–114.8 ka) and 350–500 mm during the Holocene (MIS 1) and the last interglacial periods (MIS 5) (Tarasov et al., 2007).

The glacial aridity in the Asian interior is considered to have resulted from the expansion of the ice sheet along the north Eurasian continental margin, thus reducing the transport of water vapor from the North Atlantic via the westerly winds. The influence of the continental ice sheet on the glacial climates in the Asian interior has been previously investigated using GCM (general circulation model) simulations (Manabe & Broccoli, 1985; Bush, 2004). According to the GCM study, it is estimated that the annual mean soil moisture at 18 ka decreased by 20~60% compared to present day values (15 cm in soil depth) and that the difference between the annual mean precipitation during the LGM (Last Glacial Maximum) and the present day was 0.05–0.1 cm/day.

In the BDP93-2 records during the isotopic substage 5e/d and 7e/d, the abrupt declines in the concentrations of bioSi and uranium are identified (Fig. 3a and b). This is considered to be due to extreme insolation minima (65ºN, June) during 5d and 7d (Fig. 3d, Karabanov et al., 1998; Prokopenko et al., 2002). Such drops in bioSi concentration in the Lake Baikal sediment are also observed in the BDP96-2 core collected from the underwater Academician ridge in Lake Baikal (Fig. 1, Prokopenko et al., 2001a, 2002). This geochemical evidence suggests a decrease in bioproductivity over the whole area of the lake. Karabanov et al. (1998) found that the 5d-sediment of Lake Baikal is composed of a glacial clay layer with low TOC and bioSi content, which suggests glacial advances in the mountains of the Sayan–Baikal region.

During the Holocene IRD cooling events, BSS06-G2 bioSi concentrations generally tend to show a slight decrease, while the uranium depicts an increase (Fig. 6), contrary to the glacial-interglacial patterns (Fig. 3). This tendency is consistent with geological and botanical evidence previously recorded in the Asian continental interior, which was influenced by westerly winds. Pollen analyses of the Ugii Nuur lake sediment in central Mongolia from the last 8.7 ka revealed that a temperature, indicated by *Chenopodiaceae* concentration, decreased, whereas the moisture index, mainly based on concentrations of *Pinus*, *Poaceae,* and *Cyperaceae*, increased during the IRD events (Wang et al., 2009). Holocene records of Gun Nuur lake sediment in northern Mongolia, obtained by analyses of organic matter, organic $\delta^{13}C$, and magnetic properties, suggested that an increase in vegetation cover and biomass in the watershed, representing the cold/wet condition, corresponded to the timing of the IRD events in the North Atlantic (Wang et al., 2004). These two Mongolian lakes are located in the watershed of Lake Baikal. Recently, a comprehensive attempt to reconstruct the climate during the Little Ice Age (corresponding to the last IRD event 0) was made in westerlies-dominated Central Asia (Chen et al., 2010). Chen et al. investigated the spatial and temporal patterns of effective moisture (precipitation) variations in the region over the last millennium, using 17 sediment records from different sites, and then demonstrated that the LIA climate conditions became humid over a wide area.

### 3.3 Synchrony in the response to climatic changes between bioSi production and uranium weathering intensity

The bioSi and uranium variations recorded in the Lake Baikal sediment follow the same patterns of global climate changes on both the glacial-interglacial (Fig. 3) and centennial-to-millennial-scales (Fig. 6). We attribute this to the sedimentary depositional systems of the Lake Baikal region which are capable of directly responding to climate changes in the form of diatom production and uranium weathering intensity.

There are two proposed hypotheses for the causes of the variation of diatom/bioSi concentration in Lake Baikal sediment: the first is the amount of nutrient (soluble Si, P, etc.) supplied from the watershed (Chebykin et al., 2002, 2004; Gavshin et al., 2001) and the second is the change in the regional temperature in response to solar insolation (Prokopenko et al., 2001a). As shown in Figs. 3 and 6, the bioSi-uranium relationships in the Lake Baikal sediment indicate that the bioSi does not always increase with the higher uranium. This implies that the deposition of these two components rarely influenced each other. If the diatom/bioSi production in the lake is dependent on nutrient supply from the watershed, the bioSi-uranium relationships must show a positive correlation since soluble uranium should flow into the lake together with soluble Si and P. Therefore, we consider the second hypothesis to be the cause of the diatom/bioSi.

A study supporting our interpretation was conducted using the bottom sediment of Lake Baikal. Prokopenko et al. (2006) investigated the temporal variations of the Holocene temperature and humidity in the Lake Baikal region using the GCM climate model simulation and pollen fossil analyses. The results show that the Baikal bioSi/diatom production is in phase with the annual and winter temperatures and is a strong counter-phase with the humidity over the Holocene period. We think that a direct influence of temperature change on Baikal diatom production would result in a correlation between the bioSi record and a proxy of global climate changes.

As shown in Figs. 3 and 6, in the Buguldeika saddle of Lake Baikal, the uranium curve bears a closer resemblance to the paleoclimate indices compared with the bioSi record. In Lake

Baikal, the dissolved and detrital uranium in the water column are derived mainly from uranium-bearing rocks in the Selenga drainage basin, and are transported to the lake via the Selenga River and its tributaries (Edgington et al., 1996, 1997). Most of this uranium is in a dissolved form. Uranium contained in the sediment is mainly preserved as an authigenic component from dissolved uranium adsorbed on the surface of suspended sediment loads in the river and lake water and diatom frustules produced in the lake, which accumulates on the lake bottom (Edgington et al., 1996, 1997; Goldberg et al., 2010; Sakaguchi et al., 2006). Therefore, there is a possibility that the uranium deposition may be affected by the supply of detritus materials and the production rate of diatoms.

On the other hand, chronological studies on the bottom sediment in Lake Baikal indicate that the depositional rates are nearly constant through the glacial-interglacial periods over the whole lake area: 14.8 cm/kyr for the last 264 kyr at the Buguldeika saddle (Colman et al., 1999) and 3.9 cm/kyr for the last 6.7 Ma at the underwater Academician Ridge (Kravchinsky et al., 2003). The observed sedimentary features of Lake Baikal suggest that the amount of materials on which the uranium adsorbed would vary little over the entire sequence, and the concentration of uranium in the bottom sediment would be only slightly dependent on the abundances of detritus materials and diatom frustules. Therefore, we believe that the uranium variation strongly influences the input of uranium into the lake, reflecting the weathering in the Selenga drainage basin which was associated with changes in moisture levels in the region.

## 4. Conclusion

Comparing the bioSi and uranium variations, the Lake Baikal records from the Buguldeika saddle and the paleoproxy records of global climate change showed only slight differences on glacial-interglacial and centennial-to-millennial scales — there are statistically no significant differences between these two records. Thus, we conclude that the bioSi and uranium records of Lake Baikal sediment follow the same degree of global climate changes on time scales covering centuries to tens of millennia.

## 5. Acknowledgement

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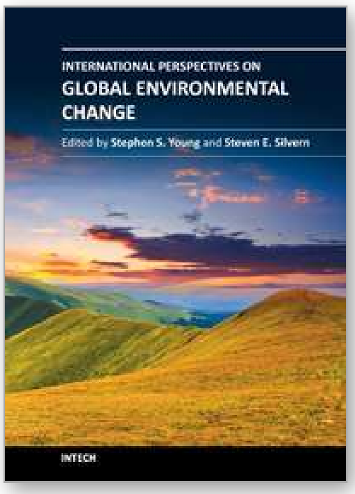


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