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Late Quaternary clay mineral record in Central Lake Baikal (Academician Ridge, Siberia)

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Abstract

We investigated the mineralogical composition of two cores recovered on the Academician Ridge (Central Lake Baikal, Siberia). Sedimentological features show that the cores are unaffected by turbidity currents. However, hemipelagic deposition is not continuous, but intermittently disturbed by syn- or post-sediment reworking (e.g., bioturbation, slumps, faulting). Such modes of deposition are consistent with the complex uplift history of the ridge. Bulk mineralogy suggests that terrigenous sediment supplies are constant through glacial/interglacial stages, and diluted by diatom-rich intervals related to warmer interglacial stages. The core stratigraphy is based on the correlation of the diatom zonation and opal abundance with the marine oxygen isotope reference curve SPECMAP. The ~ 8-m cores partly recover the last four interglacial/glacial cycles, i.e., since oxygen isotope stage 8. We test the use of clay minerals as a proxy for paleoclimatic reconstruction. The clays are more weathered during the diatom-rich intervals in agreement with warmer climate conditions. However, the mean clay composition does not change significantly through glacial/interglacial stages. This observation implies that, in the Academician Ridge sediments, a simple smectite/illite ratio (S/I) does not alone provide a reliable indicator of climatic variation. It reflects the complex clay assemblages, especially the smectite group, delivered to Central Lake Baikal. Smectites include primarily illitesmectite mixed layers, made of a mixture of montmorillonite and beidellite. According to their behavior after cation saturation, the illite-smectite mixed layers are primarily transformed smectites, with some neoformed smectites intermittently observed. In addition, Al-smectites occur in minor proportions. We conclude that the S/I ratio has a climatic significance only if it evolves in parallel with the weathering stage of the clays and is confirmed by a change in the composition of the smectites.

Keywords: clay mineralogy ; central Asia ; Lake Baikal ; Quaternary ; paleoclimate ; lacustrine sedimentation

1. Introduction

When reconstructing climate evolution, the response of the continental system to climate changes and its interaction with oceanic and atmospheric circulation patterns must be taken into account. The study of lacustrine sediments as an accurate climate archive has been developed and improved recently by international programs like PAGES (e.g., PEPII, Dodson and Lui, 1995). Among the lakes, the Lake Baikal sedimentary record provides a good climate archive (e.g. Minoura, 2000). The lake is the deepest and one of the oldest on Earth, with ~7.5 km of sediments dating back to the Middle Eocene (Hutchinson et al., 1992). It is located within the Asian continent and is far from any direct influence from oceans and ice sheets. Paleoclimate studies on Baikal have been intensified through the Baikal Drilling Program (BDP) (e.g., Kuzmin et al., 1993; BDP-Members, 1997). This project has focused on uplifted areas, such as the Academician Ridge, which is not influenced by turbidites (Kuzmin et al., 2000; Nelson et al., 1995). Sediments from this site are composed of diatom-rich intervals alternating with clay-rich intervals, and previous climatic reconstructions have been mainly based on biotic proxies, such as diatoms (e.g., Granina et al., 1992; Grachev et al., 1998; Mackay et al., 1998; Karabanov et al., 2000; Bangs et al., 2000), pollen and spores (Bradbury et al., 1994; Demske et al., 2000), chrysophytes (Karabanov et al., 2000), or $\delta^{13}C$ in organic carbon (Prokopenko et al., 1999).

Abiotic parameters, clay minerals in particular, have been used only in a few studies on Baikal sediments (Melles et al., 1995; Williams et al., 1997; Yuretich et al., 1998; Horiuchi et al., 2000; Solotchina et al., 2002). However, detrital clays can be a powerful tool for climatic reconstruction, as emphasized in many studies on marine sediments (e.g. Chamley, 1989; Fagel et al., 1994). Our aim is to investigate the evolution of clay assemblages in the Baikal lacustrine record of the last ca. 250 ky. Previously reported clay mineral data from BDP cores from the Southern Central Basin (Buguldeika Saddle, Yuretich et al., 1998, see Fig. 1) evidenced a systematic increase of smectites during diatom-bearing intervals, indicating that a climate signature can be recorded within the clay-size fraction. The changes have been explained by warmer conditions. Assuming all smectite is derived from soils, such a mineralogical trend would reflect an increased hydrolysis within the

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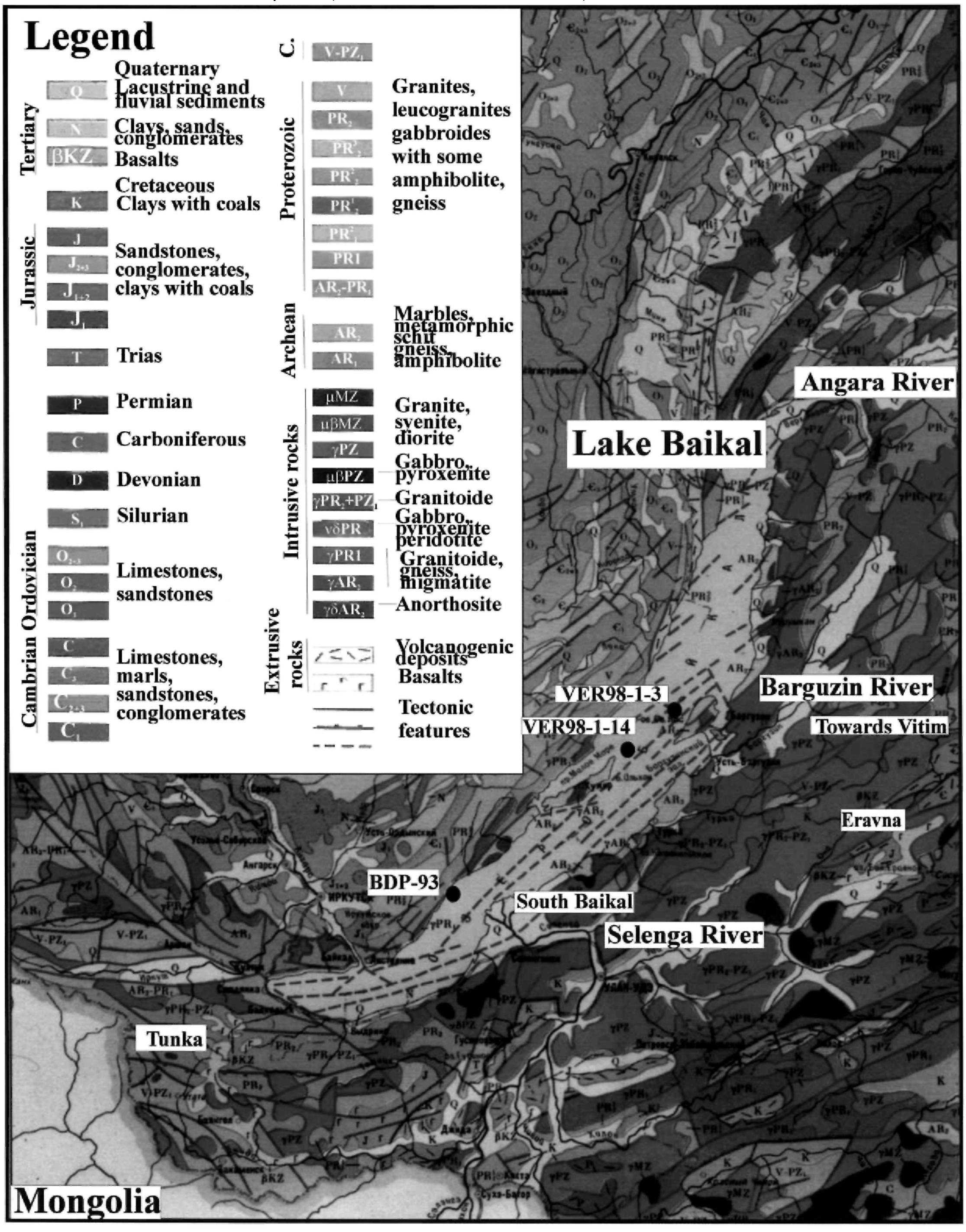
watershed during interstadial and interglacials (Yuretich et al., 1998). It is more likely that only a part of the clay assemblage reflects weathering conditions in the watershed (the remainder of the clays deriving from volcanic material or being reworked from sedimentary rocks), and thus correlates with climate variability. To test this hypothesis, we have conducted a detailed X-ray diffraction (XRD) study of the clay assemblages within two cores from the Academician Ridge, Lake Baikal, with particular emphasis on the investigation of the smectite fraction. Cation (Li, K) saturation techniques have been used in order to provide a more accurate identification of smectite and allow the differentiation of species formed through either weathering or neoformation (Thorez, 2000).

Fig. 1. *Geological map of the Lake Baikal area (modified from Galazii, 1993). Also plotted are the locations of the investigated cores, VER98-1-3 and VER98-1-14, on the Academician Ridge in the central part of the lake; the location of Core BDP-93 (Yuretich et al., 1998) recovered on the Buguldeika Saddle (Central Basin); the main rivers, and the volcanic fields (in italics; Rasskazov, 1994).*



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2. Materials and methods

2.1. Core location and sampling

Two piston cores were collected in the central part of Lake Baikal on the Academician Ridge (Fig. 1) during a joint Russian-German-Japanese expedition in September 1998 with the *RV Vereshagin*. Core VER98-1-3 was drilled on the northern part of the ridge (108°19'02"E, 53°44'56"N) at a water depth of 373 m. The total length of the core was 1092 cm, but its lower part (835-1092 cm) was mechanically disturbed during the coring operation and therefore not investigated further.

Core VER98-1-14 was recovered from the southern part of the Academician Ridge (107°58'10"E, 53°31'23"N) at a water depth of 412 m. The total length of the Core was 980 cm, but we have restricted our investigation to the first four core sections (0-773 cm) in order to cover approximately the same time interval in the two cores.

Core sub-sampling has been done for water content (2 cm resolution, GFZ, Potsdam, Germany), diatom analyses (5 cm resolution, GFZ, Potsdam, Germany), bulk and clay mineralogy (10 cm resolution, University of Liege, Belgium), and direct opal measurements (2 cm resolution, GFZ, Potsdam, Germany).

2.2. Core lithology

For both cores, the sedimentological analysis reveals the internal organization of the clayey silty layers and their interspersing by diatom-rich intervals (Fig. 2). At least 3-dm-thick diatom-rich layers are evidenced within Core VER98-1-3, at core intervals 313.5-370 cm, 628-670 cm, and 729-764 cm. Their lower limit is sharp, underlined by an erosive scar, marked usually by a color change (from gray to greenish gray, to brownish). The diatom-rich layers are generally massive or faintly laminated. The upper boundaries of the diatom-rich layers are characterized by a gradual change of texture and color. The clay-rich intervals are either massive, coarsely to finely laminated or bioturbated. Tiny concretions occur as dispersed grains, scattered streaks or aligned and forming millimetric-scale layers. The concretions occur in many detrital intervals; their composition, deduced by EDAX, is vivianite, a Fe-phosphate. The concretion-rich intervals often underlie coarsely laminated sediments. Several clay-rich intervals are affected by syn- or post-sedimentary microslumps, and some parts of the core exhibit gently inclined bedding affecting groups of layers.

The sedimentological features in Core VER98-1-14 are quite similar to those in Core VER98-1-3 (Fig. 2). Five-dm-thick diatom-rich layers are evidenced at core intervals ~ 245-260, 270-340 cm, 395-450 cm, 570-670 cm and ~ 755-773 cm. Sediment reworking or disturbance occurs intermittently (e.g., microfaults, bioturbation). A decimetric-scale escape feature with associated slumping and cross-stratification suggests that the upper part of the core (~ 80-110 cm) shows post-depositional disturbance. Overall, the core is less disturbed by slumps than Core VER98-1-3.

2.3. Mineralogical analyses

Bulk and clay minerals were identified by XRD and carried out on unoriented powder mounts or on oriented aggregates respectively. Qualitative and semi-quantitative estimation were based on peak intensity measurements on X-ray patterns (Philips PW1390 diffractometer with CuKα radiation).

2.3.1. Bulk fraction

The powder mounts were obtained by grinding ~1 g of bulk dried sediment in a mortar. The powder was placed in an Al sample holder, using gentle pressure to limit any clay mineral orientation. Semi-quantitative estimation was obtained by applying to the measured intensity of the reflections the correction factors determined by Cook et al. (1975) and modified by Boski et al. (1998) (Fig. 3). For quartz the intensity of the 4.26 Å peak was multiplied by 100/35; the relative contribution of feldspars was calculated from the intensity of the 3.18 Å peak multiplied by 2, and the total clay contribution, excepted chlorite, by the intensity of the common 4.4 Å peak multiplied by 20. In addition, any unusual increase of the background around 4.04 Å has been assigned to opal. The usual background level has been averaged by using samples from adjacent diatom-barren intervals. Biogenic opal abundance has been determined by multiplying by 20 the height of the 4.04 Å reflection above the reconstructed background (Boski et al., 1998). The accuracy of the XRD opal content estimation has been tested by comparing those values with direct opal measurement by spectrophotometry after Na_2CO_3 leachings (see Section 3).

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Fig. 2. *Lithology of sediment cores VER98-1-3 (108°19'02"E, 53°44'56"N, at 373 m water depth), and VER98-1-14 (107°58'10"E, 53°31'23"N at 412 m water depth), Academician Ridge, Lake Baikal. Modified from F. Hauregard, unpublished data.*

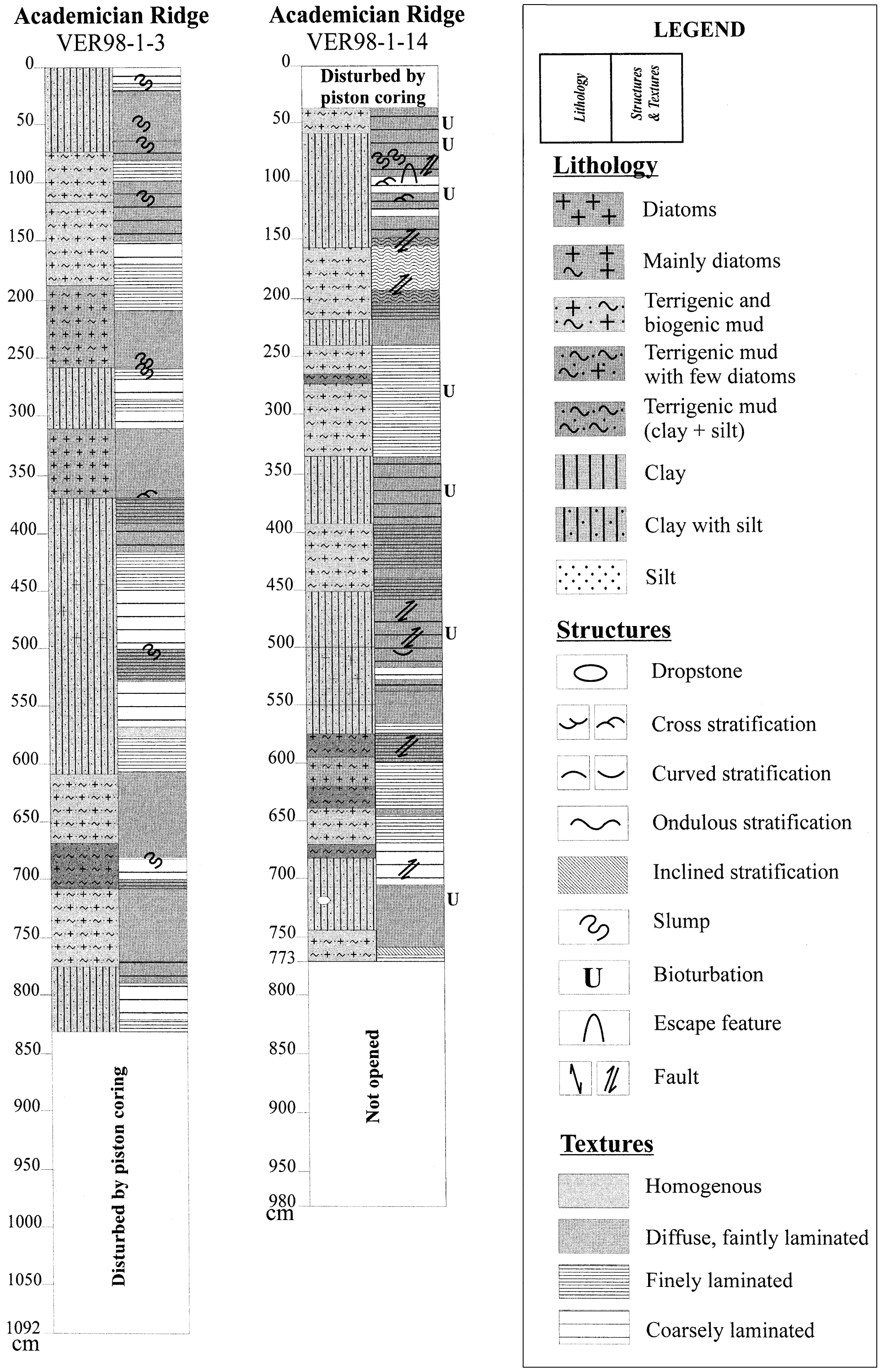

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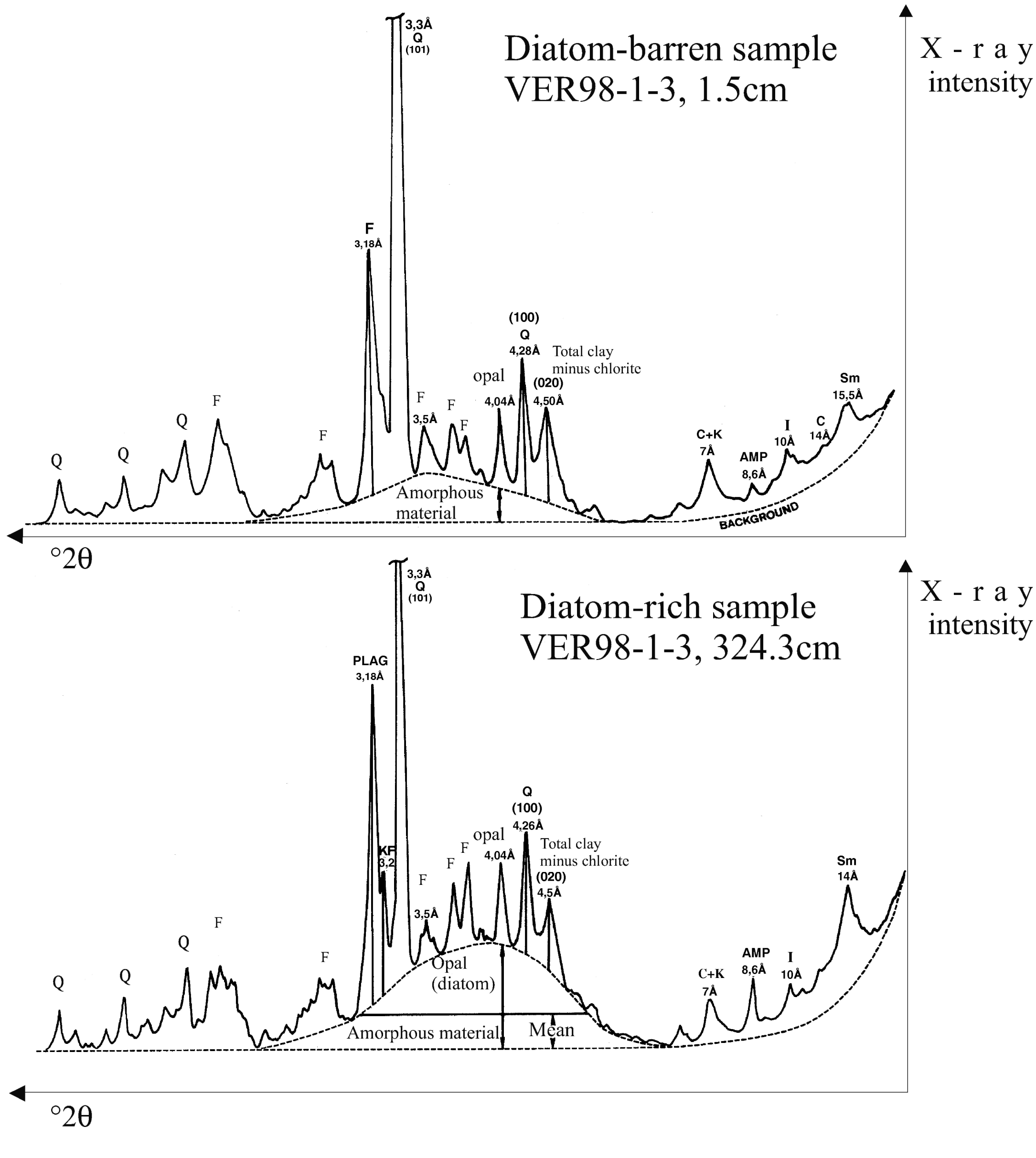
Fig. 3. *Method for semi-quantitative determination of bulk mineralogy from XRD pattern on unoriented powder mounts. Example from a VER98-1-3 clay-rich (at 1.5 cm) sample and a diatom-bearing sample (at 324.3 cm). The x-axis scale is in degree 2θ (CuKα radiation, λ =1.7890 Å, scanning speed of 2° 2θ/min). AMP, amphibole; C, chlorite; C+K, chlorite+kaolinite; F, feldspar; I, illite; KF, K-feldspar; PLAG, plagioclase; Q, quartz; Sm, smectite. See text for explanation.*



2.3.2. Clay size fraction

2.3.2.1. Sample preparation. For clay minerals, oriented aggregates on glass slides (Moore and Reynolds, 1989) were prepared from the less than 2-μm fraction obtained by suspension in distilled water of 1-2 g of the dried bulk sediment. This was followed by decantation (settling time from Stoke's law) and centrifugation at 3000 rpm. Routine XRD clay analyses included the successive measurement of an X-ray pattern in air-dried or natural condition (N), after solvation with ethylene glycol for 24 h (EG), and after heating to 500°C for 4 h (500). In addition, Li saturation and some K saturation were done on the less than 2-μm fraction in order to help identify the chemical composition and the genetic conditions of smectites, and to improve the quantification of chlorite, illite chlorite and illite-smectite mixed layers. For Li saturation (modified from Lim and Jackson, 1986), the clay suspension was washed with 2 N aqueous LiCl overnight. Samples were then rinsed with demineralized water, and prepared as oriented aggregates. XRD analyses were conducted in sequence on the air-dried slide (Li-N), heated at 300°C (Li-300, 2 h), and finally overnight glycerol solvated (Li-300Gl). K-saturated clay mounts with 2 N aqueous KCl were X-rayed successively as air-dried (K-N), heated to 110°C (K-110), and solvated with ethylene glycol (K-110EG).

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2.3.2.2. Clay identification. Semi-quantitative estimations (± 5-10%; Biscaye, 1965) of the main clay species (illite, chlorite, smectite, kaolinite, random mixed layers) were based on the height of specific reflections, generally measured on EG runs or corrected by data obtained, in parallel, after Li saturation. The intensity of the 7 Å peak is taken as an internal reference. The intensities were divided by a weight factor (1 for illite and smectites; 2.5 for chlorite and illite chlorite mixed layers (10-14c); 1.4 for kaolinite; 5 for the Al-smectite fraction) and values were summed up to 100% (Biscaye, 1965).

(a) The illite content was determined by the peak intensity at 10 Å on EG. The width at half-height of this reflection (crystallinity index) was used as an indicator for the weathering intensity.

(b) The illite-chlorite or (10-14c) mixed layer was identified by the height of the diffraction band centered on 12 Å on the EG pattern.

(c) The smectite group was divided into three components: (1) a fraction expandable at 17 Å after EG solvation, coded $(10\text{-}14Sm)_{17}$; (2) a fraction which collapsed partly to 12 Å in Li-300, and identified as $(10\text{-}14Sm)_{Al}$; and (3) a fraction already expanded at 17 Å in the air-dried sample Al_{17} or Al-smectite.

(1) $(10\text{-}14Sm)_{17}$ - This smectite fraction was deduced from the intensity of the 10 Å peak on the N and 500 patterns, as follows: $(10\text{-}14Sm)_{17} = I_{500}\text{-}I_N$. On the EG pattern, the shape of the 17 Å peak (cf. smectite classes, Thorez, 1976) and the *v/p* ratio (Biscaye, 1965) indicated that this is a random illite-smectite mixed layer $(10\text{-}14Sm)_{17}$ rather than pure smectite (Retke, 1981).

(2) $(10\text{-}14Sm)_{Al}$ - The occurrence of Al-hydrox-ylized interlayers in illite-smectite mixed layers hampered a total collapse to 12 Å upon heating to 300°C in the Li test or to 10 Å upon heating to 500°C in the 500 test. This fraction is evidenced from the intensity of the 12 Å peak on the Li-300 and EG patterns, as follows: $(10\text{-}14Sm)_{Al} = I_{Li300}\text{-}I_{EG}$. The contribution of the $(10\text{-}14Sm)_{Al}$ was not taken into account; it is partly included in the $(10\text{-}14Sm)_{17}$ estimation.

(3) Al_{17} - Some expansion was already observed at 17 Å on the air-dried sample. This reflects the occurrence of Al pillars within the interlayers of a fraction of smectite. The relative contribution of Al-smectite was based on the intensity of the residual 17 Å in the Li-300 X-ray scan, which was then reported on the EG.

(d) The contribution of chlorite was determined by the height of the 14 Å peak on the Li-300 pattern. The value is then reported on the EG pattern using the 7 Å peak as an internal reference. In Core VER98-1-3, the chlorite comprised a fresh fraction and a partially vermiculized one. The occurrence of random chlorite-vermiculite (14c-14v) mixed layer was evidenced by comparing the intensities of the diffraction band centered 12 Å in both Li-300 and 500 tests.

(e) The occurrence of kaolinite was deduced from a doublet peak around 3.5 Å, resulting from the partial overlapping of the (004) chlorite reflection at 3.54 Å and the (002) kaolinite reflection at 3.57 Å, in either the air-dried or glycolated state. Because the (001) of kaolinite and (002) of chlorite were superimposed at 7 Å, the estimation of kaolinite content was based on the intensity measured at a break or shoulder affecting the 7 Å peak on the low angle side in the EG pattern. In some cases (e.g. break not obvious), the kaolinite content was deduced from the chlorite content by using the chlorite/kaolinite peak intensity ratio measured at 3.54 and 3.57 Å.

2.3.2.3. Smectite composition and genetic origin. Within the expandable smectite fraction, the abundance of montmorillonite and beidellite was estimated by comparing the intensities of the 10 Å peak on the three Li-saturated X-ray patterns, as follows (Thorez, 1998): $(10\text{-}14Sm)Al = I_{Li300}\text{-}I_{LiN}$; montmorillonite = $I_{Li300Gl}\ \text{-}1_{LiN}$; beidellite = $I_{Li300}\text{-}I_{Li300Gl}$.

Similarly, nontronite or stevensite could be differentiated through a comparison of the intensities of the 14 Å peak on the K-saturated samples (Thorez, 2000), but these species were not detected in Core VER98-1-3. Saponite could not be identified on the basis of cation saturation, because its (060) reflection is masked by reflections belonging to non-clay minerals such as quartz (Moore and Reynolds, 1989).

The genetic origin of smectites (transformed or neoformed) was identified by K saturation (Thorez, 2000). Transformation refers to a smectite component generated by moderate or weak weathering (opening of interlayers) of a parent illite or mica. Neoformation implies a neosynthesis of a structurally similar smectite component. The occurrence and relative contribution of neoformed smectites were estimated by comparing the intensities of the 10 Å peak on the the K-saturated X-ray patterns (K-N, K-110, K-110EG), as follows: neoformed $(10\text{-}14Sm)_{Al} = I_{K110}\text{-}I_{K110EG}$; transformed $(10\text{-}14Sm)_{Al}$ if $I_{K110} = I_{K110EG}$.

3. Results

3.1. Bulk mineralogy[1]

The relative abundance of the main mineral components (quartz, feldspars, clays and opal) is plotted in Fig. 4. Amphibole though ubiquitous is only present in trace quantities. In Core VER98-1-3, the opal content as deduced from bulk XRD (see Section 2) starts to increase at 98 cm. It remains high, comprised between 17 and 48%, down to 255 cm. It drops at 265 cm and remains close to zero between 265 and 305 cm. It increases at 316.5 cm and reaches a maximum (74%) at 341.5 cm. The opal content remains constant at low values (~ 20%) from 371 to 606 cm, and is thereafter characterized by two maxima at 655 cm (68%) and 743 cm (65%). The ranges of variation of the other minerals are 10-55% for feldspars, ~ 5-35% for quartz, and ~ 5-40% for total clays (except chlorite). The fluctuations of opal deduced from bulk XRD match well with chemical measurements of opal (Fig. 4). The indirect XRD estimate usually overestimates the opal content by ~ 20% in the diatom-rich intervals. In contrast, in the diatom-barren interval, the chemical measurement gives up to 5% of opal content. This could be explained by the influence of the Na_2CO_3 leaching on other silicate components. If the opal contribution is not taken into account, the relative contributions of quartz (mean 30%), feldspars (mean 46%) and clays (mean 24%) do not significantly change with depth. Such a trend supports a uniform supply through time.

In Core VER98-1-14, the bulk mineralogy is dominated by the clay component (38-73%, mean: 60%). The relative abundances of quartz and feldspars are on average half as much as in Core VER98-1-3 (quartz: 10-21%, mean 15%; feldspars: 13-30%, mean 20%). The opal content reaches a maximum value (36%) at 420.2 cm. Its mean contribution for the whole core is quite low (6%). The indirect estimate of opal from bulk XRD is close to the direct opal measurements (Fig. 4).

3.2. Stratigraphy and age model

In both cores, the opal profile has a pattern similar to the standard marine oxygen isotope curve (SPECMAP). In sediments from Lake Baikal, the correlation between the opal content and SPECMAP, the latter calibrated with AMS^{14}C dates for the latest Quaternary and Holocene, has been used as a stratigraphic tool (Colman et al., 1995; Grachev et al., 1998) to extrapolate further back in time (i.e., before 35 kyr). Fifteen diatom complexes (I-XV) have been identified in several cores from Lake Baikal (Bradbury et al., 1994; Likhoshway, 1998). In this study, the age model (Fig. 4) is based on the correlation between the depths at which diatom Complexes VII-XV were found in Core station 18 (i.e., the reference section from Academician Ridge; Likhoshway, 1998) and Core VER98-1-3. Assuming a constant sediment accumulation rate, the biostratigraphic correlation suggests that the last ~ 54 kyr (i.e., the uppermost 2.8 m of the sediments) are missing. The 8 m of Core VER 98-1-3 probably record a time interval from OIS 4 down to OIS 8, from ~ 55 to 250 kyr. This is based on the following findings. (1) Holocene assemblages are absent. (2) Core VER98-1-3 starts with a diatom-barren clay interval, correlated to interval B. (3) The first identified diatom eco-zone is Complex VIII. (4) The abundance peak of Complex XII, corresponding to OIS 5e and having an age of 120 kyr (Colman et al., 1995), occurs in Core VER98-1-3 at ~ 340 cm.

For Core VER98-1-14, our proposed age model (Fig. 4) is based on the correlation between the fluctuations of opal and water content of Core VER98-1-3 and Core VER98-1-14. Core VER-98-1-14 spans at least from OIS 2 to 7e. The total length of interglacial OIS 5 is slightly shorter in Core VER98-1-14 (from 245 to 450 cm) than in Core VER98-1-3 (from 98 to 366 cm). The glacial OIS 6 is only half as thick in Core VER98-1-14 as in Core VER98-1-3.

[1] All bulk mineralogical data will be available on the web in the CONTINENT database (http://continent.gfz-potsdam.de) or upon request from the first author.

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Fig. 4. *Age model reconstruction based on diatom assemblages identified in several Lake Baikal cores (Bradbury et al., 1994; Likhoshway, 1998) and some AMS ^{14}C dates (Colman et al., 1995). The numbers refer to the diatom assemblages, the letters to the diatom-barren intervals. For cores VER98-1-3 and VER98-1-14, the variation with depth of the relative abundance (%) of the main mineral components estimated from bulk XRD diffraction on unoriented mounts is reported. Where available, the opal content measured by spectrophotometry after Na_2CO_3 leaching is also plotted (dotted line). The most probable correlation between the diatom abundance and SPECMAP is reported in regard to the lithology; the numbers correspond to the equivalent oxygen isotope stage (OIS).*

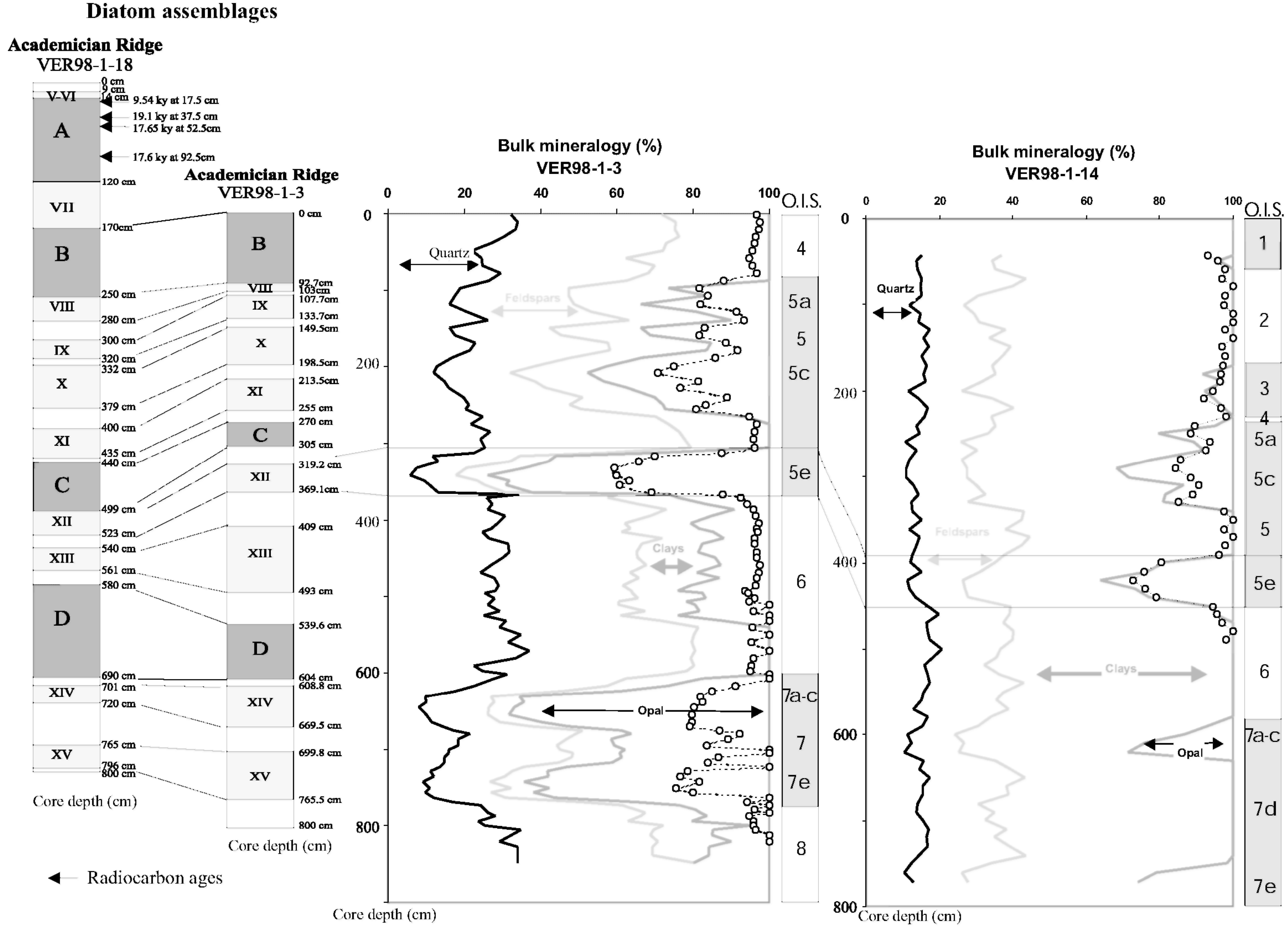

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Fig. 5. *Variation with depth in Core VER98-1-3 (a) and Core VER98-1-14 (b) of some clay mineral XRD parameters: illite crystallinity (Thorez, 1976), weathering stage of chlorite, Biscaye's (1965) v/p. The reported water content curve is obtained by comparing the wet and dry (after 3 days at 85°C) weights of a 10-ml aliquot of sediments. This supports identification of the glacial and interglacial intervals (the numbers correspond to OIS, the shadowed intervals underline the interglacials).*

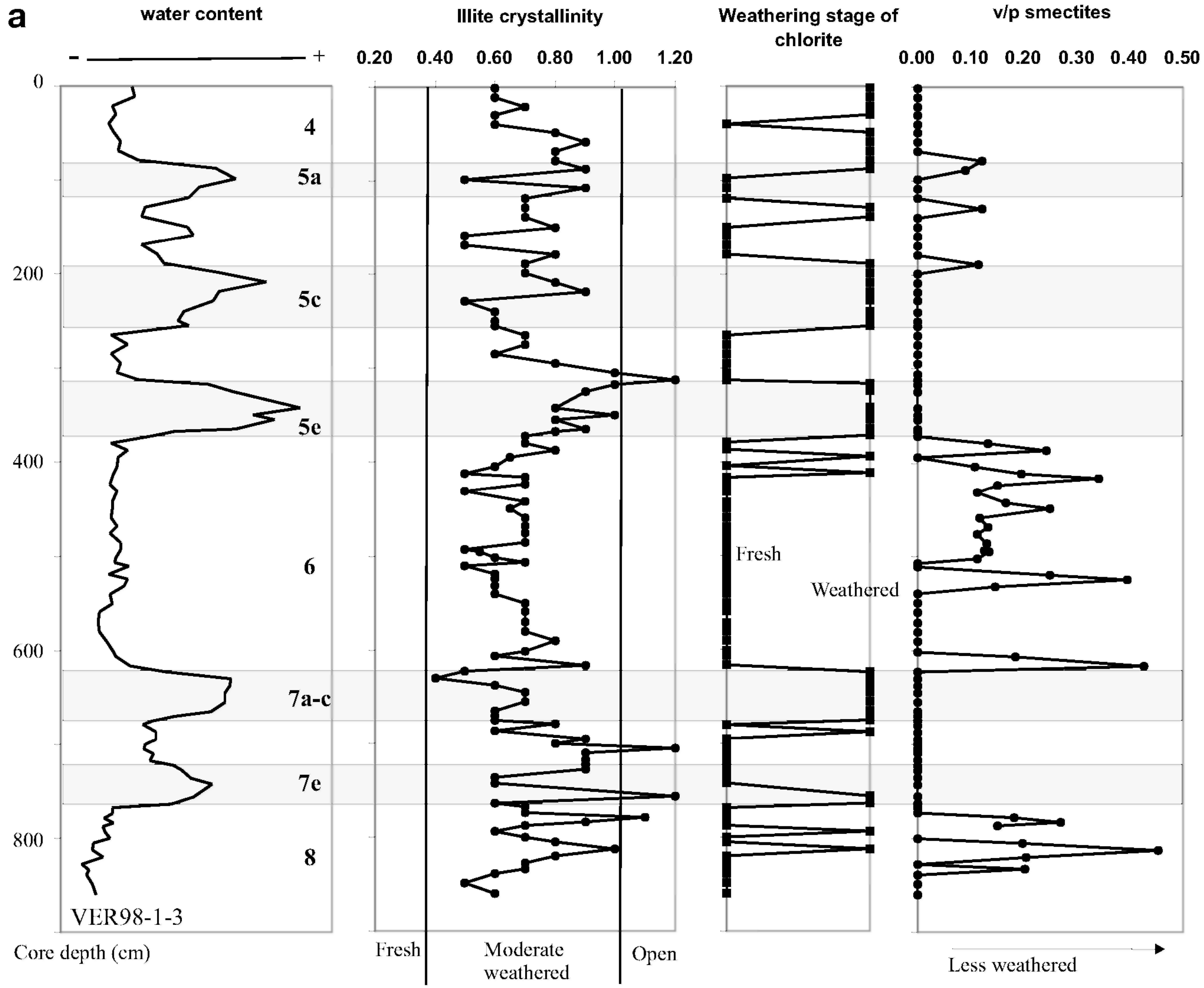

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***Fig.** 5 (Continued).*

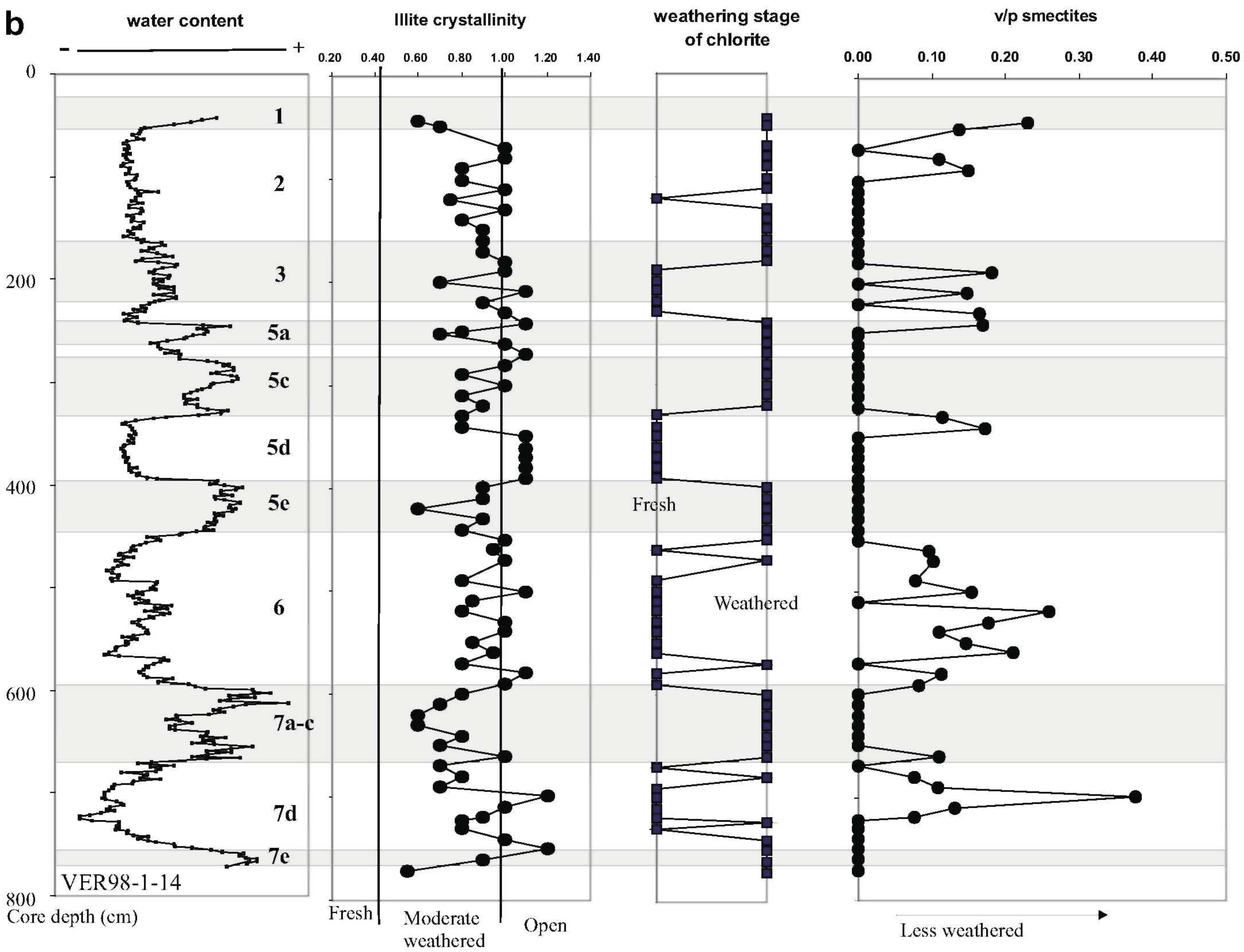

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Fig. 6. *Variation with depth in Core VER98-1-3 (a) and Core VER98-1-14 (b) of the smectite/illite ratio (17/10 EG) with regard to changes in the smectite composition. Key as in Fig. 5. The numbers in the right margin represent the mean cumulated contribution (in % of the total clay fraction) of beidellite and Al-smectite within the interglacials. The mean contribution within the total clay fraction of montmorillonite and beidellite+Al-smectite is also reported.*

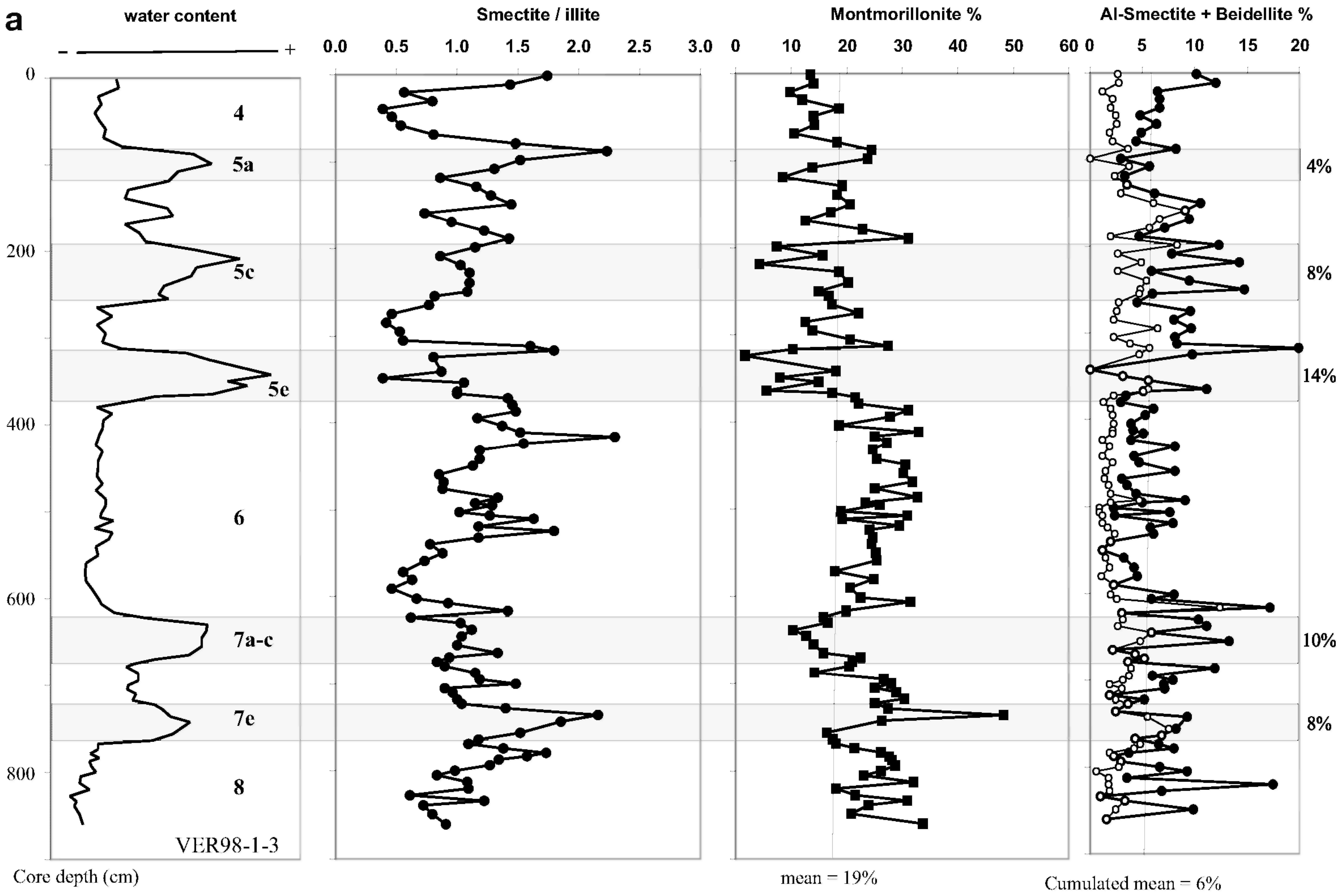

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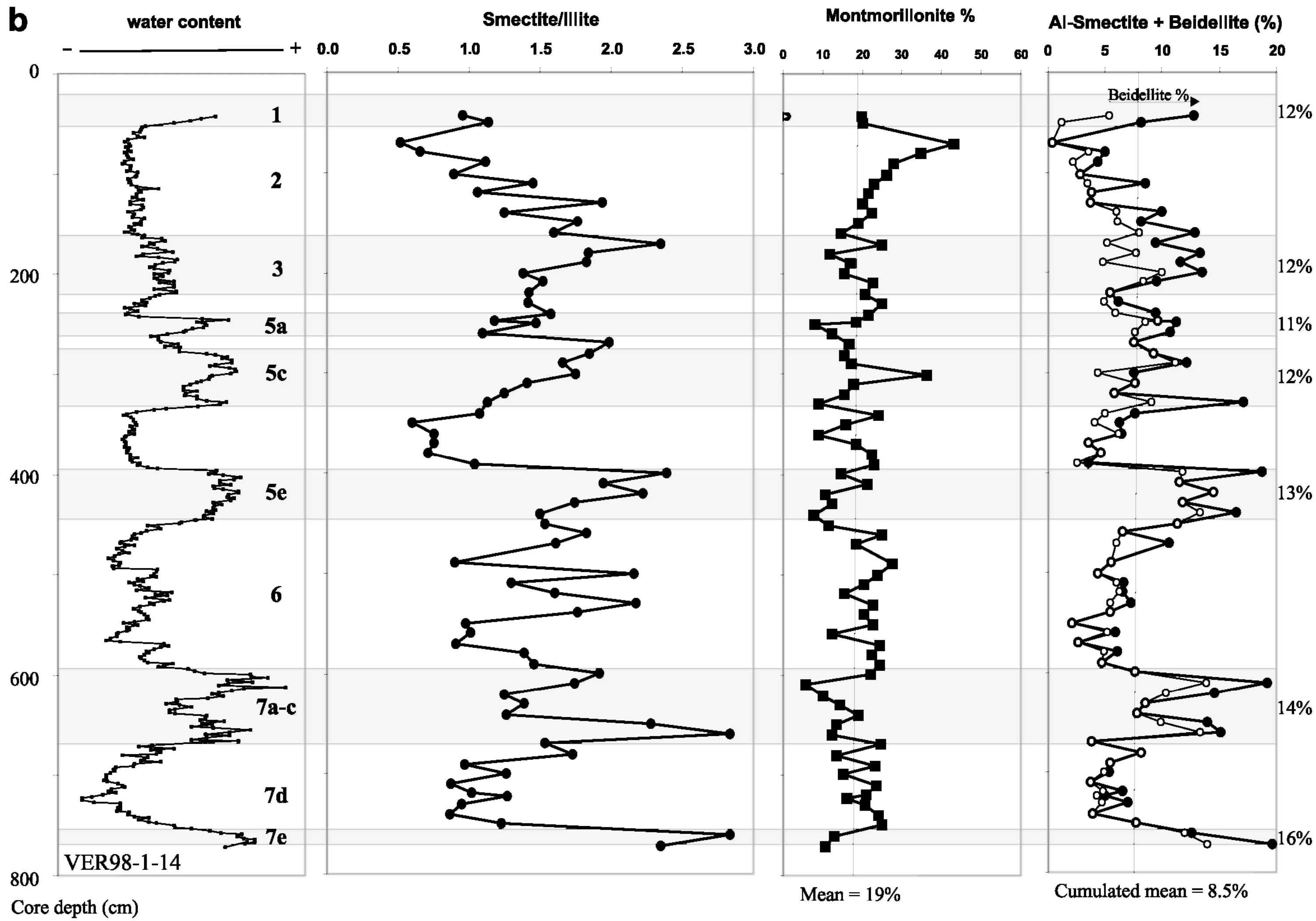
Fig. 6 *(Continued).*



3.3. Clay mineralogy[2]

In both cores, the Late Quaternary clay assemblage is complex. It includes kaolinite, fresh to slightly weathered (or open) illite, mixed layers (10-14c), fresh and/or degraded chlorite accompanied by (14c-14v), illite-smectite mixed layers $(10\text{-}14Sm)_{17}$ and Al-smectites. For the two cores, we will identify the assemblage composition, describe the weathering stages of clay minerals, and follow its fluctuations with increasing depth.

3.3.1. Core VER98-1-3

In Core VER98-1-3, the illite crystallinity varies from ~ 0.4 to 1.2° 2θ (Fig. 5a). Values ~ 0.4° 2θ correspond to a fresh mica whereas those above 1° 2θ stand for a highly weathered mineral (open illite; Thorez, 1976). A moderately weathered illite generally characterizes the diatom-poor intervals, except at the base of the core (below 700 cm) where the index remains high (1-1.2° *2 θ*). Open illite occurs within the diatom-rich intervals or just after the transition (~ 30-40 cm, e.g. index peak at 312 cm and 616 cm, diatom peak at 340 and 655 cm, respectively). Chlorite is a Mg variety. Fresh chlorite is primarily observed in diatom-poor intervals (Fig. 5a). In contrast, degraded chlorite, including a vermiculite fraction, occurs within diatom-rich intervals. The Biscaye *v*/*p* ratio ranges from 0 to 0.45 (number without unit); the higher value indicates a maximum of 70% swelling interlayers within the random illite-smectite mixed layer, whereas a zero value corresponds to a swelling of less than 35% (Retke, 1981). All *v*/*p* ratios > 0.15 (i.e. > 50% of swelling interlayers, Retke, 1981) primarily correlate with diatom-poor intervals (Fig. 5a), indicating a less intensified clay weathering in those levels.

For the whole core, the mean clay mineral composition comprises 40% illite, 25% smectites, ~ 3% Al-smectite, 16% kaolinite, 11% chlorite, and 5% illite chlorite (10-14c). The composition of the diatom-poor intervals is quite similar to the composition of diatom-rich intervals, except for a slight decrease in chlorite counterbalanced by an increase in smectite.

The total smectite content usually varies between 10 and 35% (50% as an exception at 736 cm). The smectite group is composed of three components (Fig. 6a): $(10\text{-}14Sm)_{17}$, $(10\text{-}14Sm)_{Al}$ and Al-smectites (Al_{17}). Li saturation evidences a mixing of montmorillonite and beidellite. K saturation confirms the lack of nontronite (and stevensite), and demonstrates that smectites are related to transformation and/or neoformation processes. Transformation smectite dominates in the whole series, except in two montmorillonitic-rich samples (at 129 cm and 812.6 cm). Among the smectite fraction, Al-smectite only represents a few percent, but its occurrence is genetically important. It is more common in diatom-rich intervals than in adjacent clayey intervals. Montmorillonite dominates, except in two samples (324 cm and 317 cm, proximal to the top of OIS 5e). On average, montmorillonite is more abundant in clay-rich intervals (~ 30%) than in diatom-rich ones (~ 15%). Beidellite is not always present (e.g., at 736 cm although the smectite content is highest at this depth); its content systematically increases up to 10% within diatom-rich intervals, and reaches exceptionally 15% (at 317 cm, proximal to the top of OIS 5e). The highest beidellite content is observed at the base of the core (at 820 cm) where this component accounts for 70% of smectites. This beidellite was formed by transformation. The total smectite/illite ratio (S/I), i.e., 17 Å/10 Å height ratio measured in EG by grouping $(10\text{-}14Sm)_{17}$, $(10\text{-}14Sm)_{Al}$ and Al_{17}, is shown in Fig. 6a together with variations in the smectite composition.

3.3.2. Core VER98-1-14

Variations in different weathering indices in Core VER98-1-14 are shown in Fig. 5b. As in Core VER98-1-3, a high *v*/*p* Biscaye index of smectite and fresh chlorite usually characterizes the clay-rich intervals. The general evolution of the illite crystallinity is not regular from diatom-rich to diatom-poor intervals, and the contrasts between the two lithologies are consequently less obvious than in core VER98-1-3. For instance, open and moderately weathered illites are reported in diatom-poor intervals OIS 2 and 6. We note that the freshest illites (crystallinity < 0.6° 2θ) systematically appear in the diatom-rich intervals (rather than in the diatom-rich ones for core VER98-1-3).

The mean clay assemblage is close to the clay composition of Core VER98-1-3 for illite (40%) and total smectite (28%), but it is characterized by a higher content of chlorite (15%) and 10-14c (8.5%) and a lower content of kaolinite (8.5%). The smectite group is composed of 70% montmorillonite, 20% Al-smectite and 10% beidellite. Within the whole clay assemblage, the montmorillonite varies around a mean value of 19%, with the lowest values observed during the diatom-rich intervals (Fig. 6b). By contrast, both Al-smectite and beidellite increase, in parallel, especially during the diatom-rich intervals. Their mean cumulative contribution increases from a mean value of 8.5% in clay-rich intervals, up to 15-20% in diatom-rich intervals.

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[2] All clay mineralogical data will be available on the web in the CONTINENT database (http://continent.gfz-potsdam.de) or upon request from the first author.

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4. Discussion

We organize the discussion in three parts. First we comment on the origin and composition of the material delivered to the lake, especially on the Academician Ridge. Second we focus on the origin of the clay minerals. Third we follow the evolution of the clay assemblage through the glacial/ interglacial intervals in order to identify a climate proxy.

4.1. Origin of the material delivered to the lake

4.1.1. Main sediment tributaries

Principal sediment sources for Lake Baikal are from its eastern side, with subsidiary sources to the north. Along the eastern flank, significant sediment transport along the Barguzin and Selenga river systems (Fig. 1) has created large delta complexes (Moore et al., 1997; Back et al., 1998, 1999), although the present main sediment source is the Selenga River, which is characterized by a large sublacustrine mud fan which progrades up to 65 km into the Central Basin (Nelson et al., 1999). Barguzin is a paleodelta, comprising Late Miocene-Early Pliocene sediments (Kaz'min et al., 1995). At the northern end of the lake, the Angara River (see Fig. 1) is the other major source of sediment. Some mineralogical discrepancies between the two cores could be explained by their location relative to these main tributaries.

In Core VER98-1-3, the higher feldspar and quartz content could reflect more proximal supplies from the Barguzin area. Also the supplies from Barguzin may not even reach the location of Core VER98-1-14 due to a north-south active fault located between the two studied cores (see line 8, fig. 1 in Hutchinson et al., 1992).

4.1.2. Geology of the Lake Baikal area

The northwest margin of Lake Baikal is mainly composed of Cambrian sandstones and limestones of the Siberian platform (Fig. 1; Galazii, 1993). They are surrounded to the SE (i.e., along the northern flank of the lake) by a belt of Proterozoic metamorphic schists and quartzites. The southeast margin consists of Proterozoic and Archean granites and granitoids. Metamorphic rocks including schists, gneisses, or amphibolites occur locally. Sedimentary rocks are represented by a belt of Jurassic and Cretaceous sandstones and claystones with coal outcropping within the Selenga River watershed.

Volcanic activity is related to the late Cretaceous-Paleocene evolution of the eastern Siberia rift system (Rasskazov, 1994). Lake Baikal is surrounded to the SW and the NE by three volcanic fields, the Tunka, South Baikal and Eravna basins (see Fig. 1). Sixty meters of basalts interbedded with sediments were formed in the Tunka Basin. In the South Baikal Basin, basaltic lavas did not outcrop in the Selenga delta but have been mapped 100-200 km south of Lake Baikal. In the Eravna Basin, ~ 250 m of late Cretaceous and Paleocene-Eocene sediments and volcanogenie deposits have accumulated. They also outcropped on the Vitim plateau to the northeast of the Eravna Basin.

4.1.3. Sedimentation conditions on the Academician Ridge

Paleoclimate reconstructions require a continuous sedimentary record, with no lacune or post-depositional reworking. Most cores until now were recovered in Lake Baikal on intra-basin highs, like the Academician Ridge, where hemipelagic sedimentation occurred, undisturbed by turbidite processes. The observed sedimentological features, in the two cores, which are separated by a distance of ~ 50 km, emphasize that the sediment accumulation rate on the Academician Ridge was not uniform during the last 250 kyr. For instance, sharp erosive contacts at the base of diatom-rich intervals show periods of non-deposition, the occurrence of early diagenetic vivianite concretions suggests periods of low sedimentation rates, and the bioturbation destroys the lamination in clayey layers. Furthermore, in Core VER-98-1-3, repeated small centimetric-scale slumps or gently inclined stratification may indicate intense sediment reworking by for example bottom currents. Core VER98-1-14 seems to be less affected by such sediment reworkings, except in its upper part. According to seismic profiles, sediment deposition on the Academician Ridge must have been highly variable from one site to another due to complex morphological features (Mats et al., 2000): several pronounced unconformities were recognized in seismic profiles on top of the ridge (Scholz et al., 1993); the structural scheme of the ridge displays a complicated fault pattern, with evidence of (re-)activation in the Late Pleistocene (Kaz'min et al., 1995); the variable thickness of sediment units was interpreted by a winnowing of the crests by bottom currents and a subsequent redeposition of sediments in adjacent depressions (De Batist, 1999).

4.2. Origin of the clay minerals in the Academician Ridge sediments

To identify a climate proxy in the clay mineral assemblage, we must determine the formation processes of the clays and make the point between the detrital and the neoformed clays. For paleoclimate reconstruction, only detrital clays are an indicator of weathering conditions, namely the hydrolysis conditions, within the watershed. However, clays (even detrital) reworked from older sedimentary rocks have no significance for recent climate reconstruction.

4.2.1. Detrital clays

In recent lacustrine sediments, clay minerals result predominantly from detrital input which expresses the weathering conditions at the source as well as the effects of dispersion processes (Chamley, 1989). Among the detrital clay minerals, illite and chlorite are considered primary minerals (Weaver, 1989). According to the geology of the Lake Baikal area, they could be derived from physical erosion of metamorphic parent rocks. In addition, illite could also be formed by weathering of non-layer silicates like feldspars from granites under moderate hydrolysis conditions or it could be derived from degradation of micas. In Cores VER98-1-3 and VER98-1-14, opening of the illite interlayers and partial vermiculitization of chlorite document a moderate weathering of parent minerals within the watershed (Thorez, 1985). In both cores, (10-14c) most probably reflects a partial chloritization (by insertion of Al-hydroxyls) affecting distended interlayers of illite or biotite (Thorez, 1985).

Pedogenic smectite and kaolinite are considered secondary minerals, derived through chemical weathering of parent alumino-silicates (e.g., feldspar, mica) or ferro-magnesian silicates (e.g., pyroxene, amphibole) under warm and humid conditions (Pédro, 1976; Weaver, 1989). Smectite is formed in confined environments, by recombination of released cations. Kaolinite is formed after a rapid leaching of cations, for instance on steep slopes. Soil-derived smectites display a composition ranging from between montmorillonite and beidellite, with a high content of octahedral bound Fe^{3+} (> 20%, Wilson, 1987). Such smectites originate from a transformation process (Robert and Barshad, 1972), and can be identified by the collapse of interlayers after K saturation. Moreover, Al-smectite testifies to the onset of a more intense secondary pedogenic chloritization affecting the distended interlayers of a former smectite.

4.2.2. Neoformed clays

In Lake Baikal sediments, clays and smectites in particular could be a product of neoformation by (a) dissolution of diatoms or (b) weathering of volcanic ashes.

(a) Baikal sedimentary conditions may be favorable for neoformation of smectite due to diatom dissolution at the sediment/water interface (e.g., Mackay et al., 1998). Released silica may contribute to the formation of nontronite (e.g., Lake Malawi, Millier and Forstner, 1973; Lake Washington, Jones and Browser, 1978, p. 210) or stevensite (e.g., saline lakes in Bolivia, Badaut et al., 1983; Lake Chad, Carmouze et al., 1977). However, in Core VER98-1-3, no nontronite or stevensite has been evidenced, suggesting that such neoformation of smectite has not taken place.

(b) Basalts and volcanogenic deposits outcrop along the SE margin of Lake Baikal (see Fig. 1). The occurrence of volcanogenic-derived smectite in sediments will be marked by a change in the mineralogical, physical, and/or geochemical properties. In Core VER98-1-3, K saturation evidences a fraction of neoformed smectite (i.e., a smectite fraction not affected by K saturation and preserving its expandability upon glycolation/glycerolation). For instance, the highest smectite content (> 50%) with a pure montmorillonitic composition at 736 cm may be compatible with a volcanic origin. In addition, two other levels (at 129 cm and 812.3 cm) dominated by a neoformed montmorillonite, almost pure in composition (with 0-5% beidellite), may also reflect weathering of volcanic material. However, volcanic glass in the more than 20-µm fraction has not been recognized with a light microscopic (E. Juvigné, pers. commun.).

4.3. Last climate cycle variability: a climate proxy supplied by clays?

The smectite abundance or smectite/illite (S/I) ratio is usually taken as a proxy for chemical weathering rate in the watershed. Smectites are usually considered strictly as a product of pedogenetic weathering, with their abundance being indicative of warm and wet climate conditions (Chamley, 1989; Weaver, 1989). In contrast, the abundance of a well-crystallized illite should increase during colder or drier conditions.

In the case of Late Quaternary Lake Baikal sediments, the S/I ratio includes several components of smectite, such as pedogenically derived material, reworked sedimentary rocks and, possibly, volcanogenic minerals. Within the smectite group, only transformation smectites and neoformed beidellite result from soil formation in the watershed and are therefore relevant for climate reconstruction. Reworked smectite fraction inherited from older sedimentary rocks, or neoformed montmorillonite derived from volcanic ashes will have no climate significance. In the next sections, we will examine (a) the evolution of the clay mineral weathering indices, (b) the evolution of the smectite content and, (c) the evolution of the S/I ratio with depth. Our aim is to define a proxy for climate changes within the Baikal watershed.

4.3.1. Evolution of the weathering indices of clay minerals

In Core VER98-1-3, the mineralogical parameters measured on the X-ray scans, such as illite crystallinity, the *v/p* Biscaye index of smectite and the weathering stage of chlorite (Fig. 5a) emphasize that the clays are more degraded during diatom-rich intervals than in adjacent clayey intervals. For instance, except at the base of Core VER98-1-3, the illite crystallinity roughly follows SPECMAP although there are some places where it is out of phase. Note that the time interval between the period of weathering and that of sedimentation has to be taken into account for climate reconstructions based on clay minerals (Thiry, 2000): weathered material can accumulate during a period characterized by a less aggressive climate, although it was produced during previous more aggressive conditions.

In Core VER98-1-14, the *v/p* index of smectites and the weathering stage of chlorites also record a more pronounced weathering of the clay minerals during the warmer, diatom-rich intervals (Fig. 5b).

Significant differences in the structures of clay minerals between glacial and interglacial intervals have already been reported in two samples from BDP cores from Academician Ridge (Solotchina et al., 2002). In agreement with our data, the content of smectite layers in the studied interglacial sample is slightly higher than in the glacial one. Moreover, they observed that the chlorite did not contain smectite layers in glacial samples whereas it included 15% of smectite layers in interglacial ones. According to Solotchina et al. (2002), 'this observation might be indicative of more active weathering processes in Lake Baikal watershed during warm periods'.

4.3.2. Evolution of the smectite content

During the colder, clayey intervals, the formation of smectites by pedogenesis should be less active. As a consequence, the detrital material delivered to the lakes should be characterized by relatively lower values for smectite content and S/I ratios. However, in both Cores VER98-1-3 and VER98-1-14 on the Academician Ridge, there is no obvious or systematic decrease in the total smectite content within the glacial intervals. For instance in Core VER98-1-3, the lower smectite contents (~ 10%, at 324 cm and 349 cm) are observed within the richest diatom interval (related to OIS 5e). In Core BDP-93 (Fig. 1), on the Buguldeika Saddle, most intervals containing the highest smectite content have been identified as interglacial periods; conversely, illite and chlorite exhibit a relative increase during glacial periods (Yuretich et al., 1998). Although the smectite abundance is highly variable, it correlates with SPECMAP for the last 250 kyr, with maxima of smectite around 200-250 kyr, 100-150 kyr and during the Holocene.

The absence of any obvious evolution of the clay assemblage with core depth in Academician Ridge cores, unlike for Core BDP-93, may not be due to a different clay quantification method (see comparison between German and US estimates for Core BDP-93 material, in Yuretich et al., 1998). These differences in clay assemblage may rather reflect the different core locations relative to the main sediment source, i.e., the Selenga River. On the Buguldeika Saddle, the sedimentation is more affected by overflows generated from the Selenga River. The enrichment of smectite in the finest grain-size fraction, through transport and settling, could overprint, by dilution, the primary climatic signature within the detrital clay assemblage finally delivered at the Academician Ridge (i.e., located ~ 200 km from the Selenga River system) although the weathering rate of each clay species is preserved.

4.3.3. Evolution of the smectitelillite ratio

In Core VER98-1-3, the S/I ratio fluctuates by a factor of 4 throughout the core. On average, the S/I ratio displays the highest values during or near the termination of the diatom-rich intervals or just shortly after, except for OIS 6. In detail, the S/I curve does not perfectly match the glacial/interglacial cycles: for instance, the lowest ratio (< 0.5) characterizes the diatom-richest interval equivalent to OIS 5e. During glacial OIS 6, the relatively high smectite content and S/I ratios record a high supply of montmorillonite to the ridge. This could be explained by active erosion of inherited clays from reworked sediments. The greater sediment thickness of OIS 6 in Core VER98-1-3, as compared with Core VER98-1-14, contrasts with similar thicknesses for OIS 5 in both cores. This discrepancy could be explained by enhanced erosion during glacial OIS 6 of the Barguzin paleodelta. The interpretation of the S/I ratio in terms of paleoclimate changes has then to be treated with caution.

In Core VER98-1-14, the S/I ratio varies by a factor of 6 (0.5-3, Fig. 6b). A rough correspondence between the S/I ratio and SPECMAP may be observed. The highest ratios usually occur during the interglacial diatom-rich intervals. In those intervals, the composition of the smectite group changes, characterized by lower contributions from montmorillonite and higher contributions from Al-smectite and beidellite. Within Core VER98-1-14, the high S/I ratios measured during OIS 6 are not associated with high weathering indices of illite, chlorite or smectite (Fig. 5b). The increase is only due to higher supply of montmorillonite. We propose that the S/I evolution has only a climate significance if its evolution is related to changes in the weathering indices of the clays.

5. Conclusion

(1) The composition of the hemipelagic Late Quaternary sediments varies along the Academician Ridge. In its northern part (Core VER98-1-3), the mean composition of the sediments is 34% feldspar, 23% quartz, 26% biogenic opal (diatom) and only 17% clay minerals. Southwards (Core VER98-1-14), the sediments are dominated by clays (59%) with 20% feldspar, 15% quartz and 6%, on average, of opal. The increased proportions of feldspars and quartz at Core VER98-1-3 may be due to the proximity of the Barguzin River.

(2) On average, the clay assemblage of Core VER98-1-3 is made up of 40% illite, 28% illite— smectite mixed layers (3% of Al-smectites included), 16% of fresh and/or degraded chlorite accompanied by (14c-14v) and (10-14c) and 16% kaolinite. In Core VER98-1-14, the clay assemblage is characterized by a higher contribution (24%) of chlorite and (10-14c) counterbalanced by a relatively lower amount of kaolinite (9%) and by a higher contribution of Al-smectite (7%).

(3) Since ~ 250 kyr, the weathering indices of the clay minerals record the influence of glacial/ interglacials stages: open illites, weathered chlorites, illite-smectite mixed layers with a low contribution of smectite layers are more common in interglacial stages (due to more chemical weathering under warm climate) than in glacial intervals.

(4) The evolution of the smectite/illite ratio is not systematically correlated with the glacial/interglacial stages: the correlation is good in Core VER98-1-14 but poor in Core VER98-1-3. This probably reflects the complex composition of the smectite group (montmorillonite, beidellite, Al-smectite) and its different origins (transformed or neoformed). In both cores, the smectite group is dominated by montmorillonite (mean 19%), a mineral of multiple origins (pedogenetic volcanic, or reworked). The climate significance of the S/I ratio is probably partly erased by the broad fluctuations of the montmorillonites in Core VER98-1-3. In Core VER98-1-14, the montmorillonite is more stable. In each interglacial interval in Core VER98-1-14, the S/I ratio increases in parallel with a systematic and significant higher contribution of both beidellite and Al-smectite, i.e., two minerals formed by pedogenesis. In Lake Baikal sediments, the S/I ratio could be interpreted as a climate proxy only if its fluctuation is related to changes in clay weathering indices and changes in the smectite composition.

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