

Inorganic geochemistry of El'gygytgyn Lake sediments (northeastern Russia) as an indicator of paleoclimatic change for the last 250 kyr

P. S. Minyuk · J. Brigham-Grette · M. Melles ·
V. Ya. Borkhodoev · O. Yu. Glushkova



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Abstract The inorganic geochemistry of sediments from El'gygytgyn Lake shift in phase with interpreted paleoclimatic fluctuations seen in the record over the past 250 ka. Warm periods, when the lake was seasonally ice free and fully mixed, are characterized by increased concentrations of SiO₂, CaO, Na₂O, K₂O, and Rb, by decreased contents of TiO₂, Fe₂O₃, Al₂O₃, and MgO, and by a lower chemical index of alteration (CIA). Increased levels of SiO₂ reflect increases in limnic productivity whereas many of the other elements

and the CIA likely reflect increased hydrological activity coincident with an increase in coarser sand and silt content and a decrease in clay mineral content. For cold/cooler periods when perennial lake ice cover lead to a stratified water column and anoxic bottom waters, the opposite is generally observed suggesting a decrease in hydrological activity and an increase in post-depositional chemical alteration.

Peaks in P₂O₃ and MnO, coincident with an increased abundance of vivianite, suggest possible linkages to the paleoproductivity of local fish fauna regardless of climate change across the region surrounding Lake El'gygytgyn. Strontium is high in concentration during warmer intervals and may also be linked to paleoproductivity. Enrichment of the post-Eemian portion of the sediment record in niobium, and yttrium appears independent of glacial–interglacial change; rather it may reflect a gradual shift in the geomorphology of the catchment, particularly the hydrology of large alluvial fans along the western side of the lake. In contrast to some lake records, changes in Zr concentration over time suggests only a weak, if any, increase in eolian sediment supply during colder periods.

This is the *ninth* in a series of eleven papers published in this special issue dedicated to initial studies of El'gygytgyn Crater Lake and its catchment in NE Russia. Julie Brigham-Grette, Martin Melles, Pavel Minyuk were guest editors of this special issue.

P. S. Minyuk (✉) · V. Ya. Borkhodoev ·
O. Yu. Glushkova
North-East Interdisciplinary Science Research
Institute, FEB RAS, 685000 Magadan, Russia
e-mail: Minyuk@neisri.magadan.ru

J. Brigham-Grette
Department of Geosciences, University of
Massachusetts, Amherst, MA 01003, USA
e-mail: juliebg@geo.umass.edu

M. Melles
Institute for Geophysics and Geology, University
Leipzig, D-04103 Leipzig, Germany
e-mail: melles@rz.uni-leipzig.de

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Introduction

The El'gygytyn impact crater, located in central Chukotka, northeastern Russia (67°30' N; 172°05' E), was created 3.6 Ma in Upper Cretaceous volcanic rocks (Fig. 1) (Gurov and Gurova 1981, 1982; Belyi and Raikевич 1994; Layer 2000). The catchment surrounding the crater consists primarily the ignimbrites and tuffs of the Pykarvaam Formation. The Voronin (ignimbrites, tuffs), Koekvun (andesite-basalts, tuffs, tuffaceous sands) and Ergyvaam (tuffs, ignimbrites) formations are also present but not as prevalent in

the catchment area (Belyi et al. 1998; Belyi and Raikевич 1994). The Pykarvaam Formation is found surrounding the lacustrine basin to the north, west, and south (Belyi and Raikевич 1994). Chemical and physical weathering products from these rocks make up the majority of clastic deposition into the lake.

In 1998, a 12.9 m sediment core (PG1351, Brigham-Grette et al. 2007) was taken from the center of El'gygytyn Lake (Fig. 1). The purpose of studying this pilot core was to investigate the potential of this Arctic lake system as a recorder of millennial scale paleoclimatic change some

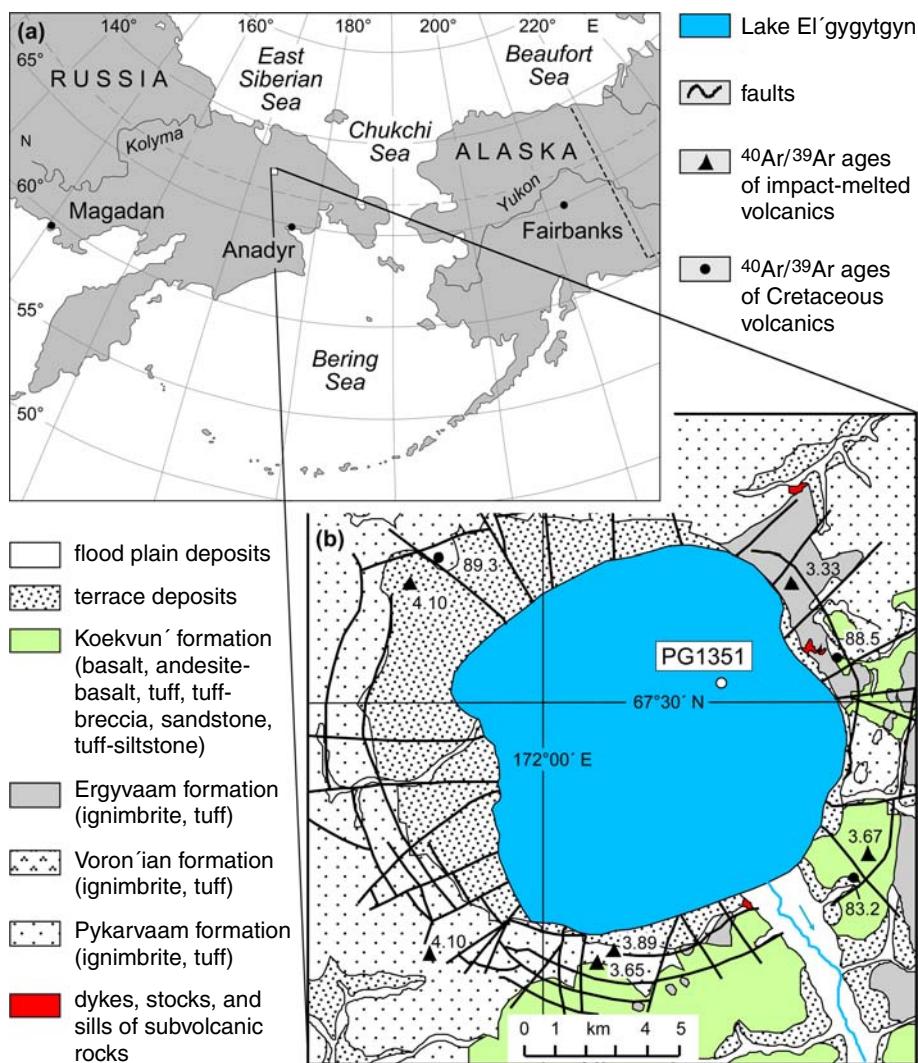


Fig. 1 Location and bedrock maps of El'gygytyn Lake in the Anadyr Mountains of Chukotka, Russia (Belyi and Raikевич 1994, modified from Nowaczyk et al. 2002)

100 km north of the Arctic Circle. The purpose of measuring the inorganic composition of core PG1351 was to explore new information on the climatic and environmental history of Lake El'gygytgyn during the past 250 ka. The core is well dated as outlined in Nowaczyk et al. (2007).

Methods

Along the length of core PG1351, samples for geochemical analysis were taken every 5–7 cm. The chemical composition and major elements in the sediments were analyzed using a multi-channel X-ray fluorescence (XRF) spectrometer SRM-25 (USSR). Dry sediment samples were fused with lithium tetroborate ($\text{Li}_2\text{B}_4\text{O}_7$) flux, in a mass ratio of 1:1. The alloy was then ground and layered with a boric acid base, and then exposed to a radiation source. X-ray fluorescence of $K\alpha$ -lines for the determined elements were generated by X-ray tube radiation with an Rh-anode at 50 kV and 40 mA. The measurement time was 100 s. Elemental compositions were determined using the fundamental parameters method (Borkhodoev 1999).

Rare elements were determined by the XRF spectrometer VRA-30 (Germany). Sediment powders were put into a cup with a 25 μm thick mylar window. The intensity of the Rb, Sr, Y, Zr and Nb $K\alpha$ -lines were generated by radiation of the X-ray tube with an Rh-anode at 50 kV and 40 mA. The elemental composition was found using the standard-background method. The Compton scattered $K\alpha$ -lines of the X-ray tube target were used to correct matrix effects and determine the background of radiation (Borkhodoev 1998). The lower detection limits of elemental determination by this method were: Rb, Y, Sr, Zr—4 ppm, and Nb—3 ppm.

Scientific context and background

Insolation-tuning of magnetic susceptibility, total organic carbon and TiO_2 , together with pollen stratigraphy, luminescence and radiocarbon dating have shown that core PG 1351 from Lake El'gygytgyn continuously spans the last 250 ka

(Nowaczyk et al. 2007; Forman et al. 2007; Lozhkin et al. 2007). A number of proxies from sedimentology, organic geochemistry, and mineralogy (Nowaczyk et al. 2002; Asikainen et al. 2007; Cherepanova et al. 2007) clarify the relationship between changes in limnology and regional climate. These data have been interpreted to suggest that the history of the lake system reflects at least four climate modes controlled by both changes in high-northern latitude insolation and circumpolar atmospheric circulation (Melles et al. 2007).

The modes outlined by Melles et al. (2007) reflect past environmental conditions associated with relatively warm, peak warm, cold and dry, and cold but moist climate. A relatively warm climate resulted in complete summer melt of the lake ice cover and seasonal mixing of the water column. This mode prevailed during the Holocene and Marine Isotope Stages (MIS) 3, 5.1, 5.3, 6.1, 6.3, 6.5, 7.1–7.3, 7.5 and 8.1. MIS 5.5 (Eemian) was characterized by significantly enhanced aquatic primary production and organic matter supply from the catchment, suggesting peak warm conditions throughout the region. During MIS 2, 5.2, 5.4, 6.2 and 6.4 the climate must have been cold and dry, leading to perennial lake ice cover, little regional snowfall, and a stagnant water body. A cold but more moist climate during MIS 4, 6.6, 7.4, 8.2 and 8.4 is thought to have produced more snow cover across the perennial ice that covered the lake even in summer. This led to strongly reduced light penetration and decreased biogenic primary production in the lake.

Results and discussion

Major elements

All major elements with the exception of P_2O_5 and MnO show significant changes in concentration throughout the 250 ka record (Fig. 2). For instance, the position of the Pleistocene/Holocene boundary at ~80 cm core depth (Nowaczyk et al. 2007) can be easily determined by a major shift in the magnitude of these data. Sediments deposited during warm periods are characterized by relatively high concentrations of CaO , Na_2O , K_2O

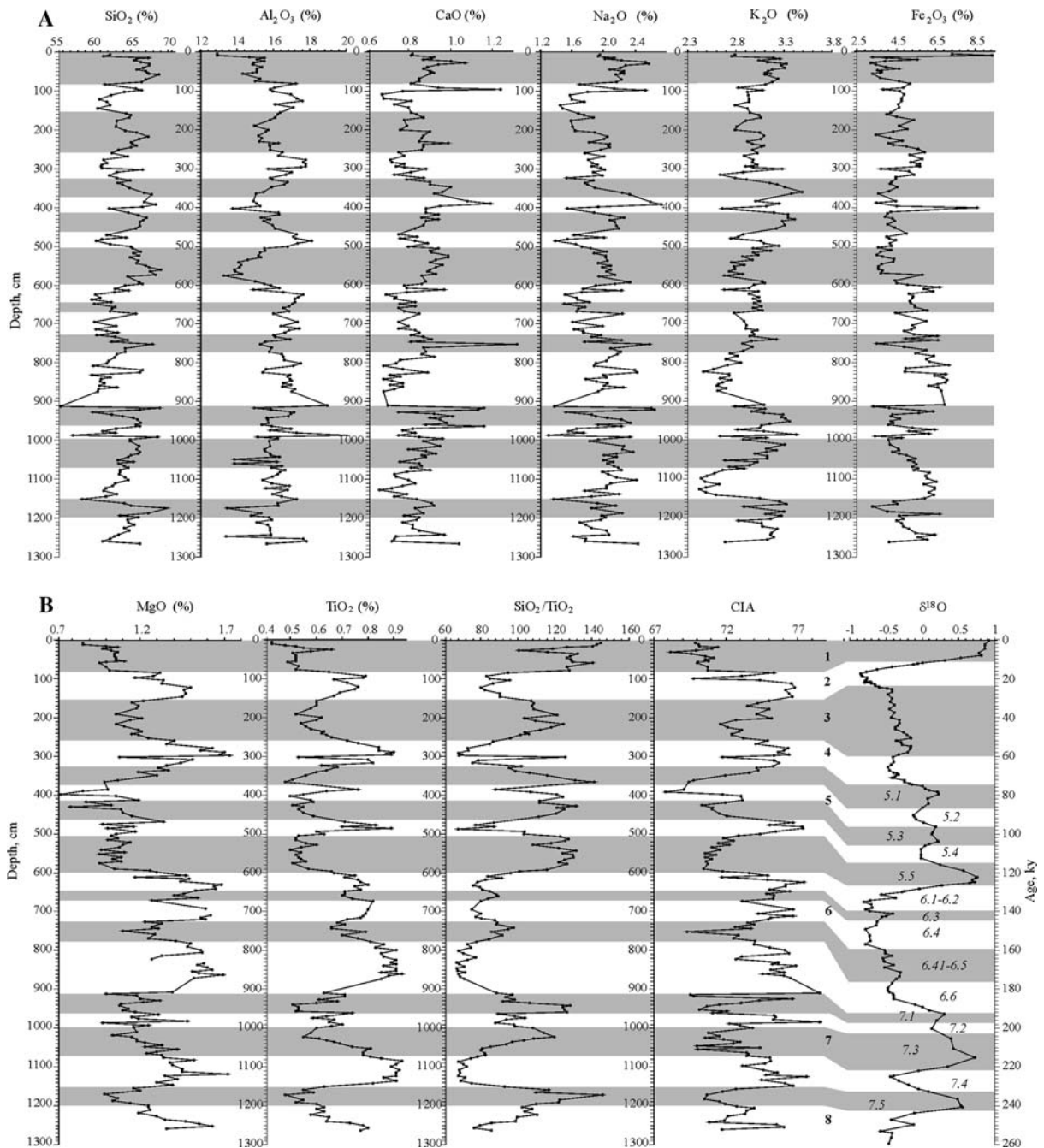


Fig. 2 Content of major elements in core PG1351 (A). Correlation of MgO and TiO₂ content, SiO₂/TiO₂, CIA with oxygen isotope data by (Martinson et al. 1987) (B).

Gray areas indicate intervals of warm diatom assemblages. Stages are marked by bold numbers, sub-stages, by italicized numbers

and particularly SiO₂, and by relatively low amounts of TiO₂, Al₂O₃, MgO, Fe₂O₃. This pattern is controlled by biogenic production in the lake as well as by weathering and sediment supply

from the catchment, both of which are controlled by paleoclimatic change.

During warm stages, massive muds with high diatom concentrations, high illite and smectite

contents, and high magnetic susceptibilities were deposited (Asikainen et al. 2007, Cherepanova et al. 2007, Nowaczyk et al. 2007). The enhanced diatom and illite flux to the lake bottom is clearly mirrored by enriched SiO_2 contents in the same sediments (Fig. 2A). The predominance of illite is also reflected by high K_2O , Na_2O , CaO contents.

During cold stages, laminated muds were deposited, which exhibit relatively low opal contents and maxima in chlorite concentrations (Asikainen et al. 2007; Cherepanova et al. 2007; Melles et al. 2007). The former is reflected by low SiO_2 contents in the sediments (Fig. 2A). Indicators of higher chlorite content comes from high MgO and Fe_2O_3 values in the respective sediments (Fig. 2B) because magnesium and iron are two of the major elements in chlorite (Brownlow 1996). In addition, titanium may occur as an elemental replacement in clay minerals, explaining the enhanced TiO_2 contents in sediments formed during cold intervals. The positive correlation between Fe_2O_3 and TiO_2 (Fig. 3) has a coefficient of correlation (C) of 0.68 (corresponding to an R^2 value of 0.465). According to mineralogical data, detrital magnetite and ilmenite dominate the heavy mineral extracts from the lake sediments (V. Smirnov, personal communication 2005). The presence of pure magnetite is supported by X-ray analyses (Minyuk et al. 2003) as well as low-temperature measurements of magnetic susceptibility (Nowaczyk et al. 2002) of the lake sediments. These data suggest that titanium is not associated with magnetic minerals, otherwise it would be in high concentration during warm periods. In fact, what we observe is that titanium is low during warm periods (Fig. 2B). Therefore, only a small quantity of total titanium can constitute any part of the detrital ilmenite concentration.

A generally good correlation also exists between SiO_2 and Al_2O_3 ($C = -0.82$; $R^2 = 0.666$), TiO_2 and MgO ($C = 0.77$; $R^2 = 0.588$), K_2O and TiO_2 ($C = -0.74$; $R^2 = 0.543$), Al_2O_3 and TiO_2 ($C = 0.73$; $R^2 = 0.529$), SiO_2 and TiO_2 ($C = -0.71$; $R^2 = 0.508$), SiO_2 and MgO ($C = -0.78$; $R^2 = 0.610$), MgO and CaO ($C = -0.66$; $R^2 = 0.429$) (Fig. 3). Assuming these elements are representative of the chemical composition of the clay mineral assemblages in sediments, the concentration of these elements

(and various ratios between them) can be effectively used and compared to create a time-series of changing paleoenvironments, including the weathering of rock in the catchment, transport processes, as well as diagenetic processes. Many of these elements and ratios are routinely used as paleoenvironment proxies in marine sediments (Wei et al. 2004; Li et al. 2003; Zabel et al. 2001; Hinrichs et al. 2001; Richer et al. 2000; Arnaboldi and Meyers 2003), lake sediments (Benson et al. 1998), and loess sequences (Jahn et al. 2001).

A select group of these major elements are routinely used to quantify the degree of alteration utilizing the “chemical index of alteration”, where $\text{CIA} = [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})] * 100$ (Nesbitt and Young 1984, 1989; Fig. 2B). This index provides an objective, relative measure of the degree to which rocks and sedimentary components are altered in any way while stored on the landscape, transported to the lake; this index also includes a measure of any post-depositional diagenesis. We determined that concentrations of SiO_2 , CaO , Na_2O , and K_2O are higher in the surrounding catchment volcanic rocks than in the lake sediments (Table 1). CIA values for the sediments recording warm climate modes are higher than the index for either rocks from the Pykarvaam Formation or beach pebbles. Even higher CIA values for cool climate modes suggests that while some chemical alteration is occurring today, the process of alteration was even more intense during past cold periods.

Lake Elgygytyn is located in the zone of hypoaerctic tundra (Yurtsev 1973) where weathering is largely controlled by the presence or absence of permafrost. The modern climate is cold (Nolan and Brigham-Grette 2007) and during glacial stages it is estimated to have been much colder (Anderson and Lozhkin 2002). Under such conditions mechanical weathering dominates the long term breakup of rocks. These exceptional climatic conditions and short summers, coupled with the high relief of the catchment and short transport distance, conspire to inhibit much chemical weathering. Chlorite, in general, is a weathering product of cold, high latitude climates (Singer 1984) representing cold (glacial) intervals in some sections (Cagatay et al. 2002; Vanderaverroet et al. 1999; Lamy et al. 1999). In Lake

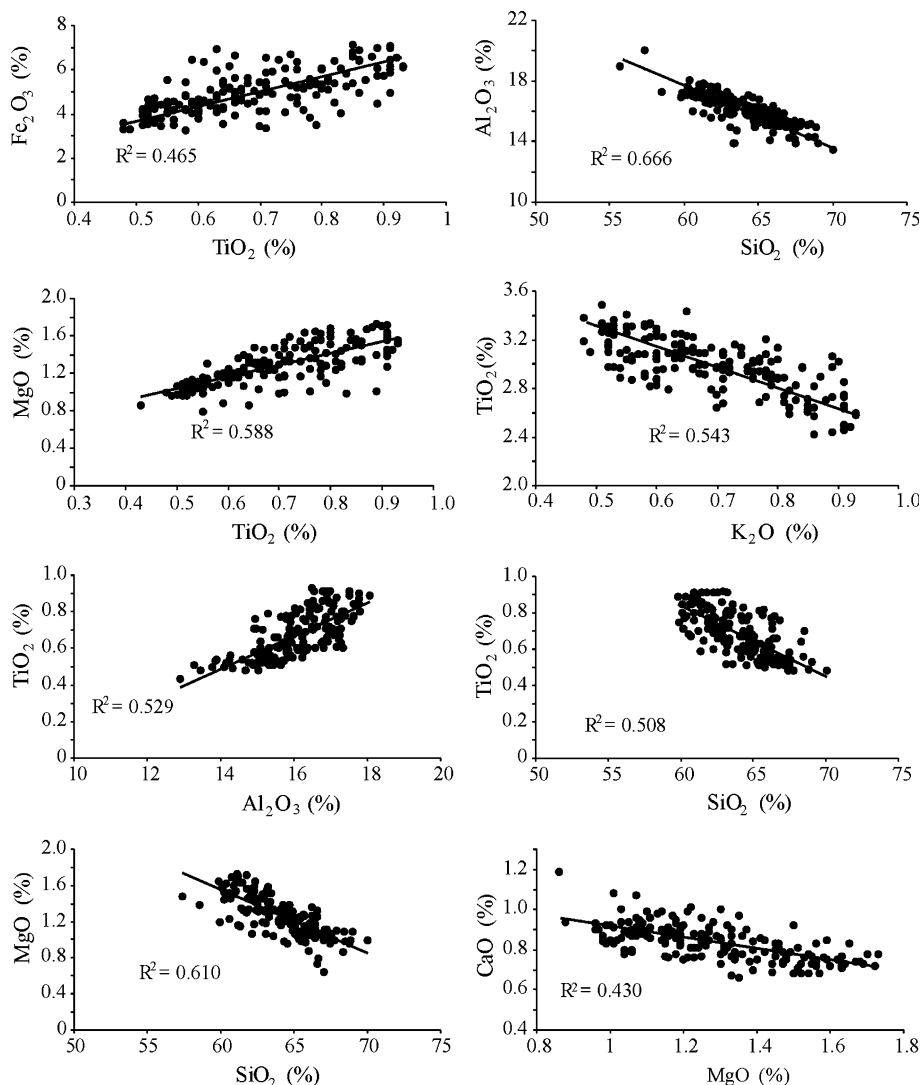


Fig. 3 Scatter plots of Fe_2O_3 versus TiO_2 ; Al_2O_3 versus SiO_2 ; MgO versus TiO_2 ; TiO_2 versus Al_2O_3 , SiO_2 , K_2O ; MgO versus SiO_2 ; CaO versus MgO

Table 1 Chemical composition (%) of rocks and sediments of El'gygytgyn Lake

Rocks, sediments	<i>N</i>	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	CIA
Pykarvaam formation ^a	11	72.57	0.29	14.92	2.60	0.06	0.47	1.59	3.53	3.90	0.06	63
Beach Pebbles ^b	11	70.34	0.31	14.28	3.51	0.07	0.65	1.38	2.99	4.08	0.09	62
Sediment from warm MIS	97	65.54	0.59	15.44	4.53	0.08	1.12	0.90	2.07	3.07	0.14	72
Sediment from cool MIS	89	62.58	0.77	16.60	5.50	0.07	1.41	0.79	1.87	2.88	0.16	75

CIA values averaged from Fig. 2b

N = Quantity of analyses

^a Belyi (1998)

^b Feldman et al. (1980)

Elgytgyn, chlorite is found to be more abundant in sediments deposited during cold intervals, consistent with more active mechanical weathering of bedrock (Asikainen et al. 2007). Even during the Eemian, the only period during the past 250 ka when climate was significantly warmer than present, chlorite was enriched in the sediments and smectite, as an index of chemical weathering (Desprairies and Bonnot-Courtois 1980; Chamley 1989), remained low.

Since the marked differences in the CIA at the stadial–interstadial level (Fig. 2B) are not due to changes in chemical weathering, the alteration must involve depositional and post-depositional processes related to diagenesis. During cold stages sedimentation was largely controlled by stratification of the water column and anoxic bottom waters. This led, for instance, to enhanced organic carbon, nitrogen and sulphur deposition and to the dissolution of magnetite (Nowaczyk et al. 2002; Melles et al. 2007). Hence the addi-

tional alteration of clay minerals can be expected. An alternative explanation or additional effect could be the eolian influx of substantially altered materials from a distal source providing more weathered material than the surrounding bedrock during cold periods.

Among the major elements, a direct correspondence is observed between P_2O_5 and MnO (Fig. 4). Phosphorus, for example, eroded from the surrounding volcanic rocks and released from soils is readily utilized by fish populations living in the lake. Today, the lake is rich with endemic *Salvelinus boganidae* Berg., *S. elgyticus* Viktorovsky et Glybokovsky, and *Salvelymis svetovidovi* Chereshev et Skopetz (Chereshev and Skopets 1993). We hypothesize that as fish bones decompose on the floor of the lake, this creates favorable conditions in the sediment porewaters for vivianite ($(Fe)_3(PO_4)_2 \cdot 8H_2O$) diagenesis, a process geochemically preferred in the absence of sulphur to produce pyrite. Vivianite was first

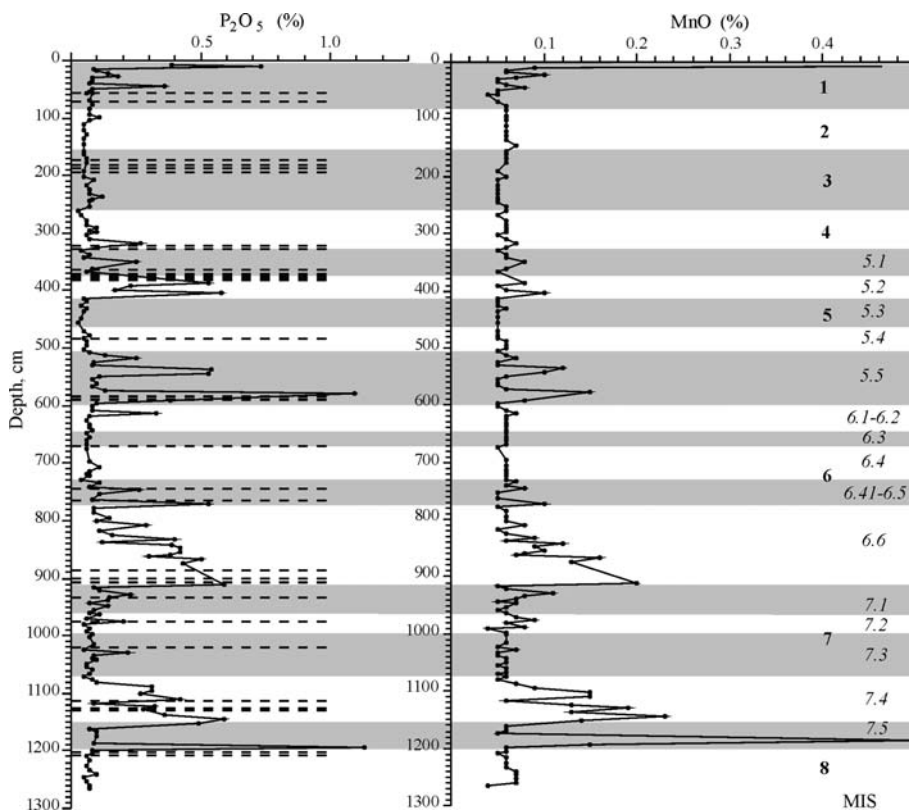


Fig. 4 Content of P_2O_5 and MnO in core PG1351. Gray areas indicate intervals of warm diatom assemblages. Stages are marked by bold numbers, substages, by italicized numbers

identified optically and found distributed throughout the core, regardless of inferred climate mode (Fig. 4); mineral identification was later confirmed by XRF measurements (Asikainen 2004). It typically occurs as rounded aggregates and rarely as crystals (Minyuk et al. 2003; Asikainen et al. 2007). A thermogram of these aggregate grains is characterized by an endothermal effect at 100–305°C. Water is removed from the mineral lattice and the exothermal peaks are present at 640°C and 790°C when iron oxidation occurs. Electron microprobe analysis shows that manganese is a minor component of vivianite (Table 2). The P₂O₅/MnO ratio in the studied sediments is close to that of vivianite (Fig. 5).

Phosphorus peaks are clear along the length of the core when vivianite is present as a diagenetic product requiring anoxic conditions (Fig. 4). Because vivianite occurs independent of climate mode, it must be formed in the sediments regardless of changes in the oxygenation of the bottom waters. Rather, its occurrence is controlled by the diagenetic microenvironment (Stoops 1983; Stamatakis and Koukouras 2001). If sufficient quantities of phosphorus or other organic matter exists even in oxic environments, then anoxic microaureoles could form at any

time. Such conditions are favorable for the growth of vivianite aggregates, as commonly observed on organic detritus (Stoops 1983), fossil animal bones (Piepenbrink 1989), and human skeletal remains (Mann et al. 1998). Hence, the occurrence of vivianite reflects times with a consistent supply of phosphorus and manganese, and is rather independent of long term biogenic primary production and accumulation. In addition, the common occurrence of vivianite over the past 250 ka supports the suggestion that the endemic fish fauna in Lake El'gygytyn probably survived glacial times (Chereshnev and Skopets 1993).

Trace elements

Among the trace elements, strontium and, less clear, zirconium and rubidium appear to shift in phase most clearly with the inferred climatically driven hydrologic and sedimentary history at Lake El'gygytyn. The volcanic rocks surrounding the lake are the direct sources of strontium, rubidium and zirconium in the sediments. High concentrations of strontium and rubidium, and low concentrations of zirconium, are characteristic of warmer modes as defined by Melles et al. (2007), while cooler periods are characterized by the reverse concentrations. This general tendency is observed along the entire core to 13 m (Fig. 6). A positive correlation also occurs between strontium and rubidium concentrations and the quantity of diatom valves in the sediment (cf., Cherapanova et al. 2007). This suggests that strontium and rubidium concentrations in the sediments are influenced by hydrological factors that also favor periods of increased productivity. Zirconium is enriched in several cold periods with the exception of MIS 2, 5.4, and 7.2. We have two possible explanations—first it may be enriched during times of high alteration in the catchment; alternatively it may be enriched as distally transported material during times of intense eolian activity (see above).

The concentrations of niobium and yttrium, in contrast, vary independently of the pattern described above. They remain low throughout the lower part of the core and increase up core significantly starting at a depth of about 300 cm (Fig 6). According to the available chronological information (Forman et al. 2007; Lozhkin et al.

Table 2 Chemical composition (%) of vivianite occurring in core PG1351

Grain	Analysis	Fe ₂ O ₃	P ₂ O ₅	MnO
I	1	50.45	33.65	1.50
	2	52.04	36.03	1.88
	3	53.42	37.58	1.98
II	1	50.99	37.73	5.57
	2	54.30	37.54	4.85
	3	54.59	36.94	2.78
III	1	52.84	36.03	2.44
	2	54.13	33.31	2.25
	3	51.82	35.81	2.43
IV	1	52.61	38.10	4.04
	2	52.85	36.88	5.61
	3	50.86	36.96	5.65
V	1	55.76	36.76	1.71
	2	55.11	36.62	2.24
	3	49.49	30.79	0.88
VI	1	52.61	36.93	4.87
	2	52.15	36.75	3.32
	3	51.18	35.85	4.10

Analyses were done on microprobe "Camebax" by M. I. Parfenov (NEISRI FEB RAS)

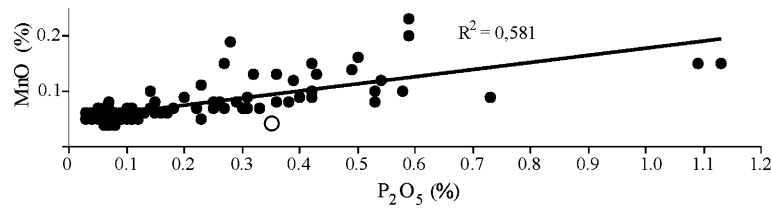


Fig. 5 Scatter plots of P₂O₅ versus MnO concentrations for sediment of core PG1351. Open symbol indicates the respective ratio of vivianite in this core

2007; Nowaczyk et al. 2007) this depth corresponds with the initiation of marine isotope stage (MIS) 4. At this time, the Upper Pleistocene silt complex (yedoma) that many researchers believe is of eolian origin (loess) began to form in north-eastern Russia (Tomordiaro 1980; Decision of interdepartmental stratigraphic conference on quaternary system of USSR 1982). The concentration of Y and Nb in volcanic rocks near the lake is 1.5–2 times lower than in sediments from the same interval (Belyi 1998). This could suggest that lake sediment enrichment by these elements might be associated with enhanced eolian sediment supply from areas far beyond the catchment. If this is true, then changes in Zr, as discussed above, cannot be due to eolian input but must be related to changes in alteration over time.

Conclusions

Inorganic geochemistry of core PG1351 confirms the general temperature-related changes in the catchment, water column and sediments of Lake El'gygytyn found to have occurred by sedimentological, biological and mineralogical proxies. This cold/warm character is reflected in primary production (SiO₂) and changes in alteration (especially as indexed by CIA based upon Al₂O₃, CaO, Na₂O, K₂O but also Fe₂O₃, MgO, TiO₂, Sr, Zr and Rb).

Precipitation changes in the region are reflected by hydrological changes in the crater basin. The inorganic geochemistry does not provide a direct proxy for the hydrological setting, but the direction of change in some indicators are consistent

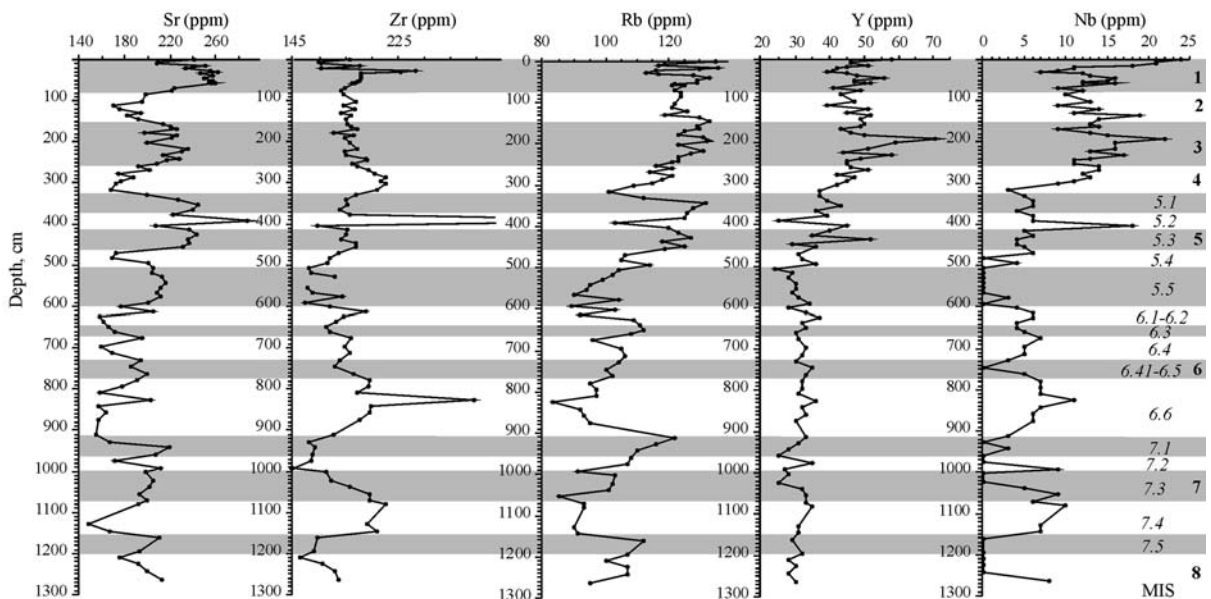


Fig. 6 Content of trace elements in core PG1351. Gray areas indicate intervals of warm diatom assemblages. Stages are marked by bold numbers, sub-stages, by italicized numbers

with inferred increases in sedimentation rate, e.g., the CIA index is lower during warmer periods when we infer more intense runoff and low residence time for sediments in transport to the lake.

Changes in distal eolian supply over time are suggested in the inorganic geochemistry data (Zr, Y, Nb); however, the results are inconsistent. The signal from the eolian proxies is most likely intertwined with complex changes in autochthonous vs. allochthonous accumulation and in post-depositional alteration.

Widely independent of climate change is the occurrence of vivianite. It occurs irregularly throughout the core given adequate supplies of phosphorus, iron, and organic detritus, thus indicating it is dependent on microenvironments in the sediments.

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