

1 **Sedimentological processes and environmental variability**
2 **at Lake Ohrid (Macedonia, Albania) between 637 ka and the**
3 **present**

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1 **Abstract**

2 Lake Ohrid (Macedonia, Albania) is thought to be more than 1.2 million years old and host
3 more than 300 endemic species. As a target of the International Continental scientific Drilling
4 Program (ICDP), a successful deep drilling campaign was carried out within the scope of the
5 Scientific Collaboration on Past Speciation Conditions in Lake Ohrid (SCOPSCO) project in
6 2013. Here, we present lithological, sedimentological, and (bio-)geochemical data from the
7 upper 247.8 m composite depth of the overall 569 m long DEEP site sediment succession
8 from the central part of the lake. According to an age model, which is based on 11 tephra
9 layers (1st order tie points) and on tuning of biogeochemical proxy data to orbital parameters
10 (2nd order tie points), the analyzed sediment sequence covers the last 637 kyrs.

11 The DEEP site sediment succession consists of hemipelagic sediments, which are interspersed
12 by several tephra layers and infrequent, thin (<5 cm) mass wasting deposits. The hemipelagic
13 sediments can be classified into three different lithotypes. Lithotype 1 and 2 deposits
14 comprise calcareous and slightly calcareous silty clay and are predominantly attributed to
15 interglacial periods with high primary productivity in the lake during summer and reduced
16 mixing during winter. The data suggest that high ion and nutrient concentrations in the lake
17 water promoted calcite precipitation and diatom growth in the epilimnion during MIS15, 13,
18 and 5. Following a strong primary productivity, highest interglacial temperatures can be
19 reported for marine isotope stages (MIS) 11 and 5, whereas MIS15, 13, 9, and 7 were
20 comparably cooler. Lithotype 3 deposits consist of clastic, silty clayey material and
21 predominantly represent glacial periods with low primary productivity during summer and
22 longer and intensified mixing during winter. The data imply that the most severe glacial
23 conditions at Lake Ohrid persisted during MIS16, 12, 10, and 6 whereas somewhat warmer
24 temperatures can be inferred for MIS14, 8, 4, and 2. Interglacial-like conditions occurred
25 during parts of MIS14, and 8.

26

27 **1 Introduction**

28 In the light of recent climate warming, it has become fundamentally important to understand
29 the characteristics and shaping of individual glacial and interglacial periods during the
30 Quaternary, as these differences can reveal information about external forcing and internal
31 feedback mechanisms in the global climatic system (Lang and Wolff, 2011). The global
32 glacial-interglacial variability has widely been studied on ice cores (e.g. EPICA-members,
33 2004; NGRIP-members, 2004) and on marine sediment successions (e.g. Lisiecki and Raymo,

1 2005). In terrestrial realms, long continuous paleoclimatic records are sparse and mostly
2 restricted to loess-paleosol sequences (e.g. Chen et al., 1999), to speleothem records (Bar-
3 Matthews and Ayalon, 2004; Wang et al., 2008) and to lacustrine sediments (e.g. Prokopenko
4 et al., 2006; Melles et al., 2012). In the eastern and southeastern Mediterranean region, long
5 terrestrial paleo-records have become available from Lake Van (Stockhecke et al., 2014a), the
6 Dead Sea (Stein et al., 2011), and from the Soreq Cave speleothem record (Bar-Matthews and
7 Ayalon, 2004).

8 In the central Mediterranean region, the only terrestrial paleo-record that continuously covers
9 more than one million years is the Tenaghi Philippon pollen record in northern Greece (cf.
10 Fig. 1), which spans the last 1.3 million years and provides valuable insights into the
11 vegetation history of the area (e.g. Tzedakis et al., 2006; Pross et al., 2015). The results from
12 Tenaghi Philippon reveal that individual interglacial and glacial periods in the Mediterranean
13 region can differ significantly in their duration and severity (e.g. Tzedakis et al., 2006;
14 Fletcher et al., 2013). However, analytical methods for paleoclimate reconstructions at
15 Tenaghi Philippon have so far been restricted to pollen analyses.

16 Lake Ohrid on the Balkan Peninsula is thought to be more than 1.2 million years old and has
17 already demonstrated its high sensitivity to environmental change and the potential to provide
18 high-resolution paleoenvironmental information for the last glacial/interglacial cycle (e.g.
19 Wagner et al., 2008; 2009, 2014; Vogel et al., 2010a). Given that Lake Ohrid is also a hotspot
20 for endemism with more than 300 endemic species in the lake (Föller et al., 2015), its
21 sediments also have the potential to address evolutionary questions such as what the main
22 triggers of speciation events are.

23 An ICDP deep drilling campaign took place at Lake Ohrid in spring 2013 using the Deep
24 Lake Drilling System (DLDS) operated by the Drilling, Observation and Sampling of the
25 Earths Continental Crust (DOSECC) consortium. More than 2100 m of sediments were
26 recovered from four different drill sites (Fig. 1). The processing of the cores from the DEEP
27 site from the central part of Lake Ohrid is still ongoing at the University of Cologne
28 (Germany). Here, we present lithological, sedimentological, and (bio-)geochemical results
29 from the upper part of the DEEP site sediment succession until 247.8 mcd (meter composite
30 depth). According to an age model, which is based on 11 tephrochronological tie points and
31 on tuning of biogeochemical proxy data against orbital parameters, the analyzed sequence
32 covers the period since 637 ka. Here, we aim to provide a chronological framework for the
33 deposits, confirm the completeness of the record, provide first insights into the

1 sedimentological, paleoenvironmental and paleoclimatological history of Lake Ohrid, and
2 form the basis of more detailed work in the future. Furthermore, our results enable a first
3 characterization of glacial and interglacial severity since marine isotope stage (MIS) 16.

4 **2 Site Information**

5 Lake Ohrid is located at the border of the Former Yugoslav Republic of Macedonia
6 (FYROM) and Albania at an altitude of 693 m above sea level (m asl, Fig. 1A). The lake is
7 approximately 30 km long, 15 km wide and covers a surface area of 358 km². Due to its
8 location in a tectonic, N-S trending graben system, the lake basin is tub-shaped with a mean
9 water depth of 150 m and a maximum water depth of 293 m (Fig. 1B). The water volume
10 calculates to 55.4 km³. The lake is mainly fed by karstic inflow (55%, e.g. Matzinger et al.,
11 2007; Vogel et al., 2010a), and by small rivers. The karstic inflow partly originates from Lake
12 Prespa, located at an altitude of 848 m asl ca. 10 km to the east of Lake Ohrid (Fig. 1B). Both
13 lakes are connected via karstic aquifers. The lake level of Ohrid is balanced by a surface
14 outflow in the northern corner (Crim Drim River, 60%, Fig. 1B), and by evaporation (40%,
15 Matzinger et al., 2006b). The large water volume and the high proportion of karstic inflow
16 induce an oligotrophic state in Lake Ohrid. A complete overturn of the water column occurs
17 approximately every 7 years (e.g. Matzinger et al., 2007), although the upper about 200 m of
18 the water column are mixed every year.

19 The catchment of Lake Ohrid comprises 2393 km² including Lake Prespa (Fig. 1B). Both
20 lakes are separated by the up to 2300 m asl high Galicica mountain range (Fig. 1B). To the
21 west of Lake Ohrid, the Mocra mountain chain rises up to about 1500 m asl. The
22 morphostructure with high mountains to the west and east of Lake Ohrid is mainly the result
23 of a pull-apart like opening of the basin during the late phases of the Alpine orogeny (Aliaj et
24 al., 2001; Hoffmann et al., 2010; Lindhorst et al., 2015). Several earthquakes in the area
25 (NEIC database, USGS) and mass wasting deposits, which occur in the lateral parts of Lake
26 Ohrid (Reicherter et al., 2011; Lindhorst et al., 2012; Wagner et al., 2012), document the
27 tectonic activity in the area until present day.

28 The oldest bedrock in the catchment of Lake Ohrid is of Devonian age, consists of
29 metasedimentary rocks (phyllites), and occurs in the northeastern part of the basin. Triassic
30 carbonate and siliciclastic rocks occur in the southeast, east, and northwest (e.g. Wagner et al.,
31 2009; Hoffmann et al., 2010; Vogel et al., 2010b). Ultramafic metamorphic and magmatic
32 rocks including ophiolites of Jurassic and Cretaceous age crop out in the west (Hoffmann et

1 al., 2010). Quaternary lacustrine and fluvial deposits cover the plains to the north and to the
2 south of Lake Ohrid (Hoffmann et al., 2010; Vogel et al., 2010b).

3 The climate at Lake Ohrid is influenced both by continental and Mediterranean climate
4 conditions (Watzin et al., 2002). Between summer and winter, monthly average air
5 temperatures range between +26°C and -1°C, respectively. The annual precipitation averages
6 to ca. 750 mm/yr⁻¹, with drier conditions during summer, and more precipitation during
7 winter. The prevailing wind directions are north and south and primarily controlled by the
8 topography of the Lake Ohrid valley (summarized by Wagner et al., 2009).

9 **3 Material and Methods**

10 **3.1 Field work**

11 The DEEP site (5045-1) is the main drill site in the central part of the lake at a water depth of
12 243 m (Fig. 1B, 41°02'57'' N, 020°42'54'' E). The uppermost sediments at the DEEP site
13 down to 1.5 m below lake floor (blf) were recovered in 2011 using a UWITEC gravity and
14 piston corer (core Co1261), as these drilling techniques provide a good core quality for sub-
15 surface sediments. In 2013, more than 1500 m of sediments were recovered from six different
16 drill holes (5045-1A to 5045-1F) at the DEEP site. The distance between each drill hole
17 averages ca. 40 m. Holes 5045-1A and 5045-1E comprise sub-surface sediments down to ca.
18 2.4 and 5 m blf, respectively. Holes 5045-1B and 5045-1C were drilled down to a penetration
19 depth of 480 m blf. At hole 5045-1D, the maximum penetration of 569 m blf was reached.
20 Spot coring down to 550 m blf was conducted in hole 5045-1F in order to fill any gaps
21 present in the other holes (see also Wagner et al., 2014). After core recovery, the sediment
22 cores were cut in (up to) one meter long segments and stored in darkness at 4°C.

23 During the drilling campaign in 2013, onsite core processing comprised smear-slide analyses
24 of core catcher material and magnetic susceptibility measurements on the whole cores in 2 cm
25 resolution using a Multi-Sensor Core Logger (MSCL, GEOTEK Co.) equipped with a
26 Bartington MS2C loop sensor (see also Wagner et al., 2014). Following the field campaign,
27 the cores were shipped to the University of Cologne for further analyses.

28 **3.2 Laboratory work**

29 A first correlation of the individual core segments to provide a preliminary composite profile
30 for the DEEP site sequence was established based on the magnetic susceptibility data of the

1 whole cores from holes 5045-1B, 5045-1C, 5045-1D, and 5045-1F. Cores incorporated into
2 the composite profile were then split lengthwise and described for color, grain-size, structure,
3 macroscopic components, and calcite content (reaction with 10% HCl). High-resolution line
4 scan images were taken using the MSCL (GEOTEK Co.). X-ray Fluorescence (XRF)
5 scanning was carried out at 2.5 mm resolution and with an integration time of 10 s using an
6 ITRAX core scanner (Cox Analytical, Sweden). The ITRAX core scanner was equipped with
7 a chromium (Cr) X-Ray source and was run at 30 kV and 30 mA. Data processing was
8 performed with the software QSpec 6.5 (Cox Analytical, Sweden, cf. Wennrich et al., 2014).
9 In order to account for inaccuracies and to validate the quality of the XRF-scanning data,
10 conventional wavelength dispersive XRF (WDXRF, Philips PW 2400, Panalytical Cor., The
11 Netherlands) was conducted at 2.56 mm resolution. The optical and lithological information
12 (layer by layer correlation) were then combined with XRF scanning data to fine-tune the core
13 correlation by using the Corewall software package (Correlator 1.695 and Corelyzer 2.0.1).

14 If an unequivocal core correlation was not possible, additional core sections from other drill
15 holes in the respective depths were opened, likewise analyzed, and used to refine the core
16 correlation. In the composite profile, the field depth measurements based on ‘meters below
17 lake floor’ (m blf) were replaced by ‘meters composite depth’ (mcd). The DEEP site
18 composite profile down to 247.8 mcd comprises two sections of core Co1261 for the
19 uppermost 0.93 mcd and a total of 386 core sections from holes 5045-1B, 5045-1C, 5045-1D,
20 and 5045-1F (Fig. 2, Table 1). The overall recovery of the composite profile calculates to
21 99.97%, as no overlapping sequences were found between core run numbers 80 and 81 in
22 hole 5045-1C. The length of the core catcher (8.5 cm) between these two runs led to one gap
23 between 204.719 and 204.804 mcd.

24 At 16 cm resolution, 2 cm thick slices (40.7 cm^3) were removed from the core half and
25 separated into four sub-samples to establish a multiproxy data set. Intermediate intervals (8
26 cm distance to the 2 cm thick slices) were subsampled for high-resolution studies by pushing
27 two cylindrical plastic vials (diameter = 0.9 cm, height = 4 cm, volume = 2.5 cm^3) into the
28 core halves. In addition, samples for paleomagnetic analyses were taken in plastic cubes
29 (volume of 6.2 cm^3) at 50 cm resolution until 100 mcd, and at 48 cm resolution below this
30 depth (cf. Just et al., 2015).

31 All sub-samples (8 cm resolution) were freeze-dried, and the water content was calculated by
32 the difference in weight before and after drying. For every other sample, an aliquot of about
33 100 mg was homogenized and ground to $<63 \mu\text{m}$. For the measurement of total carbon (TC)

1 and total inorganic carbon (TIC) using a DIMATOC 100 carbon analyzer (Dimatec Corp.,
2 Germany), 40 mg of this aliquot was dispersed with an ultrasonic disperser in 10 ml DI water.
3 TC was measured as released CO₂ after combustion at 900°C. The TIC content was
4 determined as CO₂ after treating the dispersed material with phosphoric acid (H₃PO₄) and
5 combustion at 160°C. The total organic carbon (TOC) content was calculated from the
6 difference between TC and TIC. For the measurement of total sulfur (TS) and total nitrogen
7 (TN), 10 mg of the ground material was analyzed using an elemental analyzer (vario MICRO
8 cube, Elementar Corp.) after combustion at 1150°C.

9 Biogenic silica (bSi) concentrations were determined at 32 cm resolution by means of Fourier
10 Transform Infrared Spectroscopy (FTIRS) at the Institute of Geological Sciences, University
11 of Bern, Switzerland. For sample preparation, 11mg of each sample was mixed with 500 mg
12 of oven-dried spectroscopic grade potassium bromide (KBr, Uvasol®, Merck Corp.) and
13 subsequently homogenized using a mortar and pestle. A Bruker Vertex 70 equipped with a
14 liquid nitrogen cooled MCT (Mercury–Cadmium–Telluride) detector, a KBr beam splitter,
15 and a HTS-XT accessory unit (multi-sampler) was used for the measurement. Each sample
16 was scanned 64 times at a resolution of 4 cm⁻¹ (reciprocal centimeters) for the wavenumber
17 range from 3750 to 520 cm⁻¹ in diffuse reflectance mode. After the measurements, a linear
18 baseline correction was applied to normalize the FTIR spectra and to remove baseline shifts
19 and tilts by setting two points of the recorded spectrum to zero (3750 and 2210-2200 cm⁻¹).
20 The determination of bSi from FTIR spectral information relies on spectral variations in
21 synthetic sediment mixtures with known bSi concentrations and calibration models between
22 the FTIR spectral information and the corresponding bSi concentrations based on partial least
23 squares regression (PLSR, Wold et al., 2001 and references therein). For details and
24 information regarding ground truthing of the calibration see Meyer-Jacob et al. (2014).

25 For grain-size analyses at 64 cm resolution, 1.5 g of the sample material was treated with
26 hydrogen peroxide (H₂O₂, 30%), hydrochloric acid (HCl, 10%), sodium hydroxide (NaOH,
27 1M) in order to remove authigenic matter and Na₄P₂O₇ for sample dispersion. Prior to the
28 analyses, the sample material was dispersed on a shaker for 12h and underwent one minute of
29 ultrasonic treatment. Sample aliquots were then measured three times with a Saturn DigiSizer
30 5200 laser particle analyzer equipped with a Master Tech 52 multisampler (Micromeritics
31 Co., USA) and the individual results were averaged. Data processing was carried out by using
32 the GRADISTATv8 program (Blott and Pye, 2001).

1 **4 Results and Discussion**

2 **4.1 Lithology**

3 The sediments from the DEEP site sequence down to 247.8 mcd consist of fine-grained
4 hemipelagic sediments, which are sporadically interspersed by more coarse-grained event
5 layers. From the top to the bottom of the sequence, the water content decreases from a
6 maximum of 70% to a minimum of 32% due to compaction by overlying deposits (for
7 detailed studies on the sediment compaction at the DEEP site see Baumgarten et al., 2015).

8 **4.1.1 Hemipelagic sediments**

9 The hemipelagic deposits of the DEEP site sequence were subdivided into three lithotypes
10 (Fig. 2, 3) based on information from the visual core descriptions. This includes variations in
11 the sediment color and structure.

12 The sediments of **lithotype 1** (calcareous silty clay, Fig. 2) have very dark greenish grey to
13 greenish grey colors, and appear massive or mottled (cf. Fig. 3A to 3D). Silt to gravel-sized
14 vivianite concretions occur irregularly distributed within lithofacies 1 and can be identified by
15 a color change from grey/white to blue after core opening.

16 TIC contents between 2.0% and 9.7% imply that calcite (CaCO₃) is abundant in lithotype 1
17 sediments. Changes in color correspond to different calcite and TOC contents in the deposits
18 (Fig. 3). The TOC content can be used as an indicator of the amount of finely dispersed
19 organic matter (OM) in lacustrine deposits (e.g. Cohen, 2003; Stockhecke et al., 2014b). In
20 the sediments of lithotype 1, bright colors (greenish grey) are commonly correlated with
21 massive layers and are indicative for high calcite and low OM contents. Dark (very dark
22 greenish grey, dark greenish grey) lithotype 1 deposits appear mottled and have lower calcite
23 and higher OM concentrations. The bSi content varies between 1.9% and 42.5% in the
24 sediments of lithotype 1 and is a measure for the amount of diatom frustules in the sediments
25 of Lake Ohrid (Vogel et al., 2010a). Additional contributions to the bSi content come from
26 sponge spicules, which were observed in in smear slides. Extraordinary high bSi
27 concentrations of up to 42.5% are restricted to discrete layers. Low potassium intensities (K,
28 Fig. 2) correspond to minima in the fine fraction (<4 μm, Fig. 2) of the grain size classes and
29 imply a low abundance of siliciclastic minerals in the bulk sediment composition of lithotype
30 1 deposits (cf. Arnaud et al., 2005; Wennrich et al., 2014).

1 **Lithotype 2** (slightly calcareous silty clay) sediments are greenish black and very dark
2 greenish grey in color, and appear mottled or massive (cf. Fig. 3E to 3H). Vivianite
3 concretions occur irregularly, and yellowish brown layers exhibit high amounts of siderite
4 crystals in smear-slides (Fig. 2, 4). The occurrence of siderite in the DEEP site sediments was
5 confirmed by means of XRD, EDX, and FTIRS spectroscopy (Lacey et al., 2015b).

6 TIC contents between 0.5% and 2% indicate that calcite is less abundant in lithotype 2
7 sediments. Distinct peaks in the TIC content correspond to peaks in Fe and Mn counts and to
8 the occurrence of the yellowish brown siderite layers (Fig. 4). The greenish black sediment
9 successions of lithotype 2 sediments are mottled and have high amounts of OM, as indicated
10 by TOC contents of up to 4.5%. Brighter, very dark greenish grey sections can be massive or
11 mottled and have lower TOC contents (Fig. 3). The amount of diatom frustules is moderate to
12 high, as inferred from bSi contents between 2% and 27.9%. The bulk sediment composition is
13 balanced by moderate amounts of clastic material (Fig. 2, K-intensities).

14 The bright, greenish grey sediments of **lithotype 3** (silty clay) are mottled and intercalated
15 with massive sections of up to several decimeters thickness (Fig. 3I to 3L). Vivianite
16 concretions occur irregularly, and yellowish brown siderite layers are abundant (Fig. 2).

17 The TIC values of lithotype 3 sediments rarely exceed 0.5%, which infers negligible calcite
18 contents. Occasional peaks in TIC >0.5% can be attributed to the occurrence of siderite layers
19 (Fig. 2, 4). TOC ranges between 0.4% and 4.8% (Fig.2), with higher values >2.5% close to
20 the lower and upper boundaries of lithotype 3 sediment sections, and between 3.21 mcd and
21 2.89 mcd (Fig. 2). The amount of bSi is mostly between 1.68% and 14.5%, except for several
22 peaks of up to 41.3% just above tephra layers. High potassium intensities throughout most
23 parts of lithotype 3 sediments indicate high clastic matter contents and correspond to high
24 percentages of the fine fraction (<4 μ m, Fig. 2).

25 4.1.2 Event Layers

26 The macroscopic event layers were classified as tephra deposit if a high proportion of glass
27 shards were observed in the smear slides, and as mass movement deposit (MMD) if
28 predominantly coarse grains and detrital siliciclastic components occurred (cf. Fig 2, 3).
29 Tephra layers in the DEEP site sequence appear as up to 15 cm thick layers and as lenses (cf.
30 Leicher et al., 2015). Most of the tephra layers are between 0.5 cm and 5 cm thick (e.g. Fig.
31 3H and 3P). In addition, a distinct peak in the K intensities in the DEEP site sequence at 2.775
32 mcd was identified as the Mercato crypto tephra layer (cf. Table 2) by a correlation of the K

1 XRF curve from the DEEP site to the curve of core Co1262 (Wagner et al., 2012, for
2 locations of the cores see Fig. 1). Tephrostratigraphic investigation including geochemical
3 and morphological analyses of glass shards enabled the correlation of 13 tephra layers from
4 the DEEP site sequence to known volcanic eruptions or distal tephra from the central
5 Mediterranean region (cf. Table 2 and Leicher et al., 2015).

6 The MMDs in the DEEP site sequence are between 0.1 cm and 3 cm thick, and consist of very
7 coarse silt to fine sand-sized material (cf. also Fig. 3M, 3N, and 3O). A higher frequency of
8 MMDs occurs between 117 mcd and 107 mcd, and between 55 mcd and 50 mcd, respectively.
9 Most of the MMDs show normal gradation (Fig. 3N), or appear as lenses (Fig. 3O). In some
10 very thin MMDs, the gradation is only weakly expressed. The MMD in Fig. 3M differs from
11 all other MMDs in the DEEP site sequence as it is the only one with a clay layer at the top,
12 and a 1.5 cm thick, poorly sorted, clay to fine sand-sized section at the bottom.

13 **4.2 Sedimentary Processes**

14 **4.2.1 Hemipelagic sediments**

15 Although a detailed examination of the sediment bedding structures in the DEEP site
16 sediments was frequently difficult due to secondary oxidation structures (cf. Fig. 3), the
17 mottled and massive appearance and the lack of lamination imply that anoxic bottom water
18 conditions did not occur. As massive structures commonly correspond to high calcite
19 concentrations in the sediments (high TIC), they can be explained by a high abundance of
20 calcite crystals in the sediments.

21 Scanning Electron Microscope (SEM) and X-ray Diffraction (XRD) analyses on carbonate
22 crystals from sediment traps (Matter et al., 2010) and from sediment cores spanning the last
23 40,000 kyrs (e.g. Wagner et al., 2009; Leng et al., 2010) show that the majority of carbonates
24 in Lake Ohrid sediments are endogenic calcite. Only minor contributions to the calcite content
25 come from biogenic sources, for example from ostracod valves (Vogel et al., 2010a), detrital
26 carbonates only occur in trace amounts (Lacey et al., 2015b). Endogenic calcite deposition in
27 the sediments of Lake Ohrid is predominately triggered by photosynthesis-induced formation
28 of calcite crystals in the epilimnion (e.g. Wagner et al., 2009; Vogel et al., 2010a). The
29 precipitation occurs at warm temperatures during spring and summer, as long Ca^{2+} and HCO_3^-
30 ions are not short in supply (e.g. Matzinger et al., 2007; Wagner et al., 2009; Vogel et al.,
31 2010a). High Ca^{2+} and HCO_3^- concentrations in Lake Ohrid are triggered by the intensity of
32 chemical weathering and limestone dissolution in the catchment, the karst discharge volume,

1 and the evaporation of lake water (Vogel et al., 2010a). The calcium carbonate concentration
2 in the sediments also depends on the preservation of the endogenic calcite. Dissolution of
3 calcite in lower parts of the water column, at the sediment water interface, and in the upper
4 sediment column can be caused by oxidation of OM, which triggers H_2CO_3 release from the
5 surface sediments and a lowering of the lake-water pH (Müller et al., 2006; Vogel et al.,
6 2010a). SEM analyses on endogenic calcite in the DEEP site sediments, in addition, indicates
7 the presence of microbial dissolution (Lacey et al., 2015b).

8 The high TIC contents in lithotype 1 imply high photosynthesis-induced precipitation of
9 endogenic calcite, high temperatures during spring and summer, good calcite preservation in
10 the sediments, and a somewhat higher lake water pH. Lower primary productivity, lower
11 temperatures, and a somewhat stronger dissolution of calcite can be inferred from the TIC
12 content in lithotype 2 and 3 sediments. In lithotype 2 and 3, siderite layers also contribute to
13 the TIC content (cf. Fig. 2, 4). In neighboring Lake Prespa, siderite formation has been
14 reported to occur in the surface sediments close to the redox boundary under rather acidic and
15 reducing conditions (Leng et al., 2013). In Lake Ohrid, DEEP site lithotype 2 and 3 sediments
16 contain discrete horizons of authigenic siderite crystals and crystal clusters nucleating within
17 an unconsolidated clay matrix (Lacey et al., 2015b). The open-packed nature of the matrix
18 and growth relationships between crystals suggest that, as also observed in Lake Prespa,
19 siderite formed in the pore spaces of the surface sediments close to the sediment-water
20 interface, similar to other ancient lakes such as Lake Baikal (Berner, 1981; Lacey et al.,
21 2015b).

22 The OM in lithotype 2 and 3 sediments is predominately of aquatic origin, as indicated by
23 TOC/TN ratios below 10 (cf. Meyers and Ishiwatari, 1995; Wagner et al., 2009). In lithotype
24 1 sediments, TOC/TN occasionally exceeds 10, which could imply some contributions of
25 terrestrial OM. However, due to the coring location in the central part of Lake Ohrid and the
26 relatively small inlet streams, a substantial supply of allochthonous OM to the DEEP site is
27 rather unlikely. The high TOC/TN ratios are therefore most likely a result of early diagenetic
28 selective loss of N (cf. e.g. Cohen, 2003). The aquatic origin of the OM in the sediments of
29 the deep basin of Lake Ohrid is also in agreement with the results of Rock Eval pyrolysis
30 from the nearby LINI drill site (Lacey et al., 2015a, see Fig. 1 for coring location). This
31 implies that phases characterized by higher TOC contents are representative for a somewhat
32 elevated primary productivity. This finding is confirmed by high amounts of diatom frustules
33 in the sediments (Wagner et al., 2009) and, accordingly, in high biogenic silica contents.
34 Enhanced productivity in the lake requires high temperatures and sufficient nutrient supply to

1 the epilimnion. The nutrient supply to Lake Ohrid is mainly triggered by river inflow (e.g.
2 Matzinger et al., 2006a; Matzinger et al., 2006b; Matzinger et al., 2007; Wagner et al., 2009;
3 Vogel et al., 2010a), karstic inflow from Lake Prespa (Matzinger et al., 2006a; Wagner et al.,
4 2009), and by nutrient recycling from the surface sediments (Wagner et al., 2009).
5 Phosphorous recycling from the surface sediments is promoted by anoxic bottom water
6 conditions and mixing can transport phosphorous from the bottom water to the epilimnion
7 (e.g. Wagner et al., 2009). Mixing also leads to oxidation of OM at the sediment surface and,
8 thus, to lower TOC contents. TOC preservation in the sediments can also be modified by lake
9 level fluctuations, as oxidation of OM starts in the water column during settling (Cohen,
10 2003; Stockhecke et al., 2014b). However, distinct climate induced lake level fluctuations
11 such as described for the Younger Dryas at Lake Van in Turkey (Wick et al., 2003;
12 Stockhecke et al., 2014b and references therein) have not been observed at Lake Ohrid in
13 previous studies covering the last glacial-interglacial cycle (cf. e.g. Vogel et al., 2010a;
14 Wagner et al., 2010). This can potentially be explained by the hydrological conditions at the
15 lake, the large water volume, and the relative high contribution of karstic groundwater inflow
16 to the hydrological budget of Lake Ohrid. Hence, lower (higher) TOC content are be related
17 to an overall lower (higher) productivity and/or to more (less) oxidation of OM and improved
18 (restricted) mixing conditions.

19 Overall high TOC and bSi contents in lithotype 1 sediments imply high productivity as a
20 result of high temperatures in Lake Ohrid. Less productivity and/or oxidation of OM can be
21 inferred for sediments of lithotype 2 and 3 from low TOC and bSi contents, and from
22 TOC/TN ratios <4 (cf. Leng et al., 1999). When TOC is low such as in lithotype 3 sediments,
23 TOC/TN <4 can be a result of OM degradation (decreasing organic C concentration) and
24 clay-bound ammonium supply from the catchment (increasing N concentration), such as also
25 observed in core Lz1120 from the southeastern part of the lake (Holtvoeth et al., 2015).

26 Good OM preservation, low oxygen availability, and overall poor mixing conditions could
27 have favored sulfide formation, in lithotype 1 sediments. Sulfide formation can be indicated
28 by a low TOC/TS ratio (cf. Müller, 2001; Wagner et al., 2009). In lithotype 1 sediments, the
29 high TOC/TS ratios correspond to minima in the Fe intensities (cf. Fig. 2), which suggests
30 that Fe-sulfide formation was limited by iron and/or sulfate availability (cf. also Holmer and
31 Storkholm, 2001). Sulfide formation in the sediments of Lake Ohrid is restricted to deposits
32 where Fe availability (i.e. lithotype 2 and 3 sediments) is high (cf. Just et al., 2015).
33 Furthermore, Urban et al. (1999) have shown that early diagenetic sulfur enrichment in OM is
34 low in oligotrophic lakes, as up to 90% of the produced sulfides can be re-oxidized seasonally

1 or episodically, which affects the sulfur storage over several years. At Lake Ohrid, re-
2 oxidation of sulfides may occur during the mixing season, or under present climate conditions
3 during the irregular complete overturn of the entire water column every few years. If re-
4 oxidation has biased the TOC/TS ratio, the lower ratios in lithotype 2 and 3 sediments are
5 rather a result of the overall low TOC concentrations, which is confirmed by the good
6 correspondence between the TOC/TS ratio and the TOC content.

7 Elemental intensities of the clastic matter, obtained from high resolution XRF scanning, can
8 provide information about the sedimentological composition of the deposits, and about
9 erosional processes in the catchment. Variations in the potassium intensities (K, Fig. 2) and in
10 the clastic matter content of DEEP site sequence sediments could primarily be a result of
11 changing erosion in the catchment, such as it has also been reported from other lakes on the
12 Balkan Peninsula (e.g. Francke et al., 2013). This implies that increased denudation and soil
13 erosion could be inferred for lithotype 2 and 3 sediments, while less clastic matter supply
14 occurs during deposition of lithotype 1 sediments. However, mutual dilution with authigenic
15 components such as calcite, OM and diatom frustules can bias the potassium record as
16 indicator for denudation and clastic matter supply.

17 Potassium can occur in K-feldspars, micas, and clay minerals and is mobilized particularly
18 during chemical weathering and pedogenesis, and the residual soils in the catchment become
19 depleted in potassium (Chen et al., 1999). In contrast to K, Zirconium (Zr) mostly occurs in
20 the mineral zircon, which has a high density and resistance against physical and chemical
21 weathering and is therefore commonly enriched in coarse-grained (aeolian) sediments.
22 However, in lithotype 1 and 2 sediments, low Zr/K ratios match low percentages of the <4 μ m
23 fraction (cf. Fig. 2). This implies that coarse-grained sediments (low <4 μ m percentages) are
24 depleted in Zr and enriched in K at the DEEP site. Thus, the Zr/K ratio rather provides
25 insights into the intensity of the chemical weathering in the catchment than being dependent
26 on the physical grain size. As chemically altered minerals may be stored for hundreds and
27 thousands of years in the catchment before they are eroded, transported, and finally deposited
28 (cf. Dosseto et al., 2010), the Zr/K ratio does not provide information about weathering
29 processes during the time of deposition at the DEEP site. The match of low Zr/K ratios and
30 low percentages of the <4 μ m fraction in lithotype 1 and 2 sediments imply that more coarse
31 detrital matter, predominately consisting of K-rich clastic material from young and less
32 chemically weathered soils, were deposited at the DEEP site. In contrast, high Zr/K ratios in
33 lithotype 3 sediments match with high percentages of the <4 μ m fraction and suggest that
34 more fine grained, chemically weathered, and K-depleted siliciclastics from mature soils were

1 supplied to the lake. Deviations from the correspondence between Zr/K and the <4 μ m grain
2 size fraction can be explained by additional processes that affect the deposition of clastic
3 matter at the DEEP site, such as the transportation energy in the inlet streams, lake level
4 fluctuations and the shoreline distance, and the strength of lake-internal current systems,
5 respectively (cf. also Vogel et al., 2010b).

6 4.2.2 Event Layers

7 Probable trigger mechanisms for MMDs have widely been discussed and encompass
8 earthquakes, delta collapses, flooding events, over steepening of slopes, rock falls, and lake-
9 level fluctuations (e.g. Cohen, 2003; Schnellmann et al., 2006; Girardclos et al., 2007;
10 Sauerbrey et al., 2013). At Lake Ohrid, MMDs in front of the Lini Peninsula (Fig. 1) and in
11 the southwestern part of the lake were likely triggered by earthquakes (Lindhorst et al., 2012;
12 Wagner et al., 2012; Lindhorst et al., 2015). An earthquake might have also triggered the
13 deposition of the MMD in Fig. 3M, which is composed of a turbidite succession and an
14 underlying, poorly sorted debrite (after the classification of Mulder and Alexander, 2001).
15 The disturbance generated by a debris flow can cause co-genetic turbidity currents of fine-
16 grained material in front and above the mass movement (Schnellmann et al., 2005; Sauerbrey
17 et al., 2013). As the debrite-turbidite succession occurs at 7.87 mcd in hole 5045-1F, it likely
18 corresponds to a massive slide complex north of the DEEP site (cf. hydro acoustic profile of
19 Fig. 2 in Wagner et al., 2014). Density flows that enter the central part of the Lake Ohrid
20 basin close to the DEEP site from eastern or southern directions have not been observed in
21 upper parts of hydro-acoustic profiles (cf. Fig. 2 and 3 in Wagner et al., 2014). The three
22 massive MMDs that occur in front of the Lini Peninsula to the west of the DEEP site (cf.
23 Wagner et al., 2012) are likely not related to the debrite-turbidite succession in Fig. 3M. The
24 underlying debrite does not occur in overlapping segments of holes 5045-1B, 5045-1C, and
25 5045-1D. Holes 5045-1B, 5045-1C, and 5045-1D form a N-S transect, whereas hole 5045-1F
26 is located to the east. Due to the absence of the debrite deposits in most drill holes, the
27 relatively low thickness in hole 5045-1F, and hydroplaning that generates a basal water layer
28 below a debris flow (Mohrig et al., 1998; Mulder and Alexander, 2001), erosional processes
29 at the DEEP site are likely low.

30 The sand lenses and normal graded MMDs (cf. Fig. 2, 3) can be classified as grain-flow
31 deposits (after the classification of Mulder and Alexander, 2001; Sauerbrey et al., 2013) and
32 are composed of reworked lacustrine sediments from shallow water depths or subaquatic
33 slopes close to riverine inflows. Grain flows that enter the deep parts of the Lake Ohrid basin

1 via the steep slopes might transform into a mesopycnal flow at the boundary of the
2 hypolimnion (cf. also Mulder and Alexander, 2001; Juschus et al., 2009), which would have
3 prevented erosion of the underlying sediments.

4 **5 Core chronology**

5 The chronology for the sediments of the DEEP site sequence down to 247.8 mcd was
6 established by using tephrochronological information from 11 out of 13 tephra layers (cf.
7 Table 2 and Leicher et al., 2015) and by cross-correlation to orbital parameters. The tephra
8 layers were correlated to well-known eruptions from Italian volcanoes or central
9 Mediterranean marker tephra by geochemical and morphological analyses of glass shards.
10 Radiocarbon and $^{40}\text{Ar}/^{39}\text{Ar}$ ages were transferred from the reference records to the DEEP site
11 sequence (tephrostratigraphic results of the DEEP site are provided by Leicher et al. (2015)).
12 $^{40}\text{Ar}/^{39}\text{Ar}$ ages from the literature were re-calculated by using the same flux standard in order
13 to obtain a homogenous set of ages (see Leicher et al., 2015). The stratigraphic position and
14 chronology of the OH-DP-0499/P-11 tephra layer at 49.947 mcd in different records from the
15 vicinity of Lake Ohrid is discussed in detail by Zanchetta et al. (2015). Their results imply
16 that the P-11 tephra layer has an age of 133 ± 2 ka, which was incorporated into the age-depth
17 modeling. As the 11 tephra layers provide a robust basis for the age depth model of the DEEP
18 site sequence, the tephrostratigraphic information was used as 1st order tie points.

19 The chronological information from the 11 tephra layers was also used to define cross
20 correlation points to orbital parameters, which were included into the age depth model as 2nd
21 order tie points. The tephra layers Y-5, X-6, P-11, and A11/12 were deposited when there are
22 minima in the TOC content and in the TOC/TN ratio and when there is an inflection point
23 (blue dots, Fig. 5) of increasing local summer insolation (21st June, 41°N). Thereby, the
24 inflection points coincide with an increasing winter season length (21st September to 21st
25 March, Fig. 5). Summer insolation and winter season length have a direct impact on the OM
26 content and the TOC/TN ratio, as they may trigger primary productivity and decomposition.
27 Low insolation and colder temperatures during summer reduce the primary productivity in the
28 lake, but simultaneously, a shorter winter season would have reduced mixing in the lake,
29 which reduces the decomposition of OM and increases TOC and TOC/TN (cf. Fig 5,
30 insolation and winter season length minima). A longer winter season improves the mixing,
31 but a strong insolation during summer promotes the primary productivity in the lake, which
32 also results in higher TOC and TOC/TN (cf. Fig. 5, insolation and winter season length
33 maxima). Thus, low OM preservation and low TOC and TOC/TN in the sediments may occur

1 when summer insolation strength and winter season length are balanced. In addition, the
2 inflection points coincide with the perihelion passage in March. As the highest proportion of
3 the annual radiation gets lost through surface albedo during spring (Berger et al., 1981), the
4 time period of the perihelion passage in March is characterized by cold and dry conditions in
5 the central Mediterranean region (Magri and Tzedakis, 2000; Tzedakis et al., 2003; Tzedakis
6 et al., 2006). Cold conditions at Lake Ohrid promote mixing during winter and restrict the
7 primary productivity during summer. Thus, minima in TOC and TOC/TN are tuned to
8 increasing insolation and winter season length.

9 The 11 tephra layers and 31 cross correlation points of 2nd order (supplement 1) were used
10 for the establishment of an age-depth model. An uncertainty of ± 2000 years was applied for
11 each 2nd order tie point in order to account for inaccuracies in the tuning process. For the age
12 depth modeling using the Bacon 2.2 software package (Blaauw and Christen, 2011), overall
13 stable sedimentation rates at the DEEP site (mem.strength = 60, mem.mean = 0.9, thick = 80
14 cm) and expected sedimentation rates (acc.shape = 1.5, acc.mean = 20) from first age
15 estimations for the DEEP site sequence by Wagner et al. (2014) were considered (cf. Fig. 6).
16 Finally, the age model was evaluated and refined by a detailed comparison with the age depth
17 model for the downhole logging depth scale by tuning Potassium counts (K) obtained from
18 high-resolution XRF scanning to the spectral gamma radiation (SGR) of potassium (cf.
19 supplement 2) in hole 5045-1D (Baumgarten et al., 2015, cf. Fig. 6). The obtained age model
20 reveals that the upper 247.8 mcd of the DEEP site sequence comprises the last ca. 637 kyrs.

21 On the basis of the established core chronology for the DEEP site sequence, glacial-
22 interglacial environmental variability at Lake Ohrid inferred from the TOC record is in
23 agreement with other paleoclimate records from the Mediterranean region such as the Tenaghi
24 Philippon (Tzedakis et al., 2006) and the Soreq cave records (Grant et al., 2012 cf. Fig. 6).
25 This supports the quality of the chronology for the DEEP site sediments. Whereas the
26 chronology of the Tenaghi Philippon record is, similar to the DEEP site sequence, based on
27 tuning against orbital parameters, the Soreq Cave speleothem record is the longest absolute
28 dated (U-Th) paleoclimate record currently available for the Mediterranean region.

29 **6 Overview about the Paleoenvironmental History of Lake Ohrid**

30 Variations in the TIC, bSi, TOC, K, and Zr/K records of the DEEP site sequence correspond
31 to global and regional climatic variability on glacial-interglacial time scales, such as indicated
32 by a comparison to the global benthic isotope stack LR04 (Lisiecki and Raymo, 2005), to
33 North Greenland isotope record (NGRIP-members, 2004; Barker et al., 2011), and to

1 variations in the arboreal pollen percentage of the Tenaghi Philippon record from northern
2 Greece (Tzedakis et al., 2006, cf. Fig. 7). In addition to the close match of climatic variability
3 on orbital time scales between those records and the Lake Ohrid record, a comparison of the
4 individual interglacial and glacial stages allows a first discrimination of the intensity of these
5 stages at Lake Ohrid.

6 **6.1 Interglacials**

7 Between 637 ka and present day, the DEEP site sediments deposited during the interglacial
8 periods (MIS boundaries after Lisiecki and Raymo, 2005, see Fig. 7) mainly consist of
9 lithotype 1 and 2 sediments. Moderate to high TIC, TOC, and bSi contents in lithotype 1 and
10 2 sediments imply a moderate to strong primary productivity, and thus, moderate to high
11 temperatures during spring and summer. The overall high temperatures during spring and
12 summer and a longer summer season likely resulted in an incomplete and restricted mixing of
13 the water column during winter, such as it also persists today. Poor mixing hampers the
14 oxidative mineralization of OM and thus, promotes the preservation of TOC and restricts the
15 bacterial CO₂ release at the sediment surface. A reduction in the H₂CO₃ formation and a
16 higher pH hampers the acidification in the hypolimnion and consequently improve the calcite
17 preservation. In addition, interglacial climate conditions likely promoted high Ca²⁺ and HCO₃⁻
18 concentrations and a high pH also in the epilimnion, as warm temperatures increased
19 chemical weathering of the limestones in the catchment. In addition high δ¹⁸O-calcite values
20 indicating a low Precipitation/Evaporation (P/E) ratio (Lacey et al., 2015b), suggest that
21 evaporation may also contribute to high Ca²⁺ and HCO₃⁻ concentrations in the lake water (cf.
22 Fig. 7). Supply from the catchment and increased evaporation could have also increased the
23 concentration of Si ions in the epilimnion, which could have promoted diatom productivity.
24 The sensitivity of diatom productivity to increasing Si concentrations in the water column has
25 been clearly shown in leaching experiments of tephras (D'Addabbo et al., 2015), and in high
26 resolution studies of diatom changes after the deposition of the Y-5 tephra (Jovanovska et al.,
27 2015).

28 Low supply of clastic matter from the catchment can be inferred for lithotype 1 and 2
29 sediments, where K intensities and the sedimentation rates are low (~ <0.04 cm/yr). Low
30 supply of clastic matter is likely a result of low denudation rates despite intensive chemical
31 weathering and pedogenesis during interglacial periods in the catchment. Relatively high
32 pollen concentrations in the interglacial sediments of the DEEP site core imply a dense
33 vegetation cover (cf. Fig. 7 and Sadori et al., 2015), which likely reduces erosion. In addition,

1 the low Zr/K ratios and the low proportion of the <4 μ m grain size fraction in lithotype 1 and 2
2 sediments imply that in particular K-rich minerals and the products of young soils were
3 transported to the lake.

4 Lithotype 3 deposits with negligible TIC contents only occur at the onsets and terminations of
5 interglacial periods, and during MIS7 and 3 (cf. Fig. 7). The low TIC, TOC, and bSi contents
6 of these sediments correspond to colder periods with a restricted primary productivity. In
7 addition, low temperatures during winter would have improved the mixing, which could
8 promote decomposition of OM in the surface sediments and led to lower TOC, and to lower
9 TIC by dissolution of calcite.

10 Variations of interglacial conditions since 637 ka

11 Differing OM, bSi, and TIC contents in the sediments of the DEEP site sequence
12 corresponding to interglacial time periods imply different intensities of interglacials at Lake
13 Ohrid. Comparable conditions can be inferred for MIS15 and 13 with high TIC and bSi, and
14 low OM, which indicate strong primary productivity and high temperatures during spring and
15 summer, and decomposition of OM during the mixing season. However, MIS15 and 13 are
16 regarded as relatively weak interglacials based on the synthetic Greenland isotope record
17 (Barker et al., 2011) and the global benthic isotope stack LR04 (Lisiecki and Raymo, 2005 cf.
18 also Fig. 7). Possible explanations could be that the inferred high intensity of these
19 interglacials at Lake Ohrid is due to a strong seasonality or a lower water volume, which
20 promotes a high ion concentration (high TIC, bSi) and high $\delta^{18}\text{O}_{\text{calcite}}$ values in the lake water
21 (Lacey et al., 2015b), and decomposition of OM (low TOC) during the mixing season. A
22 lower water volume can be explained by the ongoing subsidence in the lake basin, which
23 persist until today (Lindhorst et al., 2015).

24 During the first part of MIS11, between 420 ka and 400 ka, highest TIC concentrations along
25 with moderate and high bSi and TOC imply highest productivity (high TOC) and highest
26 temperatures (highest TIC), whereby the moderate bSi concentrations can be explained by
27 mutual dilution with calcite. This is consistent with other records, where strongest interglacial
28 conditions and highest temperatures since 637 ka are reported for the onset of MIS11 (Lang
29 and Wolff, 2011).

30 TIC concentration during the second phase of MIS11, between 400 ka and 374 ka, and during
31 MIS9 and 7 are generally lower and mostly restricted to confined peaks, which implies
32 overall less calcite precipitation, less primary productivity, and lower temperatures at Lake
33 Ohrid. This is consistent with the low bSi concentrations, but not with the high TOC contents,

1 and with relatively stronger interglacials subsequent to the MBE (Mid-Brunhes Event)
2 inferred from the synthetic North Greenland isotope record (Barker et al., 2011) and the LR04
3 stack (Lisiecki and Raymo, 2005). The temperatures during the second phase of MIS11 and
4 during MIS9 and 7 were likely lower compared to the first phase of MIS11 as indicated by the
5 TIC record (cf. Fig 7). In addition, the somewhat lower TIC and bSi contents also correspond
6 to lower $\delta^{18}\text{O}_{\text{calcite}}$ in the Lake Ohrid sediments, which implies that this interglacial periods
7 were isotopically fresher and less evaporated than the previous interglacial periods (Lacey et
8 al., 2015b). Between, 235 ka and 220 ka (MIS7), restricted primary productivity and
9 improved mixing are indicated by negligible TIC and low TOC and bSi concentrations.

10 Overall high TIC, TOC, and bSi concentrations during MIS5 imply a strong primary
11 productivity in the epilimnion and high temperatures during spring and summer. $\delta^{18}\text{O}_{\text{calcite}}$
12 during MIS5 is notably higher compared to the two previous interglacial periods (Lacey et al.,
13 2015b) and indicates a low P/E ratio. In particular the onset of MIS5 is reported to be one of
14 the warmest interglacial period in marine records (Lang and Wolff, 2011), which is also
15 indicated in the North Greenland temperature variations (NGRIP-members, 2004; Barker et
16 al., 2011), and in the global benthic isotope stack LR04 (Lisiecki and Raymo, 2005).

17 **6.2 Glacials**

18 Glacial periods between 637 ka and today are characterized by predominant deposition of
19 lithotype 3 sediments, with rare occurrence of lithotype 1 and 2 sediments in MIS14 and 8,
20 when TIC contents are higher. Low TOC and bSi and negligible TIC contents in lithotype 3
21 sediments imply low primary productivity and overall low temperatures during glacial
22 periods. Some minor fluctuations in productivity and temperature are indicated by TOC and
23 bSi. They are not documented in TIC, because restricted ion supply from the catchment and
24 oxidation of OM at the sediment surface due to intensified and prolonged mixing may have
25 led to a slight decrease of the bottom water pH and dissolution of calcite precipitated from the
26 epilimnion. Dissolution of calcite and the existence of a threshold can also explain the
27 delayed increase of TIC compared to TOC and bSi at the transitions of MIS16, MIS12,
28 MIS10, MIS8, MIS6, and MIS2 into the following interglacials.

29 High K, a high proportion of the fine fraction $<4\ \mu\text{m}$, and high sedimentation rates ($\sim >0.04$
30 cm/yr) despite low calcite, OM and bSi content in the glacial sediments indicate high input of
31 clastic terrigenous matter and increased erosion in the catchment. Furthermore, the high Zr/K
32 ratios suggest the supply of K-depleted, intensively weathered siliciclastics from the

1 catchment, which is also supported by a higher hematite + goethite to magnetite ratio (low S-
2 ratio) in glacial deposits of the DEEP site sequence (cf. Fig. 7 and Just et al., 2015). The
3 enhanced erosion of intensively weathered siliciclastic material can be explained by less
4 dense vegetation cover in the catchment, such as implied by low pollen concentrations in the
5 DEEP site sequence in most of the glacial periods (cf. Fig. 7 and Sadori et al., 2015), and by
6 the existence of local ice caps in the surrounding mountains of Lake Ohrid. The existence of
7 ice caps is indicated by moraines in the catchment, which are thought to have formed during
8 the last glacial cycle (Ribolini et al., 2011).

9 Variations of glacial conditions since 637 ka

10 As TIC is affected by dissolution, information about the severity of the individual glacials at
11 Lake Ohrid can only be inferred from TOC and bSi. Minima in the TOC and bSi imply that
12 most severe glacial conditions at Lake Ohrid occurred at the end of MIS16, and during
13 MIS12, 10, and 6. Somewhat higher bSi and TOC in parts of MIS14 and 8, and in MIS6, 4,
14 and 2 imply less severe glacial conditions. This suggests that the finding of glacial moraines
15 from MIS2 (Ribolini et al., 2011) is probably only due to better preservation of these glacial
16 features compared to the older glacials. Interglacial-like conditions with higher primary
17 productivity and reduced oxidation of OM in the surface sediments prevailed at the
18 occurrence of lithotype 1 and 2 sedimentation, i.e. between 563 ka and 540 ka during MIS14,
19 and between 292 ka and 282 ka during MIS8. Interglacial-like conditions along with a forest
20 expansion and more warm conditions between 292 ka and 282 ka can also be seen in the
21 pollen records of Lake Ohrid (cf. Fig 7 and Sadori et al., 2015) and Tenaghi Philippon
22 (Fletcher et al., 2013).

23 The general observation that MIS16 and 12 were more severe glacials is in broad agreement
24 with other records, such as the North Greenland isotope record (Barker et al., 2011), the
25 global benthic stack LR04 (Lisiecki and Raymo, 2005), and the Tenaghi Philippon pollen
26 record (Tzedakis et al., 2006). Thereby, the comparable low sedimentation rates in
27 combination with the negligible TIC, bSi and TOC concentrations between 460 ka and 430 ka
28 (MIS12, cf. Fig. 7) imply low supply of clastic matter and thus, low erosion in the catchment
29 despite an open vegetation cover in the catchment (cf. Fig. 7 and Sadori et al., 2015). One
30 potential explanation for the low clastic matter supply could be dry conditions and associated
31 reductions in terrestrial runoff compared to other glacials.

32 The frequent occurrence of MMDs between 280 ka and 241 ka and between 160 ka and 130
33 ka could reflect significant lake level fluctuations during MIS8 and 6. During the first period

1 (MIS8), distinct fluctuations in the pollen concentrations of the DEEP site sediments (cf. Fig.
2 7 and Sadori et al., 2015) correspond to similar fluctuations in the AP pollen percentages of
3 the Tenaghi Philippon pollen record (Tzedakis et al., 2006) and probably indicate a shift from
4 cold and dry to more warm and humid conditions in northern Greece and at Lake Ohrid.
5 During MIS6, a 60 m lower lake-level compared to present conditions and a subsequent lake
6 level rise during late MIS6 or during the transition from MIS6 to MIS5 is reported from
7 hydro-acoustic and sediment core data from the northeastern corner of Lake Ohrid (Lindhorst
8 et al., 2010). In the sediments of the DEEP site, the MIS6 to MIS5 transition occurs at ca. 50
9 mcd, which could indicate that the water depth might have not changed significantly
10 compared to the present conditions. This can be explained by the ongoing subsidence
11 (Lindhorst et al., 2015). However, the pollen record from the DEEP site also implies a phase
12 of strong aridity during MIS6 (Sadori et al., 2015), which might imply that climate-induced
13 lake level fluctuations at Lake Ohrid were probably less severe compared for example to Lake
14 Van in Turkey, where a 260 m lower lake level has been reported for the Younger Dryas (e.g.
15 Wick et al., 2003 and references therein; Stockhecke et al., 2014b). Thereby, the
16 extraordinary high sedimentation rates in particular during the first part of MIS6 in
17 combination with high K intensities and low bSi, TIC, and TOC imply intensive erosion.

18 **7 Summary and Conclusion**

19 The investigated sediment succession between 247.8 mcd and the sediment surface from the
20 DEEP site in the central part of Lake Ohrid provides a valuable archive of environmental and
21 climatological change for the last 637 kyrs. An age model was established by using
22 chronological tie points from 11 tephra layers, and by tuning biogeochemical proxy data to
23 orbital parameters. The imprint of environmental change on the lithological,
24 sedimentological, and (bio-)geochemistry data can be used to unravel the lake's history
25 including the development of the Lake Ohrid basin, and the climatological variability on the
26 Balkan Peninsula.

27 The lithological, sedimentological, and geochemical data from the DEEP site sequence imply
28 that Lake Ohrid did not experience major catastrophic events such as extreme lake-level low
29 stands or desiccation events during the last 637 kyrs. Hiatuses are absent and the DEEP site
30 sequence provides an undisturbed and continuous archive of environmental and
31 climatological change.

32 Based on the initial core description and the calcite content, the hemipelagic sediments from
33 the DEEP site sequence can be classified into three different lithotypes. This classification is

1 supported by variations in the (bio-)geochemistry proxies and matches climate variations on
2 glacial/interglacial time scales. Overall, interglacial periods are characterized by high primary
3 productivity during summer, restricted mixing during winter, and low erosion in the
4 catchment. During glacial periods, the primary productivity is low, intense mixing of the
5 water column promotes the decomposition of OM, which may have lowered the water pH and
6 led to dissolution of calcite. Enhanced erosion of chemically altered siliciclastics and a overall
7 higher clastic matter input into the lake during glacial periods can be a result of a less dense
8 vegetation cover in the catchment and melt water run-off from local glacier on the
9 surrounding mountains.

10 Following a strong primary productivity during spring and summer, the highest interglacial
11 temperatures can be inferred for the first part of MIS11 and for MIS5. In contrast, somewhat
12 lower spring and summer temperatures are observed for MIS15, 13, 9, and 7. The data also
13 suggest that high ion and nutrient concentrations in the lake water promote calcite
14 precipitation and diatom growth in the epilimnion during MIS15, 13, and 5, whereas less
15 evaporated interglacial periods exhibit lower TIC and bSi contents (MIS9 and 7).

16 Most severe glacial conditions at Lake Ohrid occurred during MIS16, 12, 10, and 6, whereas
17 somewhat warmer temperatures can be inferred for MIS14, 8, 4, and 2. Interglacial-like
18 conditions occurred during parts of MIS14 and 8, respectively.

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33 sequence, the reader is referred to Sadori et al. (2015) for a detailed list of all involved

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6

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1 Table 1. The contributions of the different core sections to the composite DEEP site profile.

Hole	Number of core runs	Number of Sections
Co1261	2 (1.1%)	2 (0.5%)
5045-1B	26 (14.2 %)	50 (12.9%)
5045-1C	72 (39.3%)	137 (35.3%)
5045-1D	75 (41.0%)	184 (47.4%)
5045-1F	8 (4.4%)	15 (3.9%)
Σ	183	388

2

3 Table 2: Correlated tephra layer in the DEEP site sequence according to Leicher et al. (2015).
 4 $^{40}\text{Ar}/^{39}\text{Ar}$ ages from the literature were recalculated at 2σ confidence levels (Leicher et al.,
 5 2015). * Calibrated ^{14}C age, + recalculated $^{40}\text{Ar}/^{39}\text{Ar}$ ages, ° age for P-11 from Zanchetta et al.
 6 (2015)

Ohrid Tephra (mcd)	Correlated eruption/tephra	Age (ka)
2.775	Mercato	$8.540 \pm 0.05^*$
11.507	Y-3	$29.05 \pm 0.37^*$
16.933	Y-5	$39.6 \pm 0.1^+$
40.486	POP2	$102 \pm 2.4^+$
43.513	X-6	$109 \pm 2^+$
49.947	P-11	$133.5 \pm 2^\circ$
61.726	Vico B	$162 \pm 6^+$
181.769	Pozzolane Rosse	$457 \pm 2^+$
195.566	SC5	$493.1 \pm 10.9^+$
201.782	A11/A12	$511 \pm 6^+$

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2 Figure 1. **A:** Location of lakes Ohrid and Prespa on the Balkan Peninsula at the border of the
3 Former Yugoslav Republic of Macedonia (FYROM) and Albania. TP: Coring location of the
4 Tenaghi Pilippon record. **B:** Map of the area of lakes Ohrid and Prespa and bathymetric map
5 of Lake Ohrid (from Lindhorst et al., 2015). Marked in white are the DEEP site and the short
6 cores Lz1120 (Wagner et al., 2009), Co1202 (Vogel et al., 2010a), and LO2004-1
7 (Belmecheri et al., 2009).

8 Figure 2. Variations of the lithological and (bio-)geochemical proxy data of the DEEP site
9 sequence plotted against meter composite depth (mcd). The core composite profile of the
10 DEEP site sediment sequence consists of core sections from core Co1261 (upper 0.93 mcd),
11 and of core sections from holes 5045-1B, 5045-1C, 5045-1D, and 5045-1F (cf. legend
12 “Composite”). Marked is also the gap in the composite profile between 204.719 and 204.804
13 mcd, where no overlapping core segments are available. The lithological information includes
14 the classification of the sediments into the three lithotypes (for color code see legend
15 “Lithology”), and information about the water content, TIC, TOC, bSi, TOC/TS, TOC/TN, K,
16 Fe, Zr/K, and grain size variability (<4 μ m grain size fraction). High-resolution XRF data was
17 filtered by using a lowpass filter (5th order, cut off frequency: 0.064 Hz) in order to remove
18 white noise from the data. Red dots mark the results of the conventional XRF analyses. The
19 occurrence of siderite layers, tephra layers, and mass movement deposits (MMD) are
20 indicated on the right column (cf. legend “Lithology”). Tephra layer and MMDs marked with
21 an asterisk are shown in Figure 3: *1: M, *2: O, *3: N, *4: P, *5: H.

22 Figure 3. High-resolution line-scan images showing characteristic core segments from
23 deposits of lithotype 1 to 3, and of Mass Movement Deposits (MMD) and tephra layers. The
24 vertical scale is in centimeter section depth. For composite depths of the line scan image see
25 supplement.

26 Figure 4. Siderite layer in core 1F-11H-3 (ca. 60 cm section depth) at 22.56 to 23.57 mcd. The
27 yellowish brown siderite layer correlates to enhanced TIC, iron (Fe), and manganese (Mn)
28 intensities in the sediments. For SEM images of the siderite, please see Lacey et al. (2015b).

29 Figure 5. **Left:** Comparison between TOC (DEEP site) and arboreal pollen percentages (AP,
30 Tenaghi Philippon, Tzedakis et al. (2006)) from 636.69 ka to the present. **Right:** Comparison
31 between TOC (DEEP site) and $\delta^{18}\text{O}$ (Soreq Cave, Grant et al. (2012)) from 160 ka to the

1 present. Red dots mark the tephrochronological age control points (1st order tie points), blue
2 dots mark the TOC versus orbital parameter tuning points (2nd order tie points). The good
3 correlation of the DEEP site TOC record with both Tenaghi Philippon and the Soreq Cave
4 temporal series supports the age model of the DEEP site succession.

5 Figure 6. Age model of the DEEP site sequence down to 247.8 mcd (637 ka). Ages were
6 calculated using the software package Bacon 2.2 (Blaauw and Christen, 2011). Overall stable
7 sedimentation rates at the DEEP site (mem.strength = 60, mem.mean = 0.9, thick = 80 cm)
8 and expected sedimentation rates (acc.shape = 1.5, acc.mean = 20) from first age estimations
9 for the DEEP site sequence by Wagner et al. (2014, cf. Fig. 6) were considered. For the ages
10 and errors of the tephra layers (red) see table 2. The cross correlation points (green) include
11 an error of ± 2000 years.

12 Figure 7. Proxy data from the DEEP site sediments plotted versus age and compared to the
13 global benthic isotope stack LR04 (Lisiecki and Raymo, 2005), the local (41°N) insolation at
14 June the 21th, the arboreal pollen concentration (AP) in the Tenaghi Philippon record in
15 northern Greece (Tzedakis et al., 2006), the North Greenland temperature derived from the
16 NGRIP $\delta^{18}\text{O}$ record (‰ VSMOW, NGRIP-members, 2004) and the $\text{GL}_T\text{-syn}$ $\delta^{18}\text{O}$ synthetic
17 isotope record (‰ VSMOW, Barker et al., 2011). The grey shaded areas indicate interglacial
18 marine isotope stages (MIS; Lisiecki and Raymo (2005). For the legend of the mass
19 movement deposits see Fig. 2. High-resolution XRF data was filtered by using a lowpass filter
20 (5th order, cut off frequency: 0.064 Hz) in order to remove white noise from the data. Note the
21 logarithmic scale for TOC and for the total pollen concentration. Pollen concentrations are
22 from Sadori et al. (2015), lake water $\delta^{18}\text{O}_{\text{calcite}}$ from Lacey et al. (2015b), and S-ratios
23 representing hematite + goethite versus magnetite from Just et al. (2015).

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