

First of all, we are very grateful for the in-depth evaluation and constructive feedback on our manuscript. We appreciate the time and detail that both reviewers took which included screening of the associated manuscripts from the Ohrid group.

We are aware that the Lake Ohrid dataset and interpretation of the proxies is not complete at the present stage but we are convinced that this manuscript will provide a valuable contribution to better understand the terrestrial environmental and lacustrine conditions in Lake Ohrid. The revision is currently underway and will greatly benefit from the suggestions by the reviewers.

Comment on the age-model by Francke et al.

The course of the revision of the suite of papers in the Ohrid Special Issue, also the age-model for the sediment core has been revised as follows:

- Based on new results from tephrostratigraphy, one tephra age-control point was adjusted.
- age-control by correlating TIC to LR04 was removed
- correlation of TOC minima to increasing local summer insolation and winter-season length can be considered robust, because of the direct influence on lacustrine conditions. Therefore, this age-depth control was retained.

For details please see revision and reply by Francke et al.

Manuscript objective 1 (14219 line 4): "The first objective is to understand whether the variability in the magnetic mineral inventories can reveal changing environmental conditions in the catchment, beyond the observed general pattern of higher (lower) terrigenous input during glacials (interglacials)."

The main problem with this objective is, both in the introduction and throughout the manuscript, that the authors, based on an assumption of magnetic susceptibility (MS) (Fig 2d) being a direct proxy for terrigenous input, have already made an interpretation of higher observed terrigenous input during glacial periods and vice versa. There are two problems with this assumption. Firstly, the assumption that terrigenous input is the main contributor to the MS signal, while a valid hypothesis, needs to be demonstrated. It is possible that the MS signal in the lake environment is at least partly caused by magnetotactic bacteria. There is an interesting literature mini-review in the manuscript about magnetotactic bacteria (14225 lines 14-30). The authors need to use their knowledge of the methods detailed by the sources in this mini-review and apply it to their own data, to analyse if magnetotactic bacteria are generating their MS or not.

#1

Thank you for this valuable comment. The phrasing in the original manuscript was not clear and led to some confusion. We don't infer an absolute increase in terrigenous materials, but rather a relative increase, compared to bioproductivity. This will be stressed more clearly in the revised version of the manuscript.

Concerning the point of a potential influence of bacterial magnetite on the susceptibility, there is one important observations that was not explicitly discussed in the original paper. When comparing MS and SIRM in our Figure 2 it is well visible that in the upper section of core (MIS2-MIS8) increases in susceptibility are not mirrored by increases in SIRM. In case the susceptibility was carried by magnetosomes, an increase in SIRM would be expected, too.

Secondly, cross-plots of remanence parameters, that are often observed to be indicative for bacterial magnetite, e.g., $kARM/SIRM$ vs. $kARM/k$ and $SIRM/k$ vs. $kARM/k$ fail for the samples from Lake Ohrid (except the few samples from the uppermost Holocene section, that was already suspected to contain magnetosomes, as stated in the original version of the paper).

Moreover, the increases in MS co-varies with potassium and iron XRF intensities and with HIRM, suggesting a detrital source for MS. However, the magnetic extraction that we carried out for the revision of the manuscript provided evidence for a relative high proportion of siderite (see also Lacey et al.). Therefore, the contribution of authigenically formed paramagnetic minerals will be incorporated in the revised version (detrital versus biogenic contribution). See also reply to reviewer 1 #3.

We will work and modify the text and include the discussion about the origin of the susceptibility signal and clarify that we intend talking about relative changes.

Secondly, assuming for the moment that terrigenous input turns out to be the main/sole contributor to the MS signal, one must consider that any MS reduction during interglacial periods could be due to a primary productivity related increase of the contributions of TOC and CaCO₃ to the overall sediment accumulation (i.e. a dilution effect) and not directly related to a change in terrigenous input flux. Until the above issues are fully investigated, much of the discussion in the manuscript about changing lithogenic sediment supply and changes in the catchment environment can be considered mostly speculative.

#2

We didn't mean that the changes were only driven by terrigenous flux, for sure for such a discussion we would have to calculate mass-accumulation rates for biogenic and terrigenous sediments (as outlined above).

However, when we talk about changes in the magnetic properties, we don't use concentration dependent data.

The most important parameter is the S-Ratio, indicating more high-coercivity minerals compared to low-coercivity minerals. The concentration parameters HIRM and SIRM are shown to evidence that decreases in S-Ratio are not related to a loss in SIRM (which could be an indicator for selective reductive diagenesis of low-coercivity minerals), but to increases in HIRM. Low S-Ratios during the glacials indicate that in those climatic phases the contribution of high-coercivity minerals increased relative to magnetite.

Manuscript objective 2 (14219 line 7): "The second objective is to investigate proxies for the occurrence of magnetic iron sulfides for their capability to reflect hydrological and environmental conditions in the lake, because their existence as early diagenetic phases is strongly linked to the accumulation and decomposition of organic material." My overall view of the manuscript with regards to Objective 2 is that the various magnetic parameters are interpreted too quickly, with arrows on the figures suggesting too simply that certain magnetic parameters correspond directly with more/less of certain magnetic minerals. The authors need to begin with a more a comprehensive and basic analytical approach whereby elementary mineral magnetic properties (super paramagnetic, single domain, pseudo single domain, multi-domain, hardness, etc.) are first catalogued and considered, long before specific magnetic minerals are named.

For example, in the introduction and methods it is already assumed that SIRM/k is a proxy for "greigite", whereby Snowball and Thompson (1990) and Nowaczyk (2012) are cited as sources. The former source uses multiple analyses to identify greigite and simply notes that greigite tends to exhibit elevated SIRM/k values, not that SIRM/k on its own can be used as a general greigite proxy. The latter source doesn't explicitly mention SIRM/k being used as a greigite proxy. The authors do use GRM as an additional greigite indicator and in section 14223 lines 15-22 it is correctly noted that GRM acquisition can indicate the presence of greigite, coinciding in many cases with high SIRM/k values. However, the authors then conclude that, for intervals where they find high SIRM/k values and no GRM acquisition, that they still have greigite present, but that it simply failed to be recorded by GRM acquisition (which is possible), and that SIRM/k should be used as a general greigite proxy on its own. Such an assertion requires a more rigorous mineral magnetic and sedimentological investigation in order to identify what type of greigite (syn-depositional bacterial or post-depositional chemical) is present in the samples, which in turn can explain genesis and preservation conditions. Options include FORC analysis, TEM+SEM.

#3

We acknowledge the suggestions for possible biases of the "greigite proxies". In the revised version, we will also convey a more comprehensive study on the magnetic mineral assemblages. We will produce additional cross-plots indicating relationships between different magnetic parameters (e.g., grain-size dependence and compositional trends). These analyses will also help to evaluate the different proxies for greigite.

To support these data-based interpretations, as suggested, we performed SEM and edx-analyses on magnetic extracts, that will be used to identify magnetic mineral assemblages and relate those to the magnetic parameters.

The fresh results we obtained from SEM/edx analyses reveal large Fe-S nodules in extracts of samples characterized by high SIRM/k and zero GRM, while microcrystalline greigite is found in samples with significant GRM acquisition. The analytical work is still ongoing, but it seems that the coarse nodules are authigenic and have the same chemical composition as the greigite microcrystals. Thus, we infer that GRM acquisition is related, as it would be expected, to SD greigite minerals, while highest SIRM/k relates to the coarse Fe-S nodules.

Seeing that the authors seek to use greigite as an indicator for lake/sediment alaeoconditions, it is imperative that they ascertain what types of greigite are present in the various parts of the core, because different types of greigite form and/or are preserved under different circumstances. Post-depositionally formed chemical greigite can form due to the downward migration of isotopically heavy sulphides in the sediment (e.g. Barker Jørgensen et al., 2004; doi:10.1016/j.gca.2003.07.017) and could simply be related to sediment features that trap sulphides (a description/discussion of the sediment features would be helpful).

The authors seek to relate the presence of “greigite” (they do not state what type) in their record to the LR04 global benthic stack (14224 lines 0-10). Any apparent association between post-depositionally formed greigite and syn-depositional climate events cannot be interpreted by way of causality, so it is therefore imperative that the authors conclusively demonstrate where they have postdepositional chemical greigite and where they have syn-depositional bacterial greigite. See the work of Vasiliev et al. (2008, doi:10.1038/ngeo335) and Reinholdsson et al. (2013, doi:10.1016/j.epsl.2013.01.029).

#4

We have found evidence that the greigite is authigenic, and the size of the nodules often exceeds 50 μm , while micro-crystalline greigite is $\sim 1 \mu\text{m}$. We therefore conclude that the greigite has formed chemically during early diagenesis by processes mediated by Fe-reducing bacteria, which would mean that it would have formed shortly after deposition of the sediments.

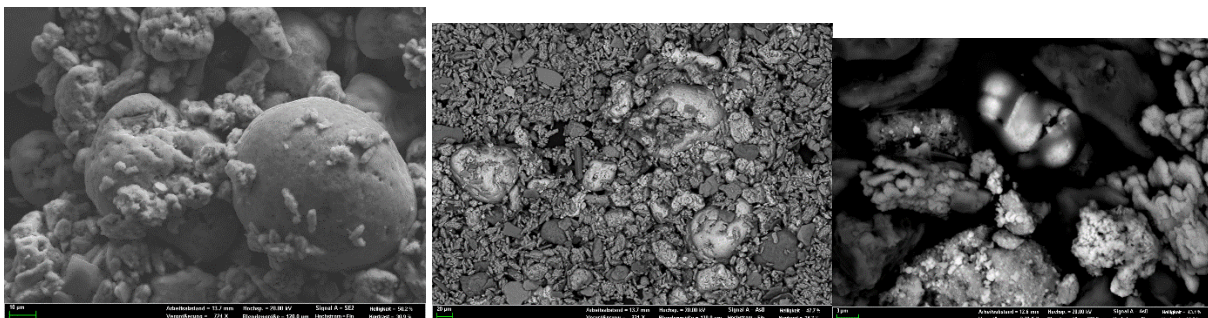
Almost syndepositional vs. post-depositional

Besides the Fe-S nodules a lot of siderite is contained in the magnetic extracts. According to Lacey (this issue) who worked on d18O of the siderite, it formed relatively fast after deposition.

Previously different scenarios were discussed for the co-occurring greigite and siderite.

If the Fe-S nodules had formed after siderite, a change in pore-water chemistry with sulfidic pore-waters would be required. We cannot test this, because no pore-water data are available. However, earlier publications from such settings show greigite overgrowth on siderite grains (Sagnotti et al., 2005, Weaver & Roberts 2005). We didn't observe so in the SEM. Below you can find an example of fine grained siderite together with some coarse Fe-S nodules.

Alternatively, siderite and greigite can form at the same time (e.g. Pye 1981, 1990) in recent environments if the rate of iron reduction is higher than the rate of sulphate reduction, which would be in-line with an inferred higher aerobic compared to anaerobic (sulfate-reducing) degradation. Some of the Fe-Sulfide nodules are perfectly round or within an organic coating. For these specific examples it appears likely that Fe-S crystallized in organic compounds (e.g., a pollen grain) and possibly also more irregular shaped particles could correspond to sulfides formed within microenvironments.



So, to sum up, based on three reasons, we expect greigite formation in the shallow subsurface:

- 1) Fe-S nodules are not growing on or replace siderite grains
- 2) Some of these nodules resemble the shape of organic compounds, or are even contained within an organic coating.
- 3) Relative Paleointensity data (just calculated as NRM/ARM) show a quite good correspondence between the Lake Ohrid core and reference records.

It is stated that “the samples containing greigite are associated to glacials occurring with low phases of eccentricity (Fig 2a).” Once again, the authors do not state what type of greigite. It’s difficult to see a significant correlation between the magnetic parameters and LR04 $\delta^{18}O$. The authors did carry out a fuzzy cluster analysis of six different magnetic, chemical and physical properties which they say “can basically be indicative of and impact the formation and preservation of greigite”. More information is required about what type of greigite is hypothesised as being formed (bacterial, chemical) and how. The rationale behind having (higher) lower TOC associated with (Cluster1)Cluster3 needs to be more clearly explained. It is likely that TOC XRF_{Fe} are causing the apparent interglacial/glacial grouping in the cluster analysis, and neither of these parameters is inherently indicative of greigite. TOC is heavily influenced by climate conditions (indeed, it was wiggle matched to LR04 by Francke et al. to produce the age model used in this manuscript). Hence, to use TOC in a cluster analysis to indicate greigite, and then claim that the cluster analysis shows a relationship between LR04 and greigite is not a valid approach. It would be interesting to see how the cluster analysis would look if TOC XRF_{Fe} were excluded and only the magnetic parameters were included.

5

We thank the reviewer for this remark on the inclusion of TOC and Fe-intensities into the cluster analyses. We agree that cluster assignment could have been influenced by the strong glacial-interglacial pattern in TOC and Fe intensities.

We repeated the cluster analyses and use now only magnetic properties. Off the same reasons for excluding TOC and Fe, we also excluded concentration magnetic parameters and only used relational proxies. Following this new analysis, the glacial-interglacial clustering in relation to the presence of greigite is still observed.

Concerning the discussion on the type of greigite, see reply #4.

Based on the two reviewer comments and together with the revision and reviewer comments on related papers from the special issue, we will re-evaluate the interpretation of extremely cold glacials in Unit 2, which contains significant greigite.

We are convinced that a distinct shift in lake conditions occurred. An alternative to the observation that those glacials were extremely cold, a rise in lake level or changes in hydrology and chemistry could be responsible for the observed shift.

- 1) Tectonics/Subsidence
- 2) Changes in water exchange between Lake Ohrid and Lake Prespa
- 3) Changes in water outflow from Lake Ohrid

At the present state, we cannot conclude which of those processes is important, and an evaluation needs additional investigations, including hydroacoustics and biological studies (e.g., shifts in communities), which have at this stage not been carried out down to the corresponding depth.

However, within this lower interval greigite is only present within glacial sediments. For this glacial-interglacial pattern, the process of a better ventilation of Lake Ohrid during glacials, and therefore increased aerobic degradation compared to anaerobic degradation (increase in sulfur reduction) is valid. At least if the greigite formed as an early-diagenetic phase (see also reply to reviewer 1).

Additionally, S-ratios can indeed help differentiate between low- and high-coercivity magnetic minerals. But why are only magnetite, goethite and hematite discussed (and also in Fig. 3c) as the only minerals affecting the magnetic assemblage coercivity? Greigite also contributes to the coercivity. The magnetic parameters will we discussed in more detail in the revised version. See Answer #3

All magnetic units need to be reported using mass specific standard notation used by mineral magnetists, to allow for easy quantitative comparison with existing publications (see technical comments). Much of the discussion mentions magnetic parameters simply as being “high” or “low”, whereas a quantitative description would enable a better comparison to existing mineral magnetic studies. Moreover, dry mass-specific units are important in such a long sediment sequence such as

the Lake Ohrid record, where downcore density changes due to sediment compaction can be expected.

6

We agree that corrections for down-core sediment compaction are worthwhile, so, we corrected all data with the wet-bulk density.

Since we are not working with pure magnetic minerals, we don't think it makes much sense to correct the magnetic parameters by dry-mass. There would still be the bias due to dilution of dia- and paramagnetic minerals, and therefore the comparison to published data would still be difficult (at least for concentration parameters).

When using ratios of magnetic parameters, a comparison is still valid.

Finally, I note that NRM data is not presented in the manuscript, nor are median destructive fields (MDF) of the NRM, which would be very useful for identifying properties of proposed magnetic minerals. The methods detail that NRM was measured with incremental demagnetisation to 100 mT, so these data should exist. Additionally, if the palaeomagnetic cubes have been subsampled with orientation in mind, then palaeomagnetic secular variation (PSV) data such as inclination and declination will also have been measured as part of the NRM measurements. Were the NRM data (and PSV data) judged to be not of sufficient quality for publication, have they been published already or will they be published in a separate manuscript? Elaboration is needed.

#7

We included MDF in our discussion. Moreover, the paleomagnetic data, especially estimates of relative paleointensity appear to be of good quality and will be published elsewhere, when the basic rockmagnetic properties will be groundtruthed.

Brief technical comments

(1) Both SIRM/k and ARM/SIRM can be indicative of magnetic grain size, depending on the number of (ferrimagnetic) magnetic minerals in the assemblage. The authors should look into this more and analyse any possible relationship between SIRM/k and ARM/SIRM

We have produced cross-plots which will show that SIRM/k is negatively correlated with ARM/SIRM, especially for those samples that carry a GRM. This discussion will be added into the revised version, at places where the occurrence of coarse grained Fe-S nodules are being discussed.

(2) There appears to be a minor typo in equation 2.

Ok

(3) The corresponding values of all the magnetic parameters from Fig 2 should be displayed for each sample in Figure 4.

We will revise Fig. 2 and include more and different magnetic parameters. We then will carefully reevaluate which parameters will be needed for the discussion of changing magnetic mineral composition in unit 1 (Fig. 4). However, there are some parameters in unit 1 (e.g. MDF, SIRM/kappa) that don't show variations in unit 1 and therefore we believe it is not necessary to include those parameters again in Fig. 4. Instead, we will show the cluster assignment again in Fig 4, because also there are a few samples that contain greigite (which will be included in the discussion of Fig. 4). By showing this color bar again, it will be easier for the reader to identify those specific intervals and cross-refer to Fig. 2.

(4) The correct mass specific unit notation that should be used:

- Magnetic susceptibility should be reported as m^3/kg
- SIRM should be reported as SIRM($A \cdot m^2/kg$)
- ARM should be reported as ARM(m^3/kg)
- SRIM/k should be reported as SIRM= (A/m)
- ARM/SIRM should be reported as ARM=SIRM($m=A$)

Will do.

(5) The division between Unit 1 and Unit 2 appears to be somewhat arbitrary. Perhaps a division based upon sedimentological properties would be more logical.

We will revise the reasoning why these units were divided (see also reply to reviewer 1, #1).