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Biogeosciences Discussions

Interactive comment on "A Holocene record of mercury accumulation in a pristine lake in Southernmost South America (53 S) – climatic and environmental drivers" *by* Y.-M. Hermanns and H. Biester

Anonymous Referee #2

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Review of Hermanns and Biester, A Holocene record of mercury accumulation in a pristine lake in southernmost South America (53oS) – Climatic and environmental drivers (Biogeosciences Discuss. 8: 6555-6588, 2011.

General

This well-written discussion paper presents an interesting hypothesis concerning the mechanisms regulating Hg deposition in pristine lakes (OM controls Hg export to lakes) and uses a detailed geochemical record covering 17,000 yr to evaluate the potential

role of climatic variability, catchment development, and external processes (e.g., volcanic activity) as mechanisms regulating metal deposition. I particularly like the use of process-oriented hypotheses to set up a general expectation for the study (increased OM influx should increase Hg accumulation in sediments); however, rather than simply use the sedimentary data to support or refute this expectation, the authors engage in highly speculative analysis of uncertain value (PCAs with low explained variance) to try and explain every apparent discrepancy. As a result, the combined Results and Discussion is long and unconvincing. Overall, I believe the main issue is that the investigators need better metrics of in situ aquatic production (e.g., fossil diatoms, pigments, lipids, etc.) as well as dissolved OM (DOM) to refine their interpretations and eliminate unwarranted guesswork. Further, I suggest that the paper better incorporate recent mass balance literature concerning the relative importance of dissolved and particulate organic matter (POM) to lakes and their sediments (e.g., Cole et al.) –at present there is the assumption that DOM influx is low relative to terrestrial POM, whereas research in diverse aquatic ecosystems suggests the opposite. As a result, assumptions concerning the capability of DOM in regulating bulk sediment properties seem unwarranted. Specific details, organized according to Discussion paper page and line number are presented below to help authors refine their analyses.

Specific

1. Introduction, p. 6556-6558. This section presents an excellent overview of how organic matter can influence the transportation of Hg in water (e.g., l. 6557). However, the literature concerning mechanisms regulating export of terrestrial DOM to lakes is less comprehensive, especially as regards the balance between terrestrial POM, DOM, and aquatic primary production as OM sources to lakes. Good reviews by J.J. Cole et al. 2007. Ecosystems 10: 171-84, Y.T. Prairie 2008. Can. J. Fish. Aquat. Sci. and L. Tranvik et al. 2009. Limnol. Oceanogr. 54: 2298-2314 provide the perspective that most carbon influx to lakes in forested catchments is derived from terrestrial DOM (about 10x more than POM influx). It would be useful to review these studies, to better

refine the specific expectations for change in DOM (and Hg) flux arising under different climate regimes.

2. p. 6558, l. 13-25. The paper suffers from imprecise predictions for the relationship between climatic variation, terrestrial development, and Hg transport. For example, if Hg influx was regulated by DOM inputs, then the two should covary most strongly when there are substantial changes in DOM flux – specifically, immediately after soils develop. Similar hypotheses could be described for each of the previously documented climatic changes (more precipitation, more DOM influx, more Hg influx, etc.). As it stands, the objectives are poorly defined, beyond a 'let's investigate' statement. This becomes more of a problem in the Discussion, where the authors expend considerable energy debating all possible mechanisms, rather than critically testing the central hypothesis.

3. Methods, p. 6559-6560 – In general, the methods seems very well done and reliable. In particular, the chronology is superb and the data density sufficient to investigate the statistical relationships among the various geochemical proxies. Unfortunately, I think the central problem of the paper is that the authors lack a unique metric to measure the various unique sources of organic matter to the sediments. For example, fossil diatoms or pigments (either extracted and quantified or measured by fluorescence) could be used to follow changes in lake production (See general books edited by J.P. Smol et al. Tracking Environmental Change Using Lake Sediments. Vol 3,. Kluwer), while recent work suggests that stable isotopes can be useful metrics of DOM influx, at least over the Holocene (e.g., Bunting et al. 2010. Limnol. Oceanogr. 55: 333-345, McKnight et al. 1997. Biogeochem. 36: 99-124).

While C/N ratios are proposed, they are mainly useful for interpretations of large-scale variation (10-20:1 ratio changes), and are much more problematic for finer details, particularly when there are high amplitude changes in terrestrial DOM influx, aquatic production, or algal composition. As a result, the narrative flips back and forth on whether the C/N is a good index of terrestrial OM or not – leading to excessive speculation and

an unconvincing argument.

I also think the authors need to use a constrained cluster analysis to better justify their three zones. I don't think it will change the story much, but it would provide a more objective demarcation of zones.

4. Results and Discussion. While I understand that the combination of Results and Discussion section is a common practise, I felt that this paper has ended up too long and speculative as a result of this narrative being in one section. In particular, details concerning the basic geochemistry tend to get repeated by stepping through OM, metals, Hg and statistics sections one at a time. Also, because there are no obviously testable objectives or hypotheses (point 1 above), the narrative is somewhat longer than needed, as it attempts to address all possible explanations.

5. Transitions among core Sections, Fig. 3, 4 and associated narrative. The most problematic aspect of the data is the apparent evidence that major changes in OM influx occur without corresponding changes in Hg deposition, such as occurs when local forest starts to develop at the base of the core, or between zones I and II. If OM were essential to transport Hg and regulate its influx, then I would have expected particularly strong covariance at these intervals. While it's possible there are extenuating mechanisms which override OM control (as touched on in the later Discussion), the more parsimonious explanation is that OM flux is not really that important – at least in these conditions.

6. p. 6563-6564. Although it's common to discuss the general relationship between proxies, I would find the arguments more convincing if statistical comparisons where used, rather than vague statements that match are 'quite good' (e.g., p. 6564, l. 14). Simple linear detrending of the time series, followed by Pearson correlation analysis would be fine, if the authors are assuming common regulatory mechanisms underlying the temporal variations.

7. p. 6563, l. 15, and l. 26. While it is probable that temperature and precipitation

had an effect, these arguments are not convincing. Similarly, p. 6565, l. 17-24 is not useful – as it says that sediments are a mix of terrestrial and aquatic OM (which is both obvious and vague).

8. p. 6566, l. 22-26, Table 3, and Methods. There is insufficient information provided to evaluate whether the soil and substrate survey was sufficiently comprehensive to allow general statements about "the catchment". Is Hg evenly distributed, or have past streams incised to deeper strata and accessed a 'point source' of Hg?

9. p. 6568-6572. Insufficient data were provided to evaluate whether the PCAs captured enough variance to be useful. At least in one case (Section II), the overall explained variation in the PCA is low (31%...on how many axes?), suggesting that the variation in proxies was not explained well be linear relationships. More importantly, I found the PCA data greatly over-interpreted and unconvincing. While the authors expend considerable effort evaluating the reasons for differences in relationships among Hg, C, N and erosion indicators in PCAs for sections 1, 2 and 3, again, I was left with the impression that they were trying to "save" a central hypothesis which had only weak support, rather than critically evaluate the mechanism.

10. p. 6572-73, section 3.5. Due to the high degree of speculation, I found several of the basic conclusions to be overstated (e.g., 6572 l. 17-19; 6573, 19-20).

Summary

The paper suffers from having a great central hypothesis, but an incomplete set of proxy metrics needed to test it. As a result, the paper is excessively long and speculative. The best means of resolving this issue would be to provide additional time series, particularly of OM sources. Alternately, the authors could focus on more statistical and critical testing, possibly resulting in the OM control mechanisms being rejected (with this data set).

Interactive comment on Biogeosciences Discuss., 8, 6555, 2011.