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Interactive comment on “Provenance of tetraether membrane lipids in a large temperate lake (Loch Lomond, UK): implications for GDGT-based palaeothermometry” by L. K. Buckles et al.

Anonymous Referee #2

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1. General Comments

In this manuscript, Buckles and colleagues examine a suite of samples from Loch Lomond and surroundings to better understand distributions and provenance of tetraether membrane lipids in lake sediments. This is a timely and important study, in that branched and isoprenoidal GDGTs in lake sediment archives are increasingly being used to reconstruct mean annual soil and lake temperatures, respectively. The application of these proxies is confounded by complex allochthonous and autochthonous sources, including growing evidence for in-situ brGDGT production within lake systems. By collecting a diverse sample suite that includes soils, river suspended sediment, river surface sediments, lake suspended sediments, lake surface sediments and

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a time-series of lake sediment trap particulates, coupled with detailed statistical analysis of the distribution of brGDGTs in both intact polar lipids and cores lipids, Buckles et al. present an attempt to disentangle the sources, production and delivery of brGDGT to lake sediments and the temperature signals these lipid distributions may carry.

This is a complex story, and the authors have done a very good job in arguing their main conclusions from this study, showing how these are supported by, or at least are consistent with, their data. These conclusions include: (i) substantial in-situ production of brGDGTs within the Loch Lomond lake system, although whether in the water column or within lake sediments (or both) is less clear; (ii) distinct patterns of brGDGT distributions among sample populations such as soils, rivers and lake sediments, suggesting that the straightforward rationale of brGDGT in soils followed by erosion, transport and deposition in lake sediments is incomplete or too simplistic; (iii) a caution that without full understanding of brGDGT distributions among possible sources to lake sediments, especially assessment of in-situ production, application of MBT/CBT-based temperature reconstructions rests on flawed forcing of calibrations between these brGDGT indices and air temperature that does not account for varying allochthonous contributions relative to autochthonous production; and (iv) a second caution that with in-situ production, the BIT index is not a robust indicator of soil OM contributions to sediments.

2. Specific comments

While overall sound, there are four aspects to this study that raise some questions. The first of these relates to the investigation of the soils as a source of brGDGTs. Clearly soils in this and many studies contain ample brGDGTs as both IPL and core lipids. However, the soil sampling design seems to emphasize vegetation and land-cover, and not the likelihood of a particular soil to actually contribute to downslope/downstream sediment archives. There is no way to know in this study that the sampled soils actually contribute to river or lake sediments; future studies may wish to better emphasize soils that are geomorphologically best primed to contribute sediments: adjacent to channels, gullies, and stream banks, along the steepest slopes, and downslope from

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any tilled/disturbed land-use. Thus, while this study shows that the soil brGDGT distributions are not identical to river SPM and sediment brGDGT distributions, this does not require that soils in general are not contributing brGDGTs to the rivers but rather only that the soils sampled in this study may not be the most important contributors to the rivers. The assertion that there may be in-situ production of brGDGTs in these rivers is potentially consistent with the data, but may also be explained by not sampling the soils/locations that best contribute particulates to the rivers. It is curious that the river sediments and river SPM samples show similar brGDGT distributions, as these cannot be the same populations of sediment. Coarse sand (river sediments) represents a different sediment source and flow regime than fine clays and silt in suspended sediments. It would be interesting to examine if the brGDGT distributions in the finest size fraction of the soil samples better matched these river sediment samples.

Secondly, the authors use intact polar lipids as reflection of “living” bacterial sources of brGDGTs, and core lipids as representing relict and more degraded material. However, little is known about the time scale for degradation of IPL. If it is slow relative to the timescale of sediment mobilization across the landscape, then even IPL may be integrating older sources and bacterial communities in soils. This can be important in regions where that has been substantial land-use change in recent decades or centuries, as brGDGT-producing soil communities may have changed yet the landscape may still export some relict IPL. The timescale of IPL degradation also is important for considering in-situ brGDGT production in lakes and especially rivers. In the Loch Lomond watershed, the timescale of storage of lipids in soils is much greater than the timescale of transport in rivers (many years versus a few days), thus it is difficult to imagine substantial IPL degradation during the short time that these compounds are transported in rivers. As the authors mention, the difference in %IPL between soil and river samples is not statistically significant, and thus in-river brGDGT is not required.

Third, it remains unclear if autochthonous production of GDGTs occurs in the lake water column as well as sediments. Clearly, the brGDGT distributions in lake sediments

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are distinct from those in lake suspended sediments and sediment trap samples, while lake suspended sediments and sediment trap samples are not very distinct from river samples. This supports brGDGT primarily within the lake sediments and not within the water column. However, the differences in lake SPM CBT ratios between core lipids and IPL, as well as the close capturing of temperature depth profiles by TEX86, supports production of branched and isoprenoidal GDGTs, respectively, within the water column.

Fourth, it would be valuable to know how river discharge and suspended sediment fluxes leading into Loch Lomond changed over the course of this study. Differences in GDGT concentrations, and for the sediment traps, the fluxes, may be in part explained by changes in suspended sediment concentration and discharge. Thus, different river flow regimes, and the landscape's geomorphic response to precipitation, runoff, and temperature, might result in different soil sources to the rivers and lake, different populations of soil-derived GDGTs as deeper soil horizons are accessed for erosion, and differences in the mixture between allochthonous sources and autochthonous production in the lake. Showing the concentration of brGDGTs in the sediment trap samples, in addition to their fluxes, may begin to inform this.

3. Technical corrections

p4190, l5-6: The authors should define here what is meant by Tierney10, Zink10, etc. This is clear much later in the manuscript that these are shorthand for different calibrations, but is not clear here where first introduced.

p4195, l1: The authors should provide more detail on the sediment trap deployment and recovery of sediment from it.

p4196, l16: The authors should describe the composition and concentration of the phosphate buffer used in this extraction

p4201, l4: The Methods section and Table 2 indicate that River Falloch SPM was

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sampled only once, in September 2011. The statement that brGDGTs in river SPM was constant throughout the year is not supported.

p4207, l21: Because some of the watershed is urbanized, and the uplands have widespread immature stony soils, it may be overstated to say that the catchment is fully soil covered.

Table A1: For clarity the authors should indicate which sample pairs in the MANOVA analysis are for IPL and which are for CL.

Figures: The ordering of figures is cumbersome, as Figures 4 and 5 are referred to before Figure 3 in the manuscript.

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