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Interactive comment on "Probing the fate of soil-derived core and intact polar GDGTs in aquatic environments" *by* F. Peterse et al.

Anonymous Referee #1

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In this paper, the authors incubated a soil sample from New Zealand in river and ocean water for a period of about 6 months and examined the abundance and distribution of core and intact polar GDGTs. There was no change in branched GDGT abundance and distribution during the experiment. In contrast, the total (i.e. core + intact polar) concentration of isoprenoid GDGTs substantially increased during the incubation, with a concomitant change in distribution leading to a decrease in BIT index of more than 0.1 unit.

The subject of the paper is of topical interest due to the increasing number of studies on GDGTs in both terrestrial and aquatic environments. The manuscript is well-written and well-organised. Nevertheless, I have several concerns about this study:

1) I do not think that the design of the experiment is appropriate to investigate the fate

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of soil-derived GDGTs in aquatic environments. The authors incubated soil samples in water using a soil:water ratio of 1:10. This corresponds to the incubation of soil in a water-saturated environment and just allows monitoring the growth of Archaea and branched GDGT source microorganisms in such conditions. Therefore, the aim and title of this paper should be modified to accurately reflect the design of the experiment. I would say something like: "Abundance and distribution of GDGTs in soils incubated under water-saturated conditions". The investigation of the fate of soil-derived GDGTs in aquatic environments should take into account the fact that soil-derived organic matter is present at low concentrations in such environments and also that natural conditions are complex.

2) I would have used a different control setup, where microbial activity is inhibited. I would have added some chemical agent such as zinc chloride to the mixture of soil and river/ocean water to stop all microbial activity. In contrast, the authors simply mixed distilled water and soil sample, leading to the growth of GDGT source microorganisms (especially Archaea) during the incubation in distilled water. Consequently, similar results were roughly obtained in terms of GDGT abundance and distribution, whatever the type of water used for the soil incubation (distilled, river or ocean water). This point is never discussed in the manuscript. In any case, the incubation in distilled water cannot be considered as a control one.

Detailed comments are given below.

Abstract

Line 12. As commented above, the authors can only say that the soil signature remains unaltered during the incubation under water-saturated conditions.

Line 21. The authors should take into account the fact that substantial amounts of brGDGTs can be produced in situ, thus overprinting the signature of soil-derived brGDGTs.

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Introduction

Page 11571, line 19. Please also refer to papers where branched GDGTs were investigated in peats.

Page 11573, lines 3-7. It would have been interesting to determine the origin of branched GDGTs in the Rakai River by comparing the abundance and distribution of these compounds in soil and water samples collected along the river.

Page 11573, lines 21-24. Please add a map showing where the soil and water samples were collected.

Page 11575, line 13. Please specify the average carryover of CLs into the IPL fraction.

Page 11576. Please specify the average analytical uncertainty of the CBT, MBT', BIT and TEX86 indices.

Page 11577, line 10. IPL-derived brGDGTs are less abundant than CL brGDGTs in all samples.

Page 11577, lines 11-13. Please be less assertive: these experiments suggest that soil-derived brGDGTs might not be sensitive to photodegradation.

Page 11577, lines 15-20. Please refer to Fig. 3.

Page 11578, lines 25-30. In the peat study by Huguet et al. (2013), there were no changes in brGDGT concentration, only in brGDGT distribution. This shows that the brGDGT distribution may be affected without any change in brGDGT abundance.

Page 11579, line 6. Remove the "but" and change "affect" by "effect".

Page 11579, lines 15-17. Please specify if the amounts reported correspond to total concentrations (i.e. CLs + IPLs) or not.

Page 11579, lines 22-23. The increase in total isoGDGT concentration is not so clear in Fig. 2, taking into account the analytical error on GDGT measurements. In addition, the

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fact that CL and IPL concentrations are reported separately does not help to visualize such an increase. Is this increase statistically significant? Please provide p-value.

Page 11579, line 26. Please refer to Supplementary Material.

Page 11580, lines 1-19. Please refer to the paper by Lincoln et al. (2014, PNAS) showing that crenarchaeol may also be produced by planktonic Euryarchaeota. The authors should discuss the fact that the isoGDGT increase was also observed in the incubation with distilled water (control setup). This implies that isoGDGTs are produced during the incubation, independently of the type of water used for the experiment. For example, IPL isoGDGT concentrations are higher in the incubations with distilled water than with river water. This partly questions the argument of nutrient availability affecting isoGDGT concentrations, since there are no nutrients in distilled water.

Page 11580, line 20. Once again, please specify if the increase in CL isoGDGT concentration is statistically significant.

Page 11580, lines 23-26. The authors never compare the results of the incubations in river and ocean water with those of the control incubations. Nevertheless, the proportional decrease of IPL-isoGDGTs with time is observed in all the incubations, even the control ones. This point should clearly be discussed in detail in a revised manuscript.

Page 11581, lines 1-5. This paragraph is not convincing. Indeed, the authors previously argued that IPL brGDGTs, which very likely possess phospho head groups, are stable because their side chains are ether-bound. Therefore, the same argument should be used for IPL isoGDGTs. Nevertheless, the authors use the opposite argument here, saying that IPL isoGDGTs very likely contain a phospho head group and are therefore rapidly degraded. Please be consistent in the discussion, using the same argument for the two types of GDGTs (isoprenoid and branched).

Page 11581, line 6. I would modify this sentence: “To evaluate if and how soil GDGT signatures are modified during the incubations (. . .)”.

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Page 11581, lines 10-11. I disagree with this sentence. The authors showed that there was no obvious increase or decrease in brGDGT concentration, but changes in brGDGT distribution could have occurred. Changes in concentration/distribution can occur independently.

Page 11581, line 12. Surprinsgly, the CBT was observed to increase at the end of the control incubation.

Page 11581, line 17. Please also refer to the Supplementary Table.

Page 11581, lines 26-30. What is the limit of detection of the brGDGTs with cyclopentane moieties? These compounds may be present in the IPL fraction, but at such a low concentration that they are not detected. In the present study, 10 g of soil were incubated. The extraction of larger amounts of soil (30-50 g dry weight) may be sufficient to detect IPL brGDGTs. In any case, it seems difficult to conclude that brGDGTs with one or more cyclopentyl moieties are produced at a lower rate than those without just because they are not detected.

Page 11582, line 15. Please be more moderate: "Our incubation results suggest that (. . .)".

Page 11582, lines 23-26. Tracing the absolute amount of brGDGTs in rivers may not be a reliable tracer of soil OC in all aquatic systems. It will depend on the proportion of soil-derived and in situ produced brGDGTs in the aquatic system investigated. If brGDGTs are mainly produced in situ (in the water column and/or sediment), measuring the concentration of brGDGTs will not help in tracing soil OC.

Page 11583, lines 5-6. What do the authors mean by "growth of specific microorganisms"? Please specify.

Page 11584, lines 1-5. In order to investigate aquatic branched GDGT production, I would have incubated river/ocean water with the corresponding sediment sample. This would be more relevant than the incubation with soil sample.

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Page 11584, line 13. The difference in branched GDGT distribution between the river SPM and catchment soil sample suggests that at least some branched GDGTs are produced in situ in the river (in the water column and /or sediment), even though this result is based on only one soil and one SPM sample. The hypothesis of riverine in situ production cannot be excluded and should be taken into account.

Page 11584, lines 21-22. The distribution of brGDGTs differ between the river and ocean SPM, and between the river SPM and catchment soil (Fig. 4). Therefore, I would not say that "soil brGDGT signatures delivered to the oceans will echo those entering the corresponding fluvial network".

Interactive comment on Biogeosciences Discuss., 11, 11569, 2014.