Distributions and sources of isoprenoidal GDGTs in Lake Lugano and other central European (peri-)alpine lakes: Lessons for their use as paleotemperature proxies

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Isoprenoidal glycerol dialkyl glycerol tetraether (isoGDGT) lipids occur ubiquitously in freshwater and marine environments. Since their distribution varies with temperature, sedimentary isoGDGTs have been used as proxies for the reconstruction of past continental climate for almost two decades. Yet, their application in lacustrine sediments is still not well constrained because the niches of isoGDGT-producing microorganisms in lakes are often ill-defined. Here, we study the distribution of isoGDGTs and their hydroxy derivatives (OH-isoGDGTs) in the water column of the deep (288 m), meromictic northern basin of Lake Lugano (Switzerland) using quantitative analysis of various pools of isoGDGTs and the stable carbon isotopic composition of isoGDGT-derived biphytanes. We provide strong evidence for archaeal water column sources of the isoGDGTs, based on comparison of lipid data with microbial diversity determined by 16S rRNA next generation sequencing. We find highest concentrations (i.e., 40 ng L 1) of crenarchaeol, the isoGDGTs specific for thaumarchaea, in suspended particle matter (SPM) from deeper (30e100 m) waters below the thermocline. This correlates well with thaumarchaeal 16S rRNA gene abundances, comprised by a single thaumarchaeote of the order Nitrosopumilales. The concentrations of OH-isoGDGT with 0e2 cyclopentane rings follow this profile, suggesting an identical archaeal source. In the deeper anoxic waters, the archaeal community changes substantially and was comprised of various members of the Bathyarchaeota, Diapherotrites, Euryarchaeota, and Woesearchaeota. This change is accompanied by a changing distribution of isoGDGTs with a high contribution of GDGT-0 with a polar head group, and a more negative d13C value of the acyclic biphytane derived thereof. Comparison of the isoGDGT composition (distribution and d13C) in the surface sediment with that of the sinking particle flux studied over 1 year at three depths indicates substantial downward transport of isoGDGTs to the sediments from the waters between the thermocline and the anoxic hypolimnion, but not from the deeper (>100 m) waters. The relatively low value for the isoGDGT-based TEX86 value (ca. 0.40) in the surface sediment is similar to that of the in-situ produced isoGDGTs in the waters below the thermocline, and is consistent with the year-around low water temperatures (<6 C). We also analyzed the isoGDGT composition in surface sediments of 36 additional (peri-)alpine lakes in Central Europe; their isoGDGT concentration and distribution show a clear-cut difference based on lake size. In relatively large and deep lakes, like the Lake Lugano North Basin, the crenarchaeol concentration was much higher than in smallsized and shallow lakes, because Nitrosopumilales spp. have seemingly no niche in shallower waters (0 e30 m depth), and production, and hence preservation in the sedimentary

record, of crenarchaeol (and other thaumarchaeal isoGDGTs) is much lower than in large lakes. We argue that in the mid-latitudinal large and deep (peri-)alpine lakes Nitrosopumilales spp. generally thrive in cold waters below the thermocline, where temperature is largely not affected by fluctuations in mean annual air temperature. Therefore, the TEX86 paleothermometer is relative insensitive to atmospheric temperature changes in these lakes. In the small-sized and shallow lakes and ponds crenarchaeol can still be present but is probably mainly derived from Nitrososphaerales spp. (either from in-situ production or soil erosion). These thaumarchaea are characterized by a higher relative content of the crenarchaeol isomer than the Nitrosopumilales spp. at the same temperature, which may compromise accurate paleotemperature estimations using the TEX86 for small/shallow lakes.
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