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Identification of a novel lipid biomarker in lake sediments: Implications for the aquatic production of branched GDGTs

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Branched dialkyl glycerol tetraethers (brGDGTs) are bacterial membrane lipids that occur ubiquitously in soils worldwide. Their relative abundances (which form the MBT and CBT indices) correlate with mean annual air temperature (MAAT) and pH, making them potential proxies for paleoenvironmental reconstructions in sedimentary archives with organic matter input of from terrestrial environments. In lake sediments, however, brGDGT distributions mostly relate differently to MAAT compared to soils. A likely cause for this discrepancy is in situ production of brGDGTs within the lake water column and/or -sediments by microorganisms possessing a membrane physiology that is different from soil bacteria. Until to date, existing analytical methods did not allow the distinction between soil-and lake-derived brGDGTs, complicating paleoclimate reconstructions in lacustrine settings. In order to decipher differences in the structure and the relative distribution of brGDGTs in lake deposits versus soils, we analysed samples from 35 Swiss alpine lakes and their catchments using an improved HPLC protocol. Our data revealed a novel isomer of the hexamethylated, non-cyclic brGDGT, which was previously co-eluting with the recently described 5- and 6-methyl isomers. About half of the 35 investigated lakes contained this compound where it accounted for 1 – 11 % of total brGDGTs. We were able to isolate the previously unknown isomer by multi-step preparative HPLC. The structure of the molecule was assessed by determining the identity of its alkyl chains by ether cleavage with HI and subsequent analysis with gas chromatography quadrupole mass spectrometry (GC-MS). In addition, the carbon isotopic composition of the fragments was determined to trace their biological source. In contrast to lake sediments, soil samples analysed from the watersheds of the studied lakes did not contain the newly discovered GDGT, providing evidence for in situ production in aquatic systems. This novel isomer is the first identified, lake specific brGDGT and we expect that its recognition can significantly improve the interpretation of lacustrine brGDGTs distributions.