A MICRO AND MESOCOSM APPROACH TO UNDERSTANDING THE RESPONSE OF BRANCHED GDGTS TO ENVIRONMENTAL PERTURBATION

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Originally described in peatlands, but now found in soils, lakes, peats, and near-shore marine sediments, branched glycerol dialkyl glycerol tetraethers (brGDGTs) are thought to be membrane lipids synthesized by bacteria. The number of methyl groups and cyclopentane moieties within the core structure, accounting for 15 known forms, appears to respond to environmental factors such as pH and temperature (Weijers et al., 2007). Given the environmental sensitivity and the ubiquity of these lipids, brGDGTs have gained relevance as a possible paleoclimate proxy, leading to the proposal of the methylation index of branched tetraethers (MBT) and cyclisation index of branched tetraethers (CBT) as proxies for temperature and soil pH. Furthermore, previous research has shown that this proxy may also be applicable in lacustrine environments, although the distribution of brGDGT species in soils and lakes appears to be different (e.g., Tierney and Russell, 2009).

Despite the body of research on brGDGTs, the organisms responsible for their production largely remain elusive. This gap in knowledge has precluded in-depth study of the response of these lipids to environmental perturbation under controlled conditions, limiting the use of this proxy. To this end, we studied the production of brGDGTs in lake water using a series of micro and mesocosm experiments which allowed us to modify environmental factors while preserving the natural conditions of the sample. For this work, we collected water samples from Rose Canyon Lake, Arizona, and incubated them in either a mesocosm (two 20L tanks, one containing synthetic sediment) or a microcosm (1L bottles with varying conditions). Water samples were then filtered through a vacuum filtration system; lipids from the filters or solid samples were later extracted through an Accelerated Solvent Extraction system. Branched GDGTs were quantified comparing peak areas with a C_{46} internal standard using an HPLC-MS as described by Hopmans (2016). For this work, we calculated the indices CBT' and MBT'_{5Me} using the empirical equations proposed by De Jonge et al. (2014).

The results from the mesocosm experiments show a drastic change in both abundance and distribution of brGDGTs in the suspended particulate matter (SPM) of the two tanks as compared with samples taken directly from the lake. The experiments show a significant decrease in the total abundance of brGDGTs in the tanks, with a switch from the 5' to the 6' isomers. Interestingly, the synthetic sediment showed a continuous increase in brGDGTs which maintain a distribution similar to the SPM. The microcosm experiments tested the influence of three environmental factors: abundance of carbon sources, pH, and temperature. Incubating the microcosms without additional carbon sources other than those already present in the lake water resulted in an increase in the abundance of brGDGTs with a distribution similar to that observed in the mesocosm experiments. On the other hand, incubating the sample with 0.1 g of glucose produced a smaller increase in total abundance, but a distribution similar to that of the original lake water SPM. To test the influence of pH on brGDGT concentrations and distribution, we incubated a control experiment (pH 7.9), and two parallel experiments at pH 7.1 and 4.6 (Figure 1). We observed an increase in the tetramethylated brGDGTs and corresponding decrease in CBT' as the pH decreased, in agreement with the empirical observations of the change in CBT in modern soils (De Jonge et al., 2014). Additionally, 6' isomers appear to also respond in a direct way with pH, favoring the 5' forms at lower values. Finally, the temperature microcosms incubated at room temperature (control, 22˚C), 4.5˚C (cold) and 26.8˚C (hot) showed an increase in the tetramethylated brGDGTs and corresponding increase in MBT'_{5Me} as temperature rose (Figure 1). Our results also show a sharp increase in the 6' isomers of the acyclic brGDGTs.

Overall, these experiments provide some of the first evidence of growth and alteration of brGDGTs under laboratory conditions, which could be used to build better models to interpret this particular paleoclimate proxy in lacustrine sediments. Our data suggest that the methylation and cyclization of brGDGTs responds in the expected direction to temperature and pH, but that food or nutrient limitation may also impact brGDGT structure. Although the identity of the organisms remains unknown, constraining the optimal growth conditions could aid in generating a more effective approach to culturing them.

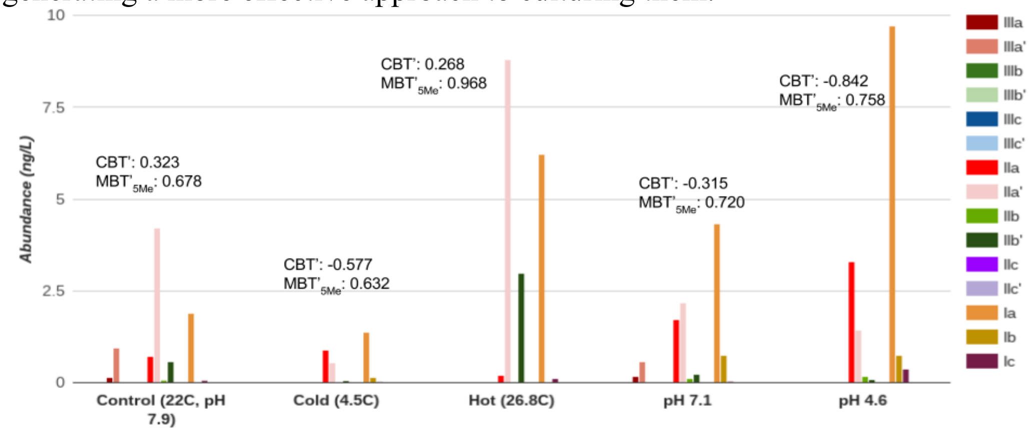

Figure 1. *Abundance (ng/L) of the brGDGT species in the pH and temperature microcosm experiments.*

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