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Extensive processing of sediment pore water dissolved organic matter during anoxic incubation as observed by high-field mass spectrometry (FTICR-MS)

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Terrestrial particulate and dissolved organic matter enters freshwater bodies, like streams, rivers, and lakes, where it is transformed by biotic and abiotic reactions. Depending on conditions, variable fractions of this organic matter are transferred to the sediments. Here, aerobic and anaerobic microbial degradation processes further change organic matter composition and structures, also producing greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2).

In this study, we used ultrahigh resolution mass spectrometry (FTICR-MS) to trace differences in the composition of solid-phase extractable pore water dissolved organic matter (SPE-DOM) isolated from surface sediments of three Swedish boreal lakes before and after 40 days of experimental anoxic incubation, alongside with determination of CH_4 and CO_2 production rates by gas chromatography. Greenhouse gas production rates varied considerably among replicates; CH_4production ranged between 5.9 $\pm$ 3.1 and 11.8 $\pm$ 5.8 nmol d^{-1} g_{dw}^{-1}, while CO_2rates were between 77.6 $\pm$ 5.9, 113.5 $\pm$ 11.6 nmol d^{-1} g_{dw}^{-1}. In general, proportions of CHO compounds slightly decreased during incubation, while CHNO compounds were enriched. Consumed molecules were less specific indicators of lake sediment alteration than particular compounds found more abundant after incubation, such as certain oxygenated aromatics and carboxyl-rich alicyclic acids (CRAM). Computation of the average carbon oxidation state in CHO molecules of sediment pore water DOM revealed depletion of both highly oxidized and reduced CHO molecules, and formation of rather non-labile fulvic acid type molecules. Svarttjärn (small lake and catchment area) presented increase in CRAM molecules during incubation, whereas lignin- and tannin-like compounds were enriched in Bisen (larger lake and catchment area), suggesting selective preservation of these non-labile aromatic compounds instead of recent synthesis. NMR spectra confirmed processing of sediment pore water organic matter during incubation, with depletion of certain lipids and formation of small oxygenated aliphatic molecules.

Hence, SPE-DOM after incubation may represent freshly synthesized compounds, leftover DOM (which should be composed of intrinsically refractory molecules) and/or microbial metabolites which were also not consumed in our experimental incubation. The pronounced change in molecular DOM composition during the incubation indicates that diagenetic modification of organic matter can be substantial compared to complete mineralization, despite the low fraction of the total DOM being mineralized to CO_2 and CH_4. These results highlight the importance of linking biogeochemical processes with studies of molecular composition using high resolution non-target approaches.