Advancing mass spectrometry imaging of sediments: lipid biomarker analysis with sub-mm resolution

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In 2014 we demonstrated the feasibility of analyzing diagnostic lipid biomarkers, more precisely archaeal GDGTs, in μm-scale sized spots directly on undisturbed sediments, without prior extraction [1]. Analyte ionization and detection involved laser desorption ionization coupled to Fourier-transform ion cyclotron resonance mass spectrometry (LDI FT-ICR-MS), and provided collections of mass spectra generated on the sample’s surface at sub-mm spatial resolution, i.e. a fine-scale image of GDGT distribution.

The final goal of our approach is to interrogate suitable archives, primarily laminated and dated sediment cores, for short-term climatic and environmental variations, and to investigate the nuances of the spatial distribution of biomarkers in their sedimentary context. Thereby we aim to advance molecular stratigraphy to a new level, regarding both the temporal resolution of records and the understanding of proxy formation.

On our way to advance this technology to the routine level, we have (a) developed suitable protocols for preparing sediment segments for analysis while maintaining their spatial integrity and (b) extended the spectrum of analyzable compounds beyond GDGTs. The combination of an optimized sample preparation pipeline, target-specific instrumental settings and the choice of a suitable chemical matrix to aid ionization (matrix assisted LDI, MALDI) now allow the detection of other informative biomarkers. For example, long-chain alkenones and associated SST estimates are useful to complement GDGT-based data, while fatty acid analysis in negative mode, or n-alkane detection enabled by the use of silver-based matrices provide access to other meaningful proxies. Latest developments in this initiative and the first applications to environmental samples will be presented.