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## Evaluating the potential of FT-ICR-MS to identify source-specific markers and trace molecular transformations in particulate organic matter

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On long timescales, carbon fluxes in and out of rock, soil and biological reservoirs control carbon dioxide concentrations in the atmosphere and therefore modulate global climate. For example, the transfer of particulate organic matter (POM) from mountain ranges into rivers and subsequent burial in the ocean constitutes a carbon sink from the atmosphere if the eroded POM is sourced from vegetation and soils. In contrast, the transfer and burial of rock-derived petrogenic POM has no effect on atmospheric carbon concentrations. However, if petrogenic POM is remineralized during transfer, often mediated by microorganisms, it constitutes a carbon source to the atmosphere. To evaluate the net effect of these processes, it is essential to understand sourcing, mobilization and fluxes of POM. Bulk stable and radiogenic isotopes as well as a range of lipid biomarkers and their stable isotope ratios have been used to trace the sourcing and transfer of POM. However, these methods are limited to the distinction of broad classes of source materials and do not contain information on potential molecular transformations during organic matter mobilization and transport.

Fourier Transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR-MS) allows the simultaneous measurement of a large range of compounds (i.e. hundreds to thousands) and has been applied in dissolved organic matter research to trace different sources as well as to identify transformations. FT-ICR-MS measurements on solvent-extractable POM provide direct information on the compositional variability of POM with a much larger analytical window than single biomarker or bulk isotope analysis and additionally might allow to trace transformations of POM upon mobilization.

Here, we test this method to decipher the different sources of POM and their mobilization in the upper catchment of the trans-Himalayan Kali Gandaki River, which sources petrogenic POM from abundant Jurassic sediments as well as biospheric POM from aged and modern soils. We evaluate the potential of the high-resolution molecular dataset to identify new marker compounds for specific organic matter sources and, by applying indicator species analysis, to statistically identify indicator compounds. In a second step, we evaluate the potential to trace transformations across the mobilization step from each specific organic matter source to particulate organic matter in

river sediments.

We found a large number of source-specific elemental formulas for biospheric carbon and strong heterogeneity for bedrock-derived organic carbon which highlights that petrogenic carbon varies in molecular composition depending on its (geological) origin. Regarding transformations, we found a loss of source-specific formulas during mobilization of organic matter, related to intrinsic chemical properties. These formulas were characterized by a higher number of double bond equivalents, a higher nominal oxidation state and higher oxygen content than formulas shared between riverine POM and source organic matter for all sources, which is consistent with the preferential loss of more labile organic matter during transport and/or mobilization. Overall, our study highlights the potential of FT-ICR-MS to identify molecular-level transformations of solvent extractable lipids along the source-to-sink pathway of sedimentary organic matter.