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Deciphering Bulk δ^{13}C and δ^{15}N Isotopic Compositions using Density-gradient Centrifugation: A Maceral-specific Approach to Isotopic Analysis

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A novel combination of density-gradient centrifugation (DGC), organic petrography, and elemental analyzer isotope ratio mass spectrometry (EA-IRMS) analysis allows maceral-specific isotopic compositions and C/N ratios to be determined for Type III (Pennsylvanian and Cretaceous coal) and Type II (Devonian black shale) kerogens. This approach has significant potential for interpreting isotopic results beyond that provided by the isotopic analysis of bulk organic carbon and nitrogen or even by compound-specific isotope analysis given the possible heterogeneity of organic matter (OM) sources and the relatively low abundance of soluble OM compared to kerogen. Our approach provides source specificity by separating kerogen macerals prior to isotopic analysis.

An important, yet often overlooked, limitation of bulk isotopic analysis of organic matter in sediments and rocks is that it reflects the relative contribution of the different constituents and their respective compositions. As a result, variations in bulk isotopic composition are difficult to properly interpret without detailed information regarding a) the relative amounts of each individual constituent (or maceral), and b) the isotopic composition of each entity. The novel combination used here allows us to constrain both relative abundance and isotopic composition of organic constituents in a sample. DGC has been used to separate individual macerals (the microscopically identifiable components of kerogen and coals) to allow analysis of relatively pure ($>$ 95%) end-member components. We have recently used this technique on Devonian black shales and have been able to determine a 1.5 per mil difference in δ^{13}C between

marine components (alginite and bituminite), as well as a 3 to 4 per mil difference between terrestrial and marine components. Similarly, analysis of Type III kerogen (coal) indicates shows a >2 per mil difference in δ^{13}C between macerals in the same sample. This approach will ultimately allow more reasoned, comprehensive interpretations of bulk isotopic trends through sedimentary sequences.