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Puzzle Game of Isotope Effects: Fingerprint and Reconstruct the Fates of Organic Molecules

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Stable isotope effects, as the key to unlock the code of kinetic, thermodynamic, biochemical, and even femto-chemical processes, have exhibited a power of blooming the breakthroughs in various areas of studies outside its origin in geology. However, its power gets limited by the current analytical techniques and theories, with which we hardly unravel a comprehensive observation of isotope effects in laboratory settings, simulations, or natural environments.

Our work aims to use the isotopic signatures of various environmental organics for environmental forensic and biogeochemistry studies. To this end we develop and validate novel stable isotope analysis methods, in view of advancing the next-generation mass spectrometry techniques (e.g., ESI-Orbitrap mass spectrometry), and establishing groundbreaking stable isotope effects theories.

Tracing the fate of various organic molecules (e.g., in soils, groundwaters, marine environments, fossils, and biomaterials) can be challenging because the precursors and/or products are either unknown/mixed, common in nature, or are labile and/or difficult to measure. Moreover, multiple transformation processes may occur simultaneously, rendering them difficult to distinguish or apportion based solely on tracking the concentrations. To resolve these challenges in environmental forensics, 2,4-dinitroanisole was used as a model compound for deciphering the transformation mechanisms using stable isotope effects, with the aim of more effective and efficient fate-tracking and source-identifications in field remediation efforts [1].

To explore broader applications in biogeochemistry, ESI-Orbitrap methods were developed and validated for a series of international standards (e.g., amino acids, urea), and in-house referencing materials (e.g., fatty acids), with nanomole-level analytes injected to achieve a precision for ^{13}C, ^{15}N, ^{18}O and ^{37}Cl of < 0.2 ‰. The capability for simultaneous high-precision analysis of multi-element isotope ratios in oxyanions and biomolecules is predicted to boost innovative discoveries and applications in geosciences and biochemistry. For example, in Earth's aqueous systems and metabolisms, reaction pathways may involve rate-determining steps of C-O or N-O bond cleavage or formation, causing relatively large primary oxygen kinetic isotope effects that could be diagnosed or clearly indicated by carbon and nitrogen isotope analysis.

References:

[1] C. Wang, M. E. Fuller, J. Murillo-Gelvez, et al. (2024) ES&T 58, 5996-6006.