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Advancing Organic Carbon Source Discrimination Using Ramped Pyrolysis Oxidation

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Organic carbon (OC) is a fundamental component of the global carbon cycle. OC of different sources vary in terms of form, age, and properties, and play distinct roles in regulating atmospheric CO_2. Therefore, accurately distinguishing between OC sources is crucial for understanding carbon cycling at Earth's surface. Traditional methods, such as carbon and nitrogen elemental ratios and their isotopes, have been widely employed as effective indicators for source discrimination. However, challenges emerge when these methods are applied in certain ecosystems, particularly in cases where carbon and nitrogen isotopic values overlap significantly between endmembers or in watersheds with highly variable compositions.

Ramped Pyrolysis Oxidation (RPO) has emerged as an innovative technique capable of deconvoluting the thermal and oxidative spectra of OC, offering a novel fingerprint for OC properties. Despite its potential, RPO remains underutilized for source discrimination when compared to traditional isotopic or biomarker methods. In this study, we present a novel application of RPO, transforming thermal decomposition profiles into matrices and utilizing an endmember mixing model as a matrix calculation tool to determine optimal source contributions. To validate the robustness of this method, we conducted binary, ternary, and quaternary endmember mixing experiments. The results demonstrated that the deviation in source contribution estimates was within 5%, with an average maximum deviation of 3±2%, outperforming conventional source discrimination methods in both accuracy and consistency.

Subsequently, we applied this approach to quantify the sources of particulate organic carbon (POC) in the Buha River and Lake Qinghai, located on the northeastern Tibetan Plateau. Using the newly validated RPO endmember model, we estimated the contributions of soil, vegetation, and rock to the POC. Our findings indicate that (1) approximately 90±3% of riverine POC originates from soil organic carbon erosion, with a mean radiocarbon age of 2,960±130 years; and (2) climate significantly influences the evolution of organic carbon erosion in Lake Qinghai. This study highlights the applicability and advantages of the RPO method in tracing OC sources and reconstructing erosion histories