

MOLECULAR AND ISOTOPIC DISTRIBUTIONS OF ALIPHATIC SIDE CHAIN IN CARBONACEOUS

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Introduction: Carbonaceous chondrites are the most primitive stony meteorites which carry a record of organic chemical evolution dating back at least 4.5 billion years. Thus, the organic matter in carbonaceous chondrites potentially records a succession of chemical histories that started with reaction in the interstellar medium and ended with reactions driven by hydrothermal alteration in the meteorite parent bodies [1]. A thorough understanding of the organic compositions of carbonaceous chondrites is highly relevant to better understand evolution of life on earth. Organic materials in carbonaceous chondrites range from a simple observed organic compound in interstellar clouds, the formic acid [2] to a complex macromolecular network of Insoluble Organic Matter (IOM) [3]. The IOM fraction has been characterized using different analytical approaches [4,5,6]. However, none of the previous studies was able to specifically characterize aliphatic side chain distributions on the aromatic nuclei. Various degradation methods yield predominantly aromatic moieties. Isotopic compositions of aliphatic side chains and their differences relative to aromatic nuclei have not been studied. The aim of this work is to understand the chemical structures and isotopic variations of the aliphatic side chains in the meteorite IOM. We studied different types of meteorites [Orgueil (CI1); ALH85013 (C2), ALH84033 (CM1/2), Murchison (CM2), EET87770 (CR2) and MET00430 (CV3)].

Procedures: The powdered samples were first extracted with double-distilled water for 24h at 110°C under vacuum. After extraction, the solution were adjusted to pH > 10 using NaOH solution and subjected to rotary evaporation. The volume-reduced solution is then transferred to a 2 mL vial for further analysis. The solvent extraction residues were treated with HCl 4 mol L⁻¹ and then demineralized using a mixed CsF-HF solution [6,7]. Basically, the CsF-HF solution attacks the silicate minerals by producing HCl soluble fluoride salts and liberating the IOM. The IOM samples were subsequently treated with ruthenium tetroxide to oxidize the aromatic cores and convert the aliphatic side chains into monocarboxylic acids (MCAs). The water solution in both pristine meteorite and IOM extractable were analyzed quantitatively using solid phase micro extraction (SPME) coupled with GC-FID, followed by GC-MS for identification and GC-IRMS for isotope ratios. SPME provides excellent recovery for monoacids.

Results and discussion: 15 to 50 MCAs were identified and quantified in both water soluble extracts and RuO₄ treated IOM samples (for examples of GC-FID traces see Fig. 1). The concentrations of monoacids in water extract-

able fractions ranged between 0.06 to 1.4 $\mu\text{mol g}^{-1}$ of meteorite, while in the IOM degradation products varied from 2 to 35 $\mu\text{mol g}^{-1}$. The molecular distributions, especially for the branched monoacids, are very similar between water soluble fractions and RuO₄ treated IOM samples for Murchison and EET87770 (Fig.1). Such structural similarity and inheritance between IOM and soluble acids suggest that these compounds have similar or the same origins, i.e., a common precursor may be responsible for their formation. There is also striking similarity in the distribution of MCAs released from RuO₄ treated samples, regardless of their petrographic classifications (e.g., CM1, CM2, CR2 and CV3). In contrast, the soluble acids from different meteorites show more variable distributions. There are fewer MCAs in soluble fractions than in RuO₄-treated IOM fractions in most samples, suggesting post synthetic processes are more likely to affect the soluble compounds than the IOM. Carbon isotope ratios of most monoacids range from 0 to -80 ‰ (vs. PDB), which are indistinguishable from terrestrial organic compounds. However, meteorite MCAs are highly enriched in deuterium, with δD values ranging from a few hundred to >+2000‰ (vs. VSMOW). There is also a good correlation between the isotope ratios from the water extractable and those released from IOM degradation. Our isotope data once again suggest a possible parent-daughter relationship between soluble and insoluble organic fraction in carbonaceous meteorites.

Conclusion: We developed a new method to characterize the aliphatic side chains of IOM at the molecular level. The new method allow rigorous structural and quantitative evaluation of side chain length and distributions that cannot be achieved by previously published methods. Up to 9 carbon atoms are present in the aliphatic side chain of aromatic nuclei of the IOM, significantly longer than previously suggested 1 to 6 carbon atoms.. The strong similarity in molecular distributions and isotopic ratios of water soluble and IOM derived monoacids indicate that IOM and soluble fractions have the same origins.

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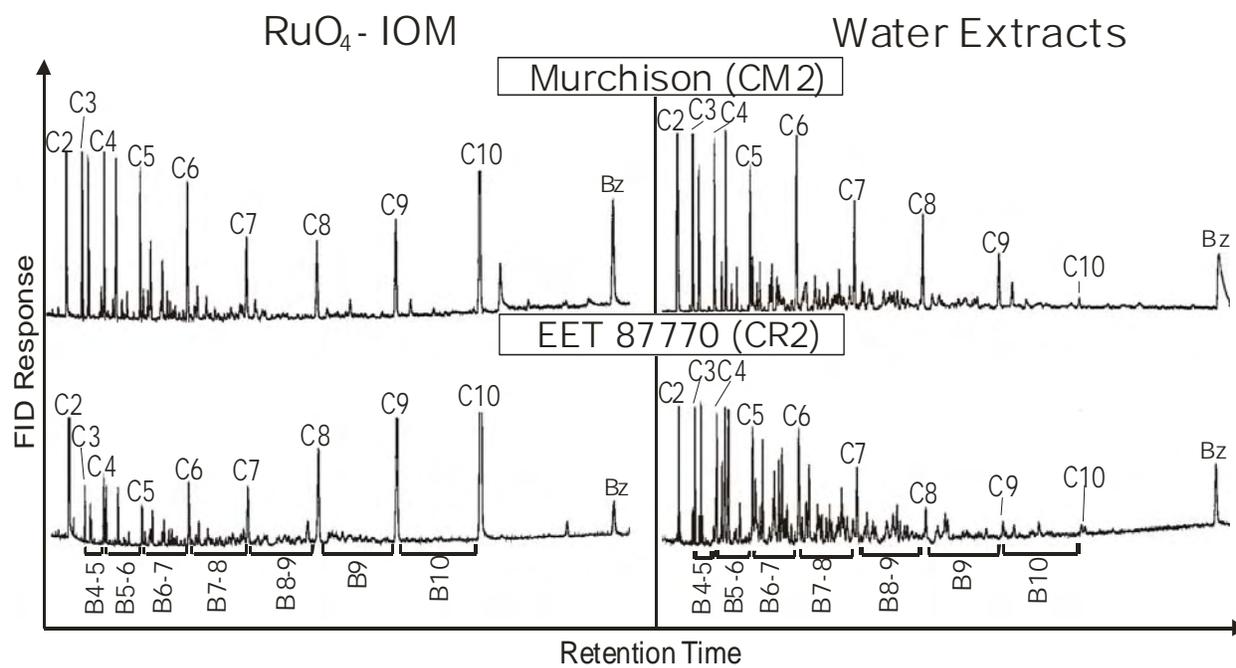


Figure 1 - Gas chromatograms showing the distribution of MCAs in Murchison and EET 87770. C2 to 10=straight chain; B=Branched and Bz= Benzoic Acid