ISOTOPIC CHARACTERISATION OF KEROGEN-LIKE MATERIAL FROM THE MURCHISON CARBONACEOUS CHONDRITE

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Stepwise extraction of C, H and N from acid-demineralised residues of carbonaceous chondrites, by either pyrolysis or combustion, reveals considerable variability in $\delta^{13}C$, $\delta^D$ and $\delta^{15}N$. Even after adjustment of the data to correct for the presence in the residues of traces of isotopically anomalous C and N of apparently circumstellar origin [1], substantial variability remains, attributable to isotopic inhomogeneity in the kerogen-like material that constitutes most of such a residue. This inhomogeneity is manifested also by pronounced intersample variability, within individual meteorites, most marked for the H isotopes.

Our data for a stepwise pyrolysis and a stepwise combustion of such a residue, taken together with results reported by Robert and Epstein [2] for pyrolysis of a less completely demineralised residue, suggest that kerogen-like material in Murchison may be characterised by three, or possibly four, isotopically distinct components. (The fourth component, revealed as low-molecular-weight hydrocarbons evolved during pyrolysis, may have been produced by isotopically selective cracking of the major aliphatic component, possibly during the extraction process itself.) The relative proportions of the different components are not constant among different Murchison samples.

Adopting a consciously oversimplified model of the kerogen-like material as consisting of a series of polycyclic aromatic moieties connected by aliphatic bridging chains [3], we identify two of the components as being aromatic in nature, with the third, and possible fourth, being aliphatic. It seems likely that the reality is more complicated than this.

Both aromatic components appear to have the same $\delta^{13}C$ value: $-18.7^{\circ}/oo$ (rel. PDB). The major aromatic component, released at relatively low temperatures, is enriched in D ($\delta^D > +1500^{\circ}/oo$, rel. SMOW) but relatively depleted in $^{15}N$ ($\delta^{15}N < -6^{\circ}/oo$, rel. AIR). The minor aromatic component apparently reverses these trends ($+350 < \delta^D < +550 ; \delta^{15}N > +60$) but the inferred values are sensitive to possible contributions by any inorganic H and/or N that might have survived demineralisation. The presence of such inorganic material is suggested by anomalously high H/C and N/C ratios for some of the high-temperature steps, but, barring exaggeratedly anomalous isotopic compositions, would merely perturb the measured composition of the real component, not generate a spurious one.

The major aliphatic component is enriched in $^{13}C$, relative to the aromatic components, by at least $6^{\circ}/oo$, and is also rich in D and $^{15}N$ ($+780 < \delta^D < +1800 ; \delta^{15}N > +40$). These values suggest that this aliphatic component may be related to the most polar extractable fractions in the meteorite. Such a relationship could imply either a common progenitor or isotopic exchange between the aliphatic component and a fluid phase on the meteorite parent body. The C data, in particular, implicate the planetesimal
environment in the evolution of the organic population. Thus, the inorganic carbonates, clearly themselves secondary, formed apparently from CO₂ like that trapped in Murchison. The same CO₂ probably constituted the major C source for the homologous series of monocarboxylic acids in Murchison [4], and thus plausibly for the bulk of the polar organics and, by extension, the aliphatic kerogen component. In contrast, the relatively high D-contents of the polar organics and the aliphatic component are difficult to reconcile with the apparently low D-content of the planetesimal aqueous phase, as recorded in the meteoritic hydrous silicates [2].

If isotopic exchange were indeed responsible for the present composition of the major aliphatic component, it is possible that it originally had the same isotopic composition as the major aromatic component. However, it does not seem possible for the latter to have been derived from the former by aromatisation. Similarly, the minor aromatic component cannot readily have been derived from any of the other identified components.

The isotopic data for C and N appear consistent with operation of purely solar-system processes, but the magnitude and range of observed D/H ratios still point towards fractionation at the low temperatures of interstellar clouds [5,6]. The mode of incorporation of such putative interstellar molecules into the kerogen-like material remains unclear, however. In particular, it is possible that measured D-contents could reflect the random distribution of some highly D-enriched entities, which may not even be chemically combined into the kerogen-like structure. Such a situation would seriously limit the usefulness of H-isotopic data in constraining theories for the origin of such structures. However, the fact that certain soluble fractions exhibit pronounced D-enrichments shows that at least some of the interstellar H was involved in organic synthesis in the early solar system. The enrichment of D in several components of differing chemical affinities indicates that the interstellar D is not carried solely by an acetylinic or cyanoacetylinic polymer as has been proposed [3].

REFERENCES