A NanoSIMS study of spatial relations between H and N isotopic anomalies in Orgueil and Murchison Insoluble Organic matter

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Abstract

Introduction: The major fraction of the organic carbon in Carbonaceous Chondrites (CC) is in the form of Insoluble Organic Matter (IOM). Although the formation of IOM still eludes our understanding, recent advances in structural, chemical and isotopic features of the IOM reveal some peculiarities. Among them, the isotopic heteoregeneity of the IOM in CC is striking: discrete H and N isotopic anomalies - the so-called *hotspots* - with δ values up to +19400‰ and +1770‰, respectively, are embedded in the bulk IOM, which has average compositions that are lower by a maximum of one order of magnitude [1]. This isotopic heterogeneity is often interpreted as a result of gas-phase interstellar-like processes occurring in the Solar Nebula or inherited from its parent molecular cloud [1]. Previous studies have observed that the D and ^{15}N *hotspots* can be either spatially correlated (i.e. at the same location in the IOM) or uncorrelated [1,2]. Here we present new Hydrogen and Nitrogen isotopic measurements with the NanoSIMS in order to shed light on these spatial correlations.

Methods : acid residues with a high content of IOM from Murchison and Orgueil [3] was pressed in pure gold foil. Two terrestrial kerogen standards (Type I and III) were analyzed in parallel. A 16 keV Cs$^+$ primary ion beam of 10pA rastered across a 20x20 μm^2 surface with an ion spot of 200 nm and a counting time of 1 ms/pixel. Three magnetic fields were used to measure successively: (i) H$^-$ and D$^-$, (ii) ^{12}C^{14}N$^-$ and ^{12}C^{15}N$^-$ and (iii) ^{13}C$^-$ and ^{13}CH$^-$. To improve the counting statistic, especially for Hydrogen isotopes, a typical measurement of IOM is composed of 100 such cycles. Under these conditions, the total acquisition time for the 100 cycles is about 7 hours. Each of these 100 cycles is systematically bracketed by 2 standard measurements to monitor the possible instrumental shifts.

Results: In Orgueil and Murchison IOM, about 57% and 80% of the *hotspots* in D and ^{15}N are spatially correlated, respectively. Hydrogen and Nitrogen isotopic compositions of Orgueil *hotspots* range from +2300‰ to +4000‰ and from +135‰ to +412‰ respectively.

For Murchison, H and N *hotspots* range from +2400‰ to +4500‰ and from -157‰ to +480‰, respectively. No correlation is observed between the magnitude of the *hotspot* anomalies, i.e. δD and δ^{15}N, in Orgueil and in Murchison. Based on H/C and N/C elemental ratios, the *hotspots* are, at a first approximation, indistinguishable from the bulk IOM chemical composition within the 25% relative error. Recent studies performed on IOM isolated from Orgueil show hydrogen isotopic variations at a molecular scale [3,4]. For instance, the Orgueil D-rich *hotspots* are a mixing between D-rich radicals (δD = +95,000‰) and the bulk IOM (δD = +1360‰) [4,5]. As for the D-rich radicals, it can be hypothesized that the spatially correlated D-rich and ^{15}N-rich *hotspots* are caused by ^{15}N-rich radical carriers. Such an assumption could be validated by EPR spectroscopy. As a corollary of this hypothesis, the occurrence of spatially uncorrelated *hotspots* suggests that some D-rich radicals do not have any Nitrogen in their chemical formula, therefore yielding a δ^{15}N equal to that the bulk IOM.

References

[1] Busemann H. et al. (2006) *Science*, 312, 727-730.

[2] Nakamura-Messenger K. et al. (2006), *Science*, 314, 1439-1442.

[3] Remusat, L. et al.(2006) *Earth and Planetary Science Letters*, 243, 15-25.

[4] Gourier D. et al. (2008) *Geochimica et Cosmochimica Acta*, 72, 1914-1923.

[5] Halbout J. et al. (1990) *Geochimica et Cosmochimica Acta*, 54, 1453-1462.