

H AND N ISOTOPES DISTRIBUTION IN INSOLUBLE ORGANIC MATTER IN ORDINARY CHONDRITES: CONSTRAINTS ON EARLY SOLAR SYSTEM PROCESSES. L. Remusat¹ and L. Piani²,

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Introduction: Insoluble organic matter (IOM) is known to be enriched in deuterium in carbonaceous chondrites (CC) and ordinary chondrites (OC) compared to terrestrial H reservoir [1]. The isotopic variations of IOMs in various types of chondrites are complex and have been related to parent body processes [1], if we assume a common precursor to the IOM in all the parent bodies. Interestingly, IOMs in OC and CC do not reveal the same behavior upon parent body evolution (metamorphism). While secondary processes tend to induce a D-depletion in CC, they result in large D-enrichments in OC, implying different mechanisms on the CC and the OC parent body [1]. Another possible explanation for this discrepancy would be that the parent body of OC accreted a small contribution of extremely D-enriched presolar organic matter [2].

It has been shown by various methods that δD of the IOM in CI and CM chondrites is heterogeneous at the molecular scale [e.g. 3] and the micron scale [e.g. 4]. The IOM of CR, CM and CI exhibit D- and ^{15}N -rich micron-sized anomalies, called hot spots [4]. In Orgueil IOM, the occurrence of D-rich hot spots has been shown to be related to the heterogeneous distribution of organic radicals characterized by $\delta D \approx 90,000$ [3]. A possible explanation is that the D-isotopic enrichment results from cold chemistry via ion-molecule like reactions - taking place in the outer areas of the protosolar nebula [3] or the interstellar space [5] - that have affected the IOM before accretion on the parent body.

In order to better constrain the origin of IOM in OC, we have investigated the distribution of heavy isotopes of H and N in the IOM of three different unequilibrated ordinary chondrites. We chose OC with different petrographic types (from 3.0 to 3.2) to track evolution due to parent body processes. Our goal was to compare the distribution of δD and $\delta^{15}N$ at the micron scale with similar observations for CC and to test the hypothesis that the IOM in OC may contain a contribution of D-rich interstellar OM.

Experimental: We have studied the IOM of 3 unequilibrated ordinary chondrites: Semarkona (LL3.0), Bishunpur (LL3.15) and GRO 95502 (L3.2). These IOM were isolated by classical HF/HCl protocol and pressed on clean indium foil, gold coated and imaged by the NanoSIMS 50 installed at MNHN. The IOM of Orgueil carbonaceous chondrite was also studied for

comparison. Secondary ion images of δD and $\delta^{15}N$ were generated using a 16 keV Cs^+ primary beam scanned over surface areas of $20 \times 20 \mu m^2$ with a raster speed of 2ms/pix and a resolution of 256×256 pixels. Before analysis, a high beam current (600 pA) was used for presputtering surface area of $25 \times 25 \mu m^2$ during 6 minutes. In a first step we acquired H^- and D^- images with a 8 pA primary beam (spatial resolution around 300 nm) and the mass resolving power of the mass spectrometer set to 4000. Then, the mass table was modified to collect $^{16}O^-$, $^{12}C_2^-$, $^{26}CN^-$, $^{27}CN^-$ and $^{32}S^-$ with a mass resolving power of 8000; we then lowered the primary current to 3 pA to increase the spatial resolution (150 nm). Data were processed using l'image software (Larry Nittler, Carnegie Institution in Washington DC).

Instrumental mass fractionation on isotopic and elemental ratios was calibrated using 4 samples: two terrestrial kerogens, Orgueil IOM and GRO 95502 IOM. We used the gas source mass spectrometry values as reference values for IOMs [1] and for terrestrial standards. Plotted against the reference values, the NanoSIMS measured values exhibit a linear correlation over several thousands of permil leading to a more precise correction than a unique standard procedure.

Results and Discussion: Compared to Orgueil, the IOM of Semarkona, Bishunpur and GRO 95502 (Figure 1) exhibit very few D-rich hot spots. Semarkona and Bishunpur IOMs are not homogeneous either. Large-scale heterogeneity occurs, with several microns large areas showing significantly large D-enrichment compared to the rest of the material having a δD slightly above Orgueil IOM. Frequency histograms from pixels values clearly indicate broader distribution in the case of Semarkona and Bishunpur, with a bimodal pattern, indicating at least two components. One component is centered on the Orgueil value. The other one has at least twice as much deuterium. In the case of GRO 95502 (more heated), the Orgueil-like component is missing. This large scale heterogeneity points to a multi-component fine, but incomplete, mixing.

$\delta^{15}N$ images also indicate a slightly broader pixel distribution in Semarkona and Bishunpur, but with only one component centered on the Orgueil distribution. Once again very few micron-sized hot spots can be distinguished. Three ^{15}N -rich hot spots were identified in the IOM of the studied OC: two in GRO 95502

(for 800 μm^2 sampled) and one in Semarkona (for 1200 μm^2 sampled). It must be noted that these ^{15}N -rich hot spots also constitute D-rich anomalies.

As Semarkona is one of the least modified chondrite since the accretion of its parent body, the lack of hot spots cannot be considered as a consequence of parent body evolution. If OC are free of abundant D-rich and ^{15}N -rich hot spots, they were likely absent at the location of the formation of the parent body.

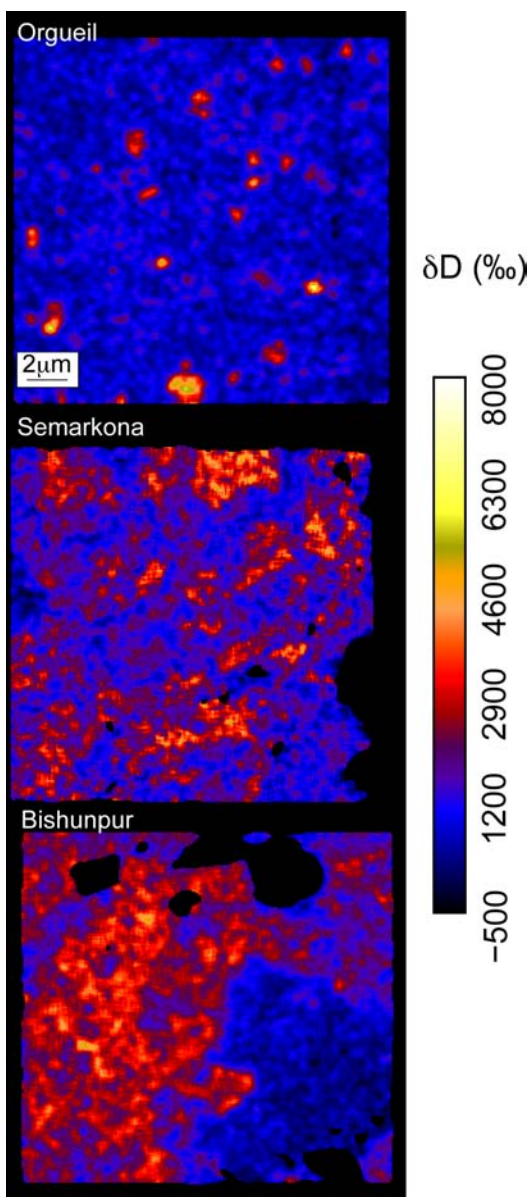


Figure 1: NanoSIMS images of δD of Semarkona and Bishunpur IOMs. A similar image of Orgueil IOM is displayed for comparison. All the images have the same size. The IOM of unequilibrated OC are heterogeneous, however, they do not exhibit numerous micron-scale anomalies as IOMs in type 1 and 2 CC like Orgueil.

Conclusions: δD and $\delta^{15}\text{N}$ in unequilibrated OC show a distinct pattern than in pristine CC. Whereas in type 1 and 2 CC, IOM exhibits numerous micron-sized D-rich and ^{15}N -rich anomalies (sometimes associated) mixed with a moderately enriched organic material, IOM in OC shows large scale heterogeneity, and very few micron-sized hot spots. Interestingly, the three hot spots we have identified are on the trend defined by IOMs in CC and corresponding to ion/molecule reactions in the protosolar nebula (figure 2).

Although we did not observe extreme D-enrichment for the hot spots in IOMs of OC, the different domains that can be defined in our images follow a mixing trend between CC-like IOM and interstellar organics (figure 2). We then conclude that our images are consistent with the hypothesis proposed by [2] that IOMs in OC are contaminated by minute amount of interstellar organics, but at a very fine (molecular?) scale. We conclude that a preaccretion mechanism rather than parent body processes should explain the differences between IOM in CC and in OC. This implies that the concept of the common organic precursor is not consistent with the total organic budget of OC.

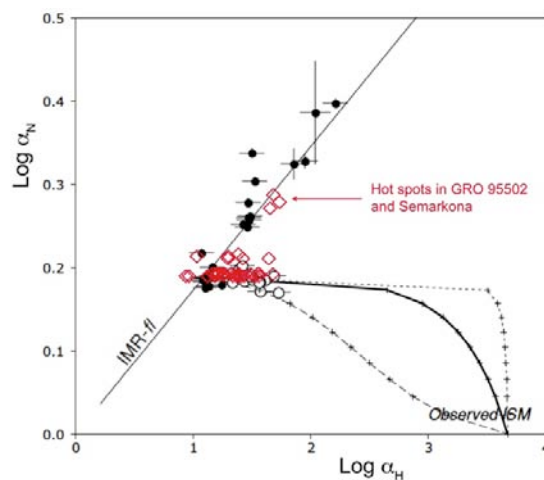


Figure 2: The N isotopic fractionation versus H isotopic fractionation between organics and the protosolar gas, from [2], modified. Black dots represent IOM in CC, Hale-Bopp HCN and hot spots in IDPs. In white dots are reported bulk IOM of OC. Our data are red open diamonds. It must be noted that three hot spots observed in Semarkona and GRO 95502 fit with the ion/molecule fractionation line (IMR-fl) of IOM in CC.

References: [1] Alexander C. M. O'D. et al. (2010) *GCA* 74, 4417-4437. [2] Aléon J. (2010) *ApJ* 722 1342-1351. [3] Remusat L. et al. (2009) *ApJ* 698, 2087-2092. [4] Busemann H. et al. (2006) *Science* 313 727-730. [5] Yang J. and Epstein S. (1983) *GCA* 47, 2199-2216.