CORRELATED DEUTRIUM AND NITROGEN ISOTOPIC ENRICHMENTS IN METEORITIC ORGANIC MATTER

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Introduction: It is well established that carbon-rich macromolecules survived the early solar system processes and were accreted in primitive carbonaceous chondrites [1−5]. These molecules often contain large isotopic enrichments in D, ^{15}N and/or occasionally in ^{13}C [1−3] hypothesized to have formed via ion-molecule reactions mediated on grain surfaces of ices coating dust grains and initiated by UV irradiation [6, 7]. To investigate the isotopic and chemical composition and origins of domains carrying correlated isotopic anomalies, i.e., hotspots anomalous in H, C and/or N, we carried out isotopic imaging of equivalent matrix regions on a series of meteorites of different petrologic types.

Samples and Methodology: We undertook NanoSIMS measurements of H, C and N isotopes in several carbonaceous chondrites: four CMs (Murray, Murchison, Cold Bokkeveld, and QUE 97990), three ungrouped chondrites (Bells, NWA 1152 and NWA 5958), and one CV (Allende). Results from C and N mapping for Murray, Murchison, Cold Bokkeveld, Bells, Allende, and QUE 97990 are reported in [8] and electron energy-loss spectroscopy of D-rich organics in QUE 97990 is described in [9]. All investigated samples were characterized, prior to NanoSIMS with a Cameca SX-100 electron microprobe at the University of Arizona. Areas of fine-grained interchondrule matrix and occasionally fine-grained rims around calcium aluminum inclusions and chondrules were searched for isotopic anomalies using the Cameca Ametek NanoSIMS 50L at Arizona State University using 2 different measurement setups. First, C and N ion imaging (setup 1) was performed with a 'chained analysis' setup. Prior to each analysis, a 20−25 pA primary beam was rastered over a 20×20 μm^2 surface area to remove the Au coating and surface contamination as well as to implant Cs^+ primary ions. The sample surface was then rastered with a 2−3 pA primary Cs^+ beam over areas of 15×15 μm^2 and the secondary isotopes of $^{12}C^-$, $^{13}C^-$, $^{12}C^{14}N^-$, $^{12}C^{15}N^-$, and $^{28}Si^-$ and secondary electrons were collected in multicollection mode. Subsequently, H ion imaging (setup 2) was done on the areas that had been scanned for C and N in the previous setup 1. The previously analyzed areas were briefly presputtered with a 20−25 pA beam, and then rastered ion images of $^1H^-$, $^2D^-$, $^{12}C^-$ and $^{18}O^-$ were collected in multicollection using a ~10pA Cs^+ beam. The analytical protocol for both setups included regular measurements on a cyanoacrylate standard. A domain was considered a hotspot if the isotopic anomaly was present in at least 3 consecutive planes and with isotopic compositions >5σ away from the terrestrial ratios.

Results: Murray, Murchison, Cold Bokkeveld, QUE 97990, Bells, NWA 1152, NWA 5958 and Allende were mapped for 5775 μm^2, 5400 μm^2, 5400 μm^2, 5625 μm^2, 2300 μm^2, 6525 μm^2, 6300 μm^2, and 5400 μm^2 respectively. All CM chondrites show presence of large H (234−4447‰) and N (-746−2544‰). The ungrouped carbonaceous chondrites NWA 1152 and NWA 5958 show no ^{13}C- or ^{15}N-rich hotspots but have some D-rich ones with compositions ranging from 244−1504‰. Murray and Bells have no C-anomalous domains, and Allende is devoid of any isotopic anomalies.

The abundance of hotspots with D excesses far exceed the number of ^{15}N-rich hotspots. In all but few occurences, the H, C and N anomalies are spatially uncorrelated. In comparison, two hotspots B1-7 and C4 in Murchison and one hotspot 3Aa9 in QUE 97990 have spatially correlated N and H isotopic anomalies. The degree of enrichments seen in D (δD~3417−4249‰) in these hotspots are high but ^{15}N ($\delta^{15}N$~289−378‰) enrichments are not large. In addition, two C-rich hotspots (average C/Si ratios ~59 indicating their carbonaceous nature) in Murchison B3 and 1A9 have correlated N and C isotopic anomalies, where the $\delta^{15}N$ of the N-anomalies are also low (77±14‰ and 324±16‰, respectively). The reaction pathways that can lead to correlated isotopic anomalies in simplest molecules can be investigated via this study, which will allow us to pin down their origins. A discussion of these results including ongoing H isotopic measuements on Cold Bokkeveld and Bells will be presented at the meeting.

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Acknowledgements: This research is funded by NASA grant (NNX15AD94G) for the program "Earths in Other Solar Systems".