NITROGEN AND CARBON ISOTOPIC VARIATIONS IN SEVERAL CARBONACEOUS CHONDRITES – A HUNT FOR THE CARRIER PHASES. M. Bose[1], T. J. Zega[2], and K. Domanik[2] [1]School of Molecular Sciences, Arizona State University, Physical Sciences Building PSD-13, Tempe AZ 85287-1604 | mbose2@asu.edu, [2]Lunar and Planetary Laboratory, University of Arizona, 1629 E. University Blvd, Tucson, AZ.

Introduction: Organic matter present in carbonaceous meteorites is most likely a mix of interstellar molecules, condensates that formed in the protosolar nebula, and those that transformed in the asteroid parent bodies. It has been established that both the soluble and insoluble organic matter (IOM) identified in meteorites and interplanetary dust particles contain material enriched in deuterium (D) and ^{15}N. Bulk ^{15}N enrichments in some carbonaceous meteorites [e.g., 1] are comparable to the values measured in comets and star-forming regions [e.g., 2]. However, meteoritic ^{15}N-rich 'hotspots' in the IOM [3–7] exhibit enrichments that varies dramatically over small spatial scales. Furthermore, meteorites belonging to the same class show enormous heterogeneity. Some of the ^{15}N enriched hotspots are associated with nanoglobules [8, 9] and may not show D enrichments [9]. They exceed the most extreme values for prestellar cores [10] or protosolar systems [11]. The origin of these hotspots, although unknown, is most likely material preserved, after being produced by low temperature chemistry in the interstellar medium or early solar system, and are therefore relics from before the solar system collapsed. This study aims to characterize the isotopic, chemical and structural characteristics of the starting inventories of organics and volatiles in meteoritic materials.

The diversity in the functional groups carrying the ^{15}N-enrichments is unknown, although some of these materials and the surrounding matrix material have been characterized [12, 13]. Hotspots with ^{15}N excess in Queen Alexandra Range 97416 and terrestrial C isotopic composition was found to contain nitrile functionality [6]. Another recent study [14] compared the average enrichments of the hotspots to the ^{15}N isotopic enrichments measured in ammonia from a large suite of meteorites, and argued that a section of these hotspots could be amines, which may have formed in the protosolar nebula. In order to assess statistically the ^{15}N enrichment routes for nitriles and amines, and identify additional N-bearing species, we collected C and N isotopic data of macromolecular material and presolar grains from Bells, Queen Alexandra Range 97990, Murray, Murchison, Cold Bokkeveld, and Allende. Additional chemical and structural data from these materials will be studied to learn more about what molecules that carry anomalous N and C isotopic compositions.

Samples and Methods: The meteorites studied are C-ung Bells, CM2.6 QUE 97990, CM2 Murray, CM2 Murchison, CM2 Cold Bokkeveld, and CV3 Allende. All investigated samples were characterized, prior to SIMS, by back-scatter electron (BSE) imaging and X-ray elemental mapping with a Cameca SX-100 electron microprobe at the University of Arizona.

Fine grained matrix areas around calcium aluminum inclusions and chondrules were searched for C and N isotopic anomalies with a 'chained analysis' setup, where the initial stage coordinates were predefined, and the stage moved in a predefined pattern. All of the isotopic analyses were performed on the Cameca Ametek NanoSIMS 50L at Arizona State University. The surfaces of the samples were Au-coated (20 nm thick) before analysis. 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 cesium. The sample surface was rastered with a primary Cs$^+$ beam over areas of 15×15 μm^2 divided into 256×256 pixels with a dwell time of 15 ms/pixel. 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 with a beam current of 2 to 3 pA. Secondary ions of ^{16}O$^-$, ^{31}P$^-$ and ^{32}S$^-$ were also occasionally collected. Several planes (5 to 10) were acquired for each area. The mass spectrometer was set to >8000 mass resolving power (Cameca definition). The analytical protocol included measurements on cyanoacrylate standard, followed by a chained analysis, and another measurement on the cyanoacrylate standard. All obtained images were processed with WinImage. Carbon and N isotopic ratios were normalized to those measured on the standard. Only regions that showed the isotopic anomaly for at least three consecutive planes and with isotopic compositions more than 5σ away from the terrestrial ratios were considered anomalous. Error bars reported in this study are 1σ errors based on poisson statistics.

Results: CM2 Murray, CM2 Murchison, CM2 Cold Bokkeveld, C-ung Bells, CV3 Allende, and CM2 QUE 97990 were scanned over areas measuring 5775 μm^2, 5400 μm^2, 5400 μm^2, 2300 μm^2, 6750 μm^2 and 4500 μm^2, respectively. The range in C and N isotopic compositions of these hotspots is shown in *Figure 1*. A total of 27 N-anomalous domains were identified by raster ion imaging of the six meteorites. No isotopic anomalies were found in Allende and two N-anomalous domains are most likely presolar grains.

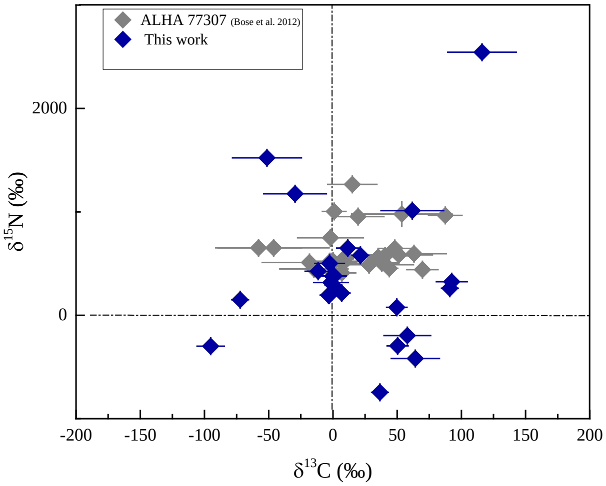

Figure 1: Carbon and nitrogen isotopic compositions of hotspots and coldspots in situ of five carbonaceous chondrites are shown in blue while the grey points are from published literature [6]. Errors are 1σ.

The remaining anomalous domains are carbonaceous grains with high C/Si ratios (C^-/Si^- ratios > 2).

The δ^{15}N values of the carbonaceous grains range from -746±61‰ to 2544±77‰ *(Figure 1)*. The five grains with ^{15}N depletions, which we refer to as 'coldspots', are from QUE 97990 and Cold Bokkeveld. The C isotopic composition of the carbonaceous grains range from -95±11‰ to 93±12‰ with seven of the grains showing a C anomaly. These seven C-anomalous grains are from Murchison, QUE 97990 and Cold Bokkeveld. The CN^-/C^- ratios for all the N-anomalous carbonaceous grains vary from 0.2 to 1.5 corresponding to 0.27 to 2.03 wt.% N, when compared to the cyanoacrylate standard. The range in C and N isotopic compositions of the hotspots are similar to those observed previously [e.g., 3, 4, 6], although the coldspots are quite abundant.

Cold Bokkeveld shows one coldspot (δ^{15}N = -746±61‰), which is anomalous in carbon as well (δ^{13}C = 37±5‰). Murray, in general shows the smallest range of anomalies, while Bells hotspots show the largest range in the ^{15}N enrichments. Two of the hotspots in Murray contain small amounts of S.

Presolar grains. Two of the anomalous grains with very large ^{15}N excesses (δ^{15}N = 4505±35‰ and 15364±48‰) correlate with Si in the ion image, suggesting that they are SiC grains. Both the SiC grains are either mainstream SiC or X-grain. QUE 97990 was also searched for C- and O-anomalous presolar grains. None were identified in the 3350 μm^2 area.

Discussion and Conclusions: Very large ^{15}N-enrichments were observed in five meteorites predominantly belonging to the CM-class of carbonaceous chondrites. Seven grains with C anomalies were also located in the same samples. It is not yet clear whether the absence of presolar grains in the relatively more pristine CM2.6 QUE 97990 compared to other CMs studied is due to aqueous alteration or that they are heterogeneously distributed and require measurement of additional areas. Oxidative metamorphism in Allende definitely led to the destruction of the organic component of this meteorite.

Our data suggest that the ^{15}N enrichments are present in CM Type 2 chondrites, irrespective of the degree of parent-body aqueous alteration. Meteorites with high degree of brecciation, like Bells, have the largest ^{15}N anomalies. ^{15}N isotopic enhancements in primitive solar system material has been explained by either non-equilibrium ion-molecule chemistry in cold, dense molecular cloud material in the interstellar medium or via N_2 self-shielding processes in the outer solar system [16, 17].

Based on the large range in N and C isotopic compositions, parent-body processing does not seem to have completely overprinted the original isotopic signatures indicative of cold chemistry. Additional measurements, including the determination of functional carriers, which are planned, are necessary in order to understand how these may have been formed and were delivered to the asteroid parent bodies.

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