

NITROGEN ISOTOPIC IMAGING OF TAGISH LAKE CARBON GLOBULES. S. Messenger¹, K. Nakamura¹, L.R. Nittler², and A. Young², ¹Johnson Space Center, Code SR, 2101 NASA Parkway, Houston TX, 77058, scott.r.messenger@nasa.gov, ²Department of Terrestrial Magnetism, Carnegie Institution of Washington, 5241 Broad Branch Road NW, Washington DC 20015

Introduction: Chondritic meteorites and interplanetary dust particles commonly exhibit significant enrichments in deuterium (D) and ¹⁵N relative to terrestrial materials. These anomalies are usually ascribed to low temperature chemical processes occurring in a presolar cold molecular cloud and/or physically similar regions of the outer solar nebula [1]. The sizes of these anomalies vary considerably among different classes of meteorites, in some cases on a microscopic scale, reflecting differing degrees of alteration and dilution of the presolar components owing to parent body processing.

Tagish Lake (CI2) is a unique meteorite that has undergone a lower degree of aqueous processing in comparison with other CI chondrites. Reflectance spectroscopy suggests that this meteorite is a rare sample of outer belt (class D) asteroids [2]. Tagish Lake (TL) possesses abundant presolar SiC and nanodiamonds [3] together with H and N isotopic anomalies that fall within the range observed among other primitive chondrites [3,4,5].

Tagish Lake has abundant organic C (~2.5 wt %, [3]) that is distinct in some respects from other carbonaceous chondrites. The organic matter in TL consists predominantly of acid insoluble, PAH-rich material, and has a surprisingly low abundance of amino acids [6]. In addition, numerous submicrometer organic globules are found distributed throughout the matrix [7]. These globules are remarkably similar to those artificially produced by photolysis of organic ices thought to coat interstellar grains [8]. While H isotopic measurements would provide a strong diagnostic test of this hypothesis, their small size (100 - 500 nm) and the naturally low abundance of D currently precludes D/H isotopic measurements on individual globules. Here we report the first C and N isotopic imaging study of samples rich in organic globules from TL.

Methods: Two types of samples were analyzed in this study. One set consisted of carbon globule-rich fragment from a 'pristine' fragment of the carbonate-poor lithology. These samples were embedded in elemental S and sliced with a diamond ultramicrotome, avoiding any possibility of contamination with organic epoxy. TEM observations of adjacent sections were performed in order to identify carbon globule-rich regions that were later targeted for N isotopic mapping. The second set of samples consisted of 5 - 20 micrometer sized fragments of matrix material

(from the carbonate-rich lithology) pressed into Au foil. These fragments were first subjected to D/H isotopic imaging with the Carnegie Institution IMS-6f ion microprobe (see companion abstract, [9]). Two particles exhibiting substantial localized D enrichments ("hotspots") were measured for their C and N isotopic compositions.

The C and N isotopic imaging studies were performed with the Washington University Cameca NanoSIMS ion microprobe. We measured C and N isotopes: ¹²C⁻, ¹³C⁻, ¹²C¹⁴N⁻, ¹²C¹⁵N⁻ and ²⁸Si⁻ simultaneously operating in multicollection mode. A 16 keV, 2 pA Cs⁺ primary ion beam was used, which had a nominal beam diameter of 100 nm. We acquired 10 - 15 sequential image layers for each sample, consuming ~100 nm of material over a period of 4 - 8 hours. The peaks were centered prior to each imaging run, with the B-field stabilized by an NMR probe to within ~1 ppm over the period of the measurement. Nearby (<50 micrometers) terrestrial 1-hydroxybenzotriazole hydrate grains were analyzed for isotopic standards, following the same imaging procedures. Since sample topography could affect the measured N isotopic ratios, we determined N isotopic ratios of submicrometer regions of the 1-hydroxy standard grains. We found sample topography of these standard grains introduced less than 40 % (1σ) spread on the measured N isotopic ratios. The sample mounts were coated with ~80 Å of Au in order to mitigate sample charging.

Results: Two pristine TL areas imaged gave essentially uniform C distributions and bulk N isotopic compositions near terrestrial, with localized δ¹⁵N enrichments in these images < +100 %. In contrast, an area targeted by TEM studies of adjacent sections had numerous submicrometer C hotspots in the secondary ion images (Fig. 1). The measured δ¹⁵N values for 24 individual C-rich hotspots ranging in size from 200 - 500 nm are shown in Fig. 2. Most of these C grains exhibited significant ¹⁵N enrichments, ranging up to a maximum of +670 %.

Both of the D-rich TL samples were also found to be ¹⁵N-rich, with bulk δ¹⁵N values of ~90 % - similar to that of the organic globule rich section described above. N isotopic images of one of these samples showed several submicrometer ¹⁵N hotspots, reaching δ¹⁵N values of +300 - 400 %. The positions of these ¹⁵N-rich hotspots are compared with the previously

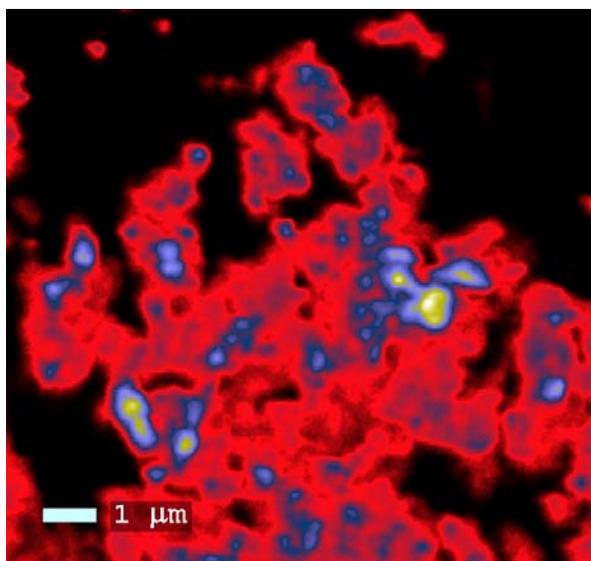


Fig 1: NanoSIMS C⁻ ion image of a region in Tagish Lake rich in submicrometer carbonaceous 'grains.'

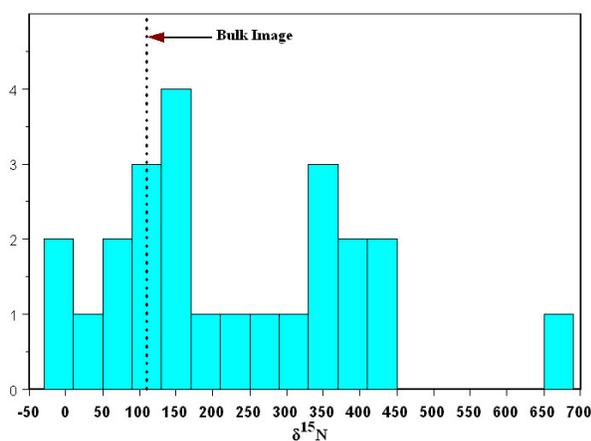


Fig 2: Histogram of δ¹⁵N values for 24 'C globules' identified in Fig. 1. The bulk δ¹⁵N value for the image including the globules is shown for comparison.

acquired D/H ratio map in Fig. 3. The D- and ¹⁵N-rich phases in this sample are clearly separated from each other in this image.

Discussion: In this initial study, we have found abundant submicrometer carbonaceous grains in pristine, carbonate-poor Tagish Lake to be significantly more ¹⁵N-rich than the surrounding matrix. While the isotopic anomalies are not so extreme as to require an exclusively interstellar origin, they do suggest a primitive origin. It is likely that these grains are partially comprised of nebular or presolar organic compounds which formed at very low temperatures. N isotopic anomalies of this magnitude

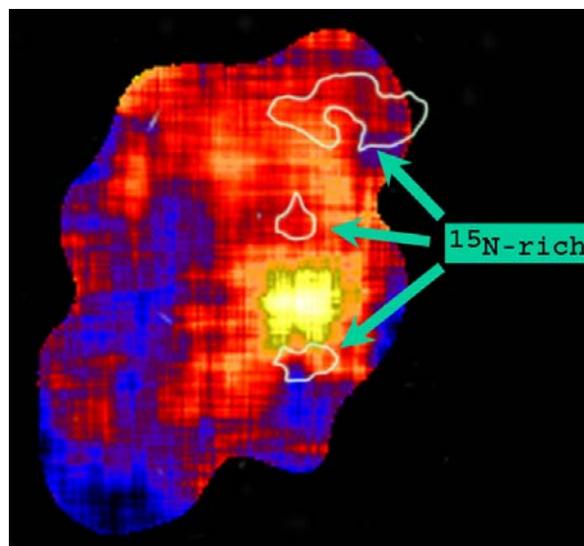


Fig. 3: D/H ratio image of Tagish Lake matrix with areas subsequently found to be ¹⁵N-rich indicated by the outlines.

are commonly observed in organic compounds from interplanetary dust particles and chondrites, although their origins are still uncertain [1,10,11]. The ¹⁵N/¹⁴N ratios of the grains appear to form a continuous distribution from normal to moderately enriched (670 ‰) implying varying degrees of dilution or exchange during or subsequent to the formation of the carbonaceous grains. More extensive studies may identify isotopically distinct sub-populations of organic globules.

While the D-rich TL sample was found to contain ¹⁵N-rich material, these two anomalous phases are not spatially associated. However, it may be possible for both H and N fractionation to occur in similar (low temperature chemical) processes contemporaneously, while residing in distinct carrier phases [11].

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