NANOSIMS STUDIES OF PRESOLAR GRAPHITE GRAINS: ARE C-ISOTOPIC RATIOS GRAIN-SIZE-DEPENDENT? P. Hoppe[1], J. Huth[1], and U. Ott[1], [1]Max-Planck-Institute for Chemistry, Particle Chemistry Dep., 55020 Mainz, Germany (hoppe@mpch-mainz.mpg.de).

Introduction: After silicon carbide (SiC), graphite is the best studied presolar mineral. A wealth of information exists on the isotopic compositions of C and of many minor/trace elements in micrometer-sized grains [e.g., 1-5]. Presolar graphite grains exhibit large C-isotopic anomalies with $^{12}C/^{13}C$ ratios between 2 and 7000. Although this range is compatible with what is observed for SiC [6, 7], the distribution of $^{12}C/^{13}C$ ratios is completely different. Most presolar graphite grains have isotopically light C (i.e., $^{12}C/^{13}C > 89$) while most SiC grains have heavy C with $^{12}C/^{13}C$ ratios between 40 and 80, indicative of different relative proportions of the stellar sources that contributed carbonaceous dust to the solar nebula. Many low-density ($\rho < 2.05$ g/cm^3) graphite grains show enrichments in ^{15}N, ^{18}O, and ^{28}Si as well as evidence for the former presence of radioactive ^{44}Ti and ^{49}V. This points to an origin in Type II supernovae (SNII) of a large fraction (several 10 %) of presolar graphite grains [3]. An important source of high-density graphite grains with isotopically light C are probably low-metallicity AGB stars [4]. This contrasts with what was inferred for the stellar sources of presolar SiC, namely, solar metallicity AGB stars for the vast majority of the grains [8] and SNII for about 1% of the grains [7,9].

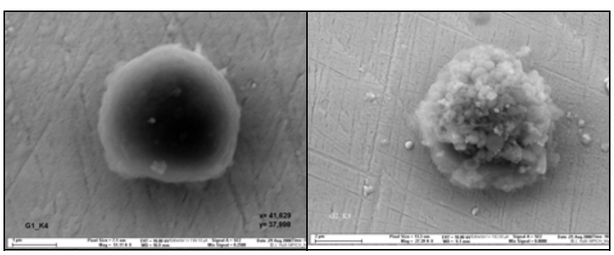

Figure 1. SEM pictures of graphite grains from Murchison separates G1 and G2. Left: Grain G1-4 (compact). Right: Grain G2-4 (composed of submicrometer-sized grains). Scale bars are 1 µm.

Since previous studies of presolar SiC had indicated grain-size dependencies of isotopic compositions [e.g., 10], a comprehensive characterization of presolar graphite grains from a broad range of grain sizes is important to get a complete picture of presolar graphite formation. Here, we report on isotope measurements of C, N, O, and Si in individual submicrometer-sized and µm-sized graphite grains separated from the Murchison CM2 meteorite. Important questions are: (i) Are there systematic differences in C-isotopic composition between submicrometer-sized and µm-sized grains? (ii) If yes, what can be inferred for the relative contributions of the different stellar sources to the population of the submicrometer-sized graphite grains?

Experimental: Presolar diamond, graphite and SiC were separated from a 91 g sample of the Murchison CM2 meteorite using procedures similar to those of [11] for their KJ series. The density separation resulted in six graphite separates: G1 (nominal density: 2.2-2.3 g/cm^3), G2 (2.1-2.2 g/cm^3), G3 (2.0-2.1 g/cm^3), G4 (1.7-2.0 g/cm^3), G5 (1.5-1.7 g/cm^3), and G6 (<1.5 g/cm^3). Grains from all separates were put on Au-coated SEM stubs. Subsequently, 49 µm-sized grains (1-19 µm; 14 from G1, 20 from G2, 9 from G3, and 6 from G4) as well as several piles of submicrometer-sized grains were selected by SEM/EDX for isotope studies. The µm-sized grains are either compact or consist of smaller subunits (Fig. 1). Samples of all types were studied for C- and N-isotopic compositions with the NanoSIMS at MPI for Chemistry. Ion images of $^{12}C^-$, $^{13}C^-$, $^{12}C^{14}N^-$, $^{12}C^{15}N^-$, and $^{28}Si^-$ were acquired by rastering a focused Cs^+ ion beam (~1 pA, 100 nm) over the grains. Four identified micrometer-sized presolar graphite grains were subsequently measured for O- and Si-isotopic compositions by recording secondary ion images of $^{16}O^-$, $^{18}O^-$, $^{28}Si^-$, $^{29}Si^-$, and $^{30}Si^-$.

Results and Discussion: We identified 6 large (1-4 µm) presolar graphite grains with $^{12}C/^{13}C$ ratios between 2.4 and 500 (Fig. 2). Four grains have isotopically light C, representing 67±33 % of the presolar graphite grains. Although statistics are limited, this is compatible with what has been observed before for micrometer-sized grains (ca. 65 %, Fig. 3). The $^{12}C/^{13}C$ ratio of the grain with the largest ^{13}C enrichment is among the lowest ratios reported so far. This grain has isotopically light N while the remaing five presolar grains have isotopically heavy N (Fig. 2). These isotopic signatures are compatible with what has been reported previously for micrometer-sized presolar graphite grains [1-5]. None of the grains shows O- and/or Si-isotopic anomalies of more than 3σ. The remaining micrometer-sized grains have close to solar C-isotopic composition ($\delta^{13}C$ = -60 to 70 ‰) with most of them being enriched in ^{15}N by typically 100-250 ‰. Graphite grains with these isotopic signatures have been observed before [1,2]. It has been suggested that these formed in the molecular cloud from which our Solar System formed, since moderate ^{15}N enrichments can be produced by ion-molecule reactions at low temperatures in molecular clouds [12].

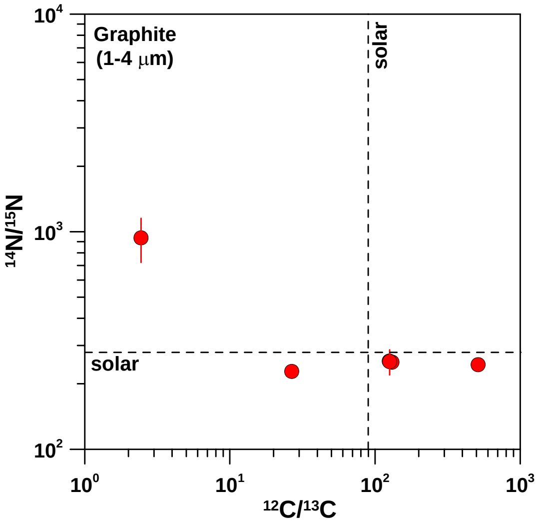

Figure 2. C- and N-isotopic compositions of six µm-sized presolar graphite grains from Murchison separates G1 and G2. Three data points overlap at $^{12}C/^{13}C$ ~130. The solar ratios are indicated by the dashed lines.

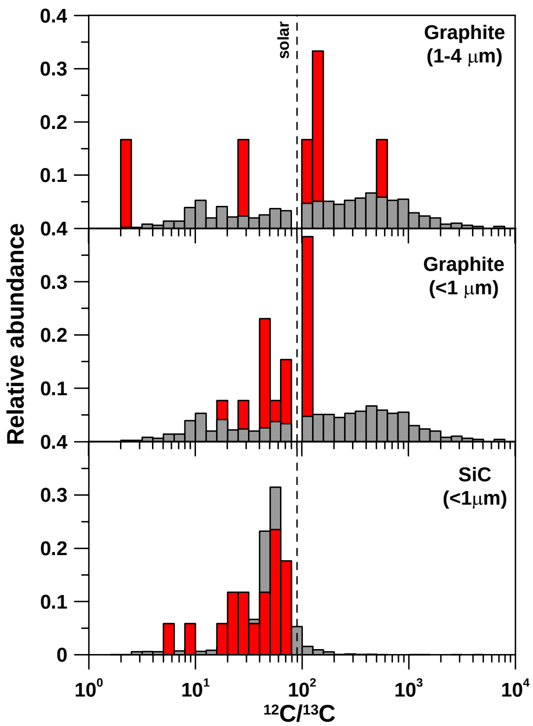

Figure 3. Histograms of $^{12}C/^{13}C$ ratios of µm-sized (top) and submicrometer-sized (middle) presolar graphite grains, and of presolar SiC (bottom). Data from this study are shown in red, those from previous studies for µm-sized graphite grains ([1,2]; the abundant group 3 grains with normal $^{12}C/^{13}C$ are not shown for clarity) and for SiC [6,7,10] in grey.

The C-isotopic ratio images of several grains in separates G1-G3 revealed the presence of submicrometer-sized objects (median size: 400 nm) with anomalous isotopic composition (Fig. 4). A total of 30 grains with anomalous C was identified. Seventeen of those grains are SiC and 13 are presolar carbon (graphite) grains. The C-isotopic ratios of these grains are plotted in Fig. 3 (middle and lower panels). The distribution of $^{12}C/^{13}C$ ratios of presolar SiC grains is similar to what has been observed for SiC before (mostly isotopically heavy C). Most of the submicrometer-sized presolar graphite grains, however, have isotopically heavy C as well, contrary to the large grains. The grains with isotopically light C constitute only 38±17 %. Statistics are limited, but this might be a hint that submicrometer-sized graphite grains have predominantly heavy C, similar to presolar SiC. It was argued that many of the micrometer-sized graphite grains with isotopically light C formed in the winds of low-metallicity AGB stars [4]. There, C/O ratios can become very large [13], which may be favourable for the growth of large grains. AGB stars with solar metallicity are not expected to develop very large C/O ratios [13] and these stars may preferentially form small (i.e. submicrometer-sized) graphite grains in addition to SiC grains.

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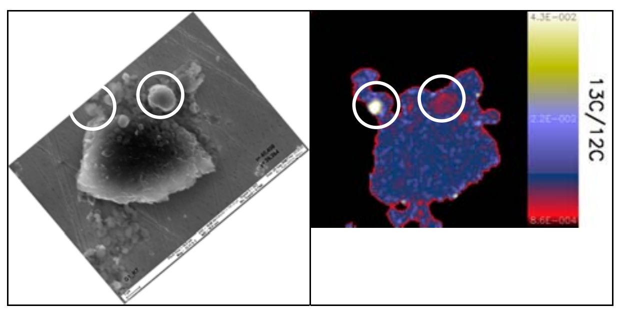

Figure 4. SEM picture (left) and corresponding NanoSIMS $^{13}C/^{12}C$ ratio image (right) of graphite grain G1-7. Two presolar grains (one depleted in ^{13}C, one strongly enriched in ^{13}C) are indicated by the circles. Scale bar in the left image is 1 µm. Field of view in the right image is 15 x 15 µm^2.

References: [1] Zinner E. et al. (1995) *Meteoritics*, 30, 209. [2] Hoppe P. et al. (1995) *GCA*, 59, 4029. [3] Travaglio C. et al. (1999) *ApJ*, 510, 325. [4] Jadhav M. et al. (2006) *New Astr. Rev.*, 50, 591. [5] Jadhav M. et al. (2008) *ApJ*, 682, 1479. [6] Hoppe P. et al. (1994) *ApJ*, 430, 870. [7] Nittler L. R. & Alexander C. M. O'D. (2003) *GCA*, 67, 4961. [8] Lugaro M. et al. (2003) *ApJ*, 593, 486. [9] Hoppe P. et al. (2000) *MAPS*, 35, 1157. [10] Hoppe P. et al. (1996) *GCA*, 60, 883. [11] Amari S. et al. (1994) *GCA*, 58, 459. [12] Charnley S. B. & Rodgers S. D. (2002) *ApJ*, 569, L133. [13] Zinner E. et al. (2006) *ApJ*, 650, 350.