FINALLY: PRESOLAR GRAPHITE GRAINS IDENTIFIED IN ORGUEIL. M. Jadhav[1], T. Maruoka[2]*, S. Amari[2] and E. Zinner[2], [1]Laboratory for Space Sciences and the Department of Earth and Planetary Sciences (mjadhav@wustl.edu), [2]Laboratory for Space Sciences and the Physics Department, Washington University in St. Louis, One Brookings Dr., St. Louis, MO 63130, USA., *present address: Department of Geosciences, Osaka City University, Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan.

Introduction: Presolar graphite grains have been extensively studied in the Murchison meteorite [1-4]. Almost everything we know about presolar graphite is based on these studies. This is because graphite is present mainly in very primitive meteorites [5] and the separation procedure of graphite is far more complicated than that of SiC [6]. Presolar graphite is known to be the carrier of Ne-E(L) [1] and according to noble gas analyses, its abundance in Orgueil is estimated to be an order of magnitude higher than that in Murchison [5]. A previous effort has been made to isolate this abundant presolar graphite in Orgueil: with the separation procedure previously applied to Murchison, Pravdivtseva et al. [7] obtained a fraction with a density of ~1.8 g cm^{-3} and grain size > 1μm. Observations in a scanning electon microscope (SEM) showed that grains from this Orgueil fraction closely resemble those of Murchison graphite, exhibiting the same onion-type morphology [4]. However, NanoSIMS isotopic analyses of C and N of 162 grains yielded only normal ratios, indicating that the grains had a solar system origin. In addition, Ne isotopic analysis of 14 individual grains, by high-transmission ion-counting noble gas mass spectrometry [8], did not detect any excesses in ^{22}Ne above the blank.

We undertook a new separation of carbonaceous and refractory presolar grains from Orgueil. The main objective was to isolate presolar graphite, which inspite of its inferred high abundance in this meteorite has been elusive until now. Here we report isotopic analyses of graphite grains from three density fractions from Orgueil. Grains of two fractions exhibit isotopic anomalies, demonstrating their presolar origin.

Experimental Methods: Our Orgueil sample was kindly provided by the National Museum of Natural History in Paris. We started with 24.16 g of the meteorite and treated it with a mixture of toluene and methanol (4 times). This removed the soluble organic matter that constituted 2.35% of the original sample. Alternating treatments with 10M HF-1M HCl and 6M HCl (14 times) dissolved the silicates. The reactive kerogens were then removed by oxidizing the remaining sample with 0.5N $Na_2Cr_2O_7$-2N H_2SO_4 at 75°C for 20 hours. After nanodiamonds were extracted by colloidal separation, the residue was further separated according to density by using sodium polytungstate [$Na_6(H_2W_{12}O_{40})$]. Ten final density fractions were obtained. The lightest density fraction, ORG1a (<1.59 g cm^{-3}) contains organic matter and the heaviest fraction, ORG1j (>2.3 g cm^{-3}) is expected to contain SiC and oxides. Any presolar graphite grains should be present in at least a few of the remaining 8 density fractions. The 8 fractions were then separated according to size with a cutoff of 1 μm.

Carbonaceous grains were identified by conducting X-ray analysis with a JEOL-840A SEM. We investigated the fractions ORG1b (1.59 – 1.67 g cm^{-3}), ORG1d (1.75 – 1.92 g cm^{-3}) and ORG1g (2.04 – 2.12 g cm^{-3}).

This initial characterization was followed by isotopic analyses of the grains in the NanoSIMS. C and O isotopic ratios were measured in multidetection mode by counting $^{12}C^-$, $^{13}C^-$, $^{16}O^-$ and $^{18}O^-$ secondary ions produced by the bombardment with a Cs^+ primary beam. For the ORG1d and ORG1g fractions, $^{12}C^{14}N^-$ was also measured.

Results: We identified 22 carbonaceous grains in ORG1b, 33 in ORG1d and 53 in ORG1g with the SEM. Most of the large grains are spherules and are very similar to Murchison presolar graphite grains; they have smooth surfaces and a platy, onion-like morphology [4]. It was difficult to determine the surface morphologies of the smaller grains (< 3 μm) due to the large concentration (much higher than in graphite separates from Murchison) of other carbonaceous material in which these grains were often found embedded. Additionally, we found at least 2-3 potato-shaped carbonaceous grains in each density fraction.

The results of the isotopic analyses of the 3 fractions are plotted in Figure 1. The C and O isotopic ratios of ORG1b grains are close to solar. They do not show the large variations that are expected in presolar graphite [4, 9]. However, 20 out of 33 grains in ORG1d, and 39 out of 53 grains in ORG1g have extremely large C isotopic anomalies. The $^{12}C/^{13}C$ ratios of the former range from 6 to 859, and those of the latter from 5 to 811 (Figures 1 and 2). Grains with higher than solar $^{12}C/^{13}C$ ratios are more abundant in the higher-density fraction ORG1g. A minor population of grains with $^{12}C/^{13}C \sim 10$ is observed in the two fractions. Similar C isotopic distributions, a higher abundance of grains with $^{12}C/^{13}C$ > solar in grains with higher density, and the presence of a population around $^{12}C/^{13}C \sim 10$, have also been observed in Murchison graphite [4].

Nine out of 33 grains in ORG1d exhibit ^{18}O excesses of up to 29 times the solar value, while all 53 grains in ORG1d have $^{16}O/^{18}O$ ratios close to solar (Figure 1). Since ^{18}O excesses are a signature of a supernova (SN) origin [9], they indicate that ORG1d contains SN graphite. In the KE3 fraction (1.65 – 1.72 g cm^{-3}) from Murchison about a third of the grains are SN graphite. The abundance of ^{18}O-rich grains decreases with increasing density and, in fact, such grains have not been observed in the highest Murchison density fraction KFC1 (2.15 – 2.20 g cm^{-3}). Thus, the density fractions of Orgueil and Murchison show similar trends in the abundances of SN graphite.

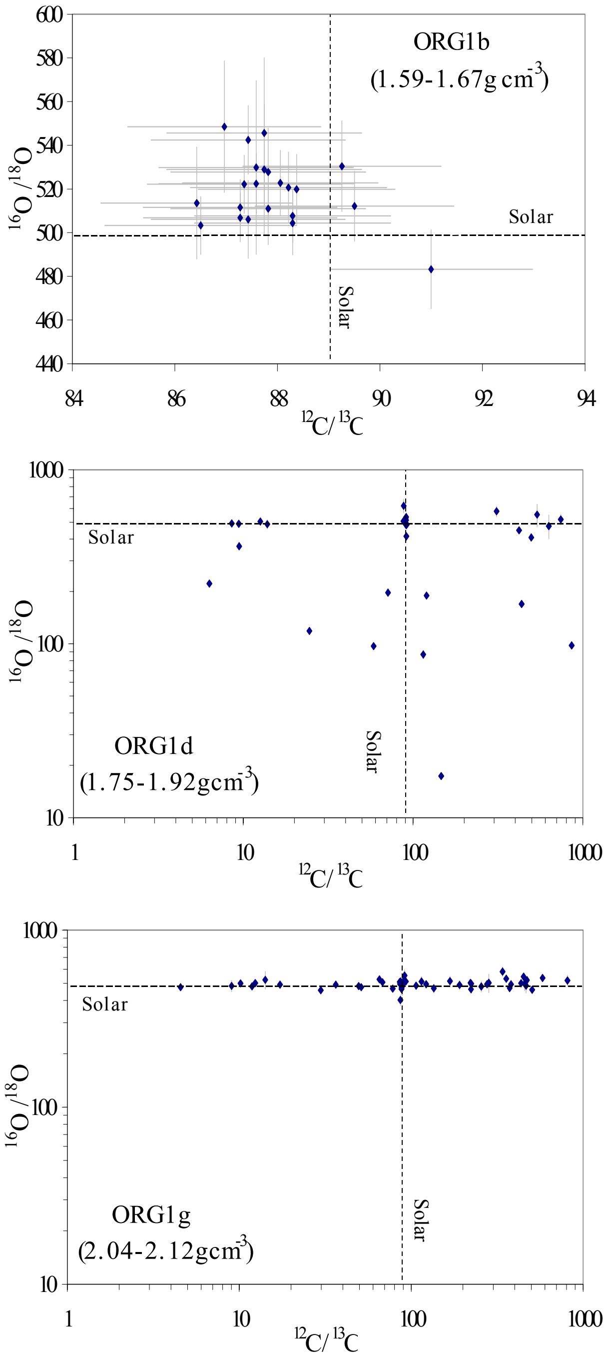

Figure 1: C and O isotopic compositions of individual carbon grains from three density fractions of Orgueil. Dashed lines indicate solar isotopic ratios: $(^{12}C/^{13}C)_{solar}$ = 89 and $(^{16}O/^{18}O)_{solar}$ = 499.

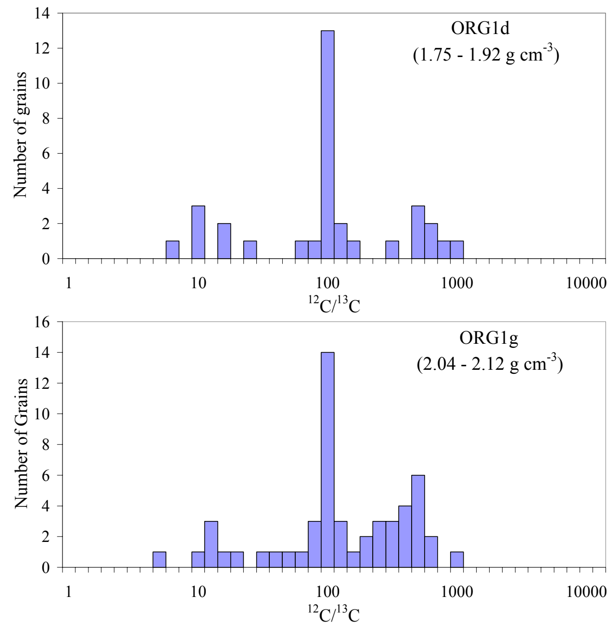

Figure 2: Distributions of $^{12}C/^{13}C$ ratios in the graphite grains.

Conclusions: Presolar graphite has been successfully isolated from Orgueil: two density fractions, ORG1d (1.75 – 1.92 g cm^{-3}) and ORG1g (2.04 – 2.12 g cm^{-3}), contain presolar graphite as indicated by isotopically anomalous carbon. Isotopic characteristics of Orgueil graphite are similar to those of Murchison graphite.

In the future, we plan to extend our analyses to the remaining five Orgueil fractions to determine the distribution of presolar graphite in this meteorite. Furthermore, Raman spectroscopy and TEM studies of these grains will reveal whether these grains are well-crystallized graphite (like the onion-type grains in Murchison [3]) or poorly graphitized carbon (cauliflower grains in Murchison [3]). Additionally, extensive isotopic analyses need to be carried out on these grains. We hope to be able to measure N, Ne, Mg-Al, Si, Ca, and Ti isotopic ratios in order to completely characterize these grains and be able to obtain information about their stellar sources.

References: [1] Amari S. et al. (1990) *Nature*, 345, 238-240. [2] Amari S. et al. (1995) *GCA* 59, 1411-1426. [3] Zinner E. et al. (1995) *Meteoritics* 30, 209-226. [4] Hoppe et al. (1995) *GCA* 59, 4029-4056. [5] Huss G. R. and Lewis R. S. (1995) *GCA* 59, 115-160. [6] Amari S. et al. (1994) *GCA* 58, 459-470. [7] Pravdivtseva O. et al. (1994) *LPS XXXV,* Abstract # 2096. [8] Hohenberg C. M. (1980) *Rev. Sci. Instr.* 51, 1075-1082. [9] Travaglio C. et al. (1999) *ApJ.* 510, 325-354.