CONTINUED ISOTOPIC STUDIES OF LOW-DENSITY GRAPHITE GRAINS FROM ORGUEIL. M. Jadhav[1], S. Amari[2], E. Zinner[2] and T. Maruoka[2]*, [1]Laboratory for Space Sciences and the Department of Earth and Planetary Sciences (manavijadhav@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: Graduate School of Life and Environmental Sciences, University of Tsukuba, Ibaraki 305-8572, Japan.

Introduction: We have reported isotopic analyses of low-density graphite grains from Orgueil at past meetings [1-3] but were unable to carry out further analyses on the same grains due to laboratory accidents (e.g., [2]) or contaminated grain mounts (e.g., [1, 3]). Here we present C, N, O, Si, Al-Mg, K and Ca isotopic analyses on new grains from the ORG1d (ρ ~ 1.75-1.92 g cm^{-3}) density fraction of Orgueil. This work is a continued effort to obtain correlated, multi-element isotopic data [4] on presolar graphite grains in order to constrain their stellar sources. Isotopic signatures in low-density graphites from Murchison [5] and Orgueil indicate that most of the grains have a Type II supernova (SN) origin while some originate from low metallicity asymptotic giant branch (AGB) stars [1-3]. However, both Murchison and Orgueil density separates have a population of grains with low $^{12}C/^{13}C$ ratios whose sources still remain enigmatic. A recent C, Ca and Ti isotopic study of this population ($^{12}C/^{13}C < 20$) in high-density Orgueil graphites indicates that these grains have multiple stellar sources: SNe and born-again AGB stars [6, 7]. This present study is also an attempt to understand grains with low $^{12}C/^{13}C$ ratios from the low-density fraction.

Experimental Methods: Twenty-three spherical, carbonaceous grains identified by EDX analysis in the SEM were picked with a micromanipulator and deposited on a gold-foil. This is now the preferred method to prepare a sample mount of graphite grains from Orgueil because it helps reduce contamination from the macromolecular carbonaceous material that these grains are often found embedded in. The mount was then coated with ~ 10 nm of gold to prevent the grains from falling off.

The isotopic analyses of these grains were carried out with the NanoSIMS at Washington University. In two independent runs, negative secondary ions of ^{12}C, ^{13}C, ^{28}Si, ^{29}Si, and ^{30}Si, and of ^{16}O, ^{18}O, $^{12}C^{14}N$, $^{12}C^{15}N$, and ^{28}Si were collected simultaneously by bombarding the sample with a Cs^+ primary beam. Positive secondary ions of ^{12}C, ^{24}Mg, ^{25}Mg, ^{26}Mg, and ^{27}Al, were produced by an O^- primary beam in order to measure the isotopic ratios in the Al-Mg system. Potassium and Ca isotopes were measured with the O^- beam in a combination of peak-jumping and multi-detection modes. The positive secondary ions of ^{39}K, ^{41}K, and ^{43}Ca, and ^{12}C, ^{40}Ca, ^{42}Ca, ^{44}Ca, and ^{48}Ti were measured at two different magnetic fields to obtain K and Ca isotopic ratios.

Results: *C, N, O, and Si isotopes*: The grains in this study exhibit a large range of C isotopic ratios ($^{12}C/^{13}C$ ~ 9 – 2280; Figures 1 and 2). Most grains have solar or sub-solar C ratios. Six grains are ^{13}C-enriched and have $^{12}C/^{13}C < 30$. Twelve of the twenty-three grains have ^{18}O excesses, ranging up to a $^{18}O/^{16}O$ ratio of ~ 17 times the solar value (Figure 2). Nitrogen-15 excesses were found in six grains (Figure 1), all of which also exhibit ^{18}O excesses. We also found twelve grains with large ^{28}Si excesses (Figure 5): ten of which have also ^{18}O excesses and six have ^{15}N excesses. These signatures in N, O and Si isotopic ratios are consistent with a Type II SN origin for these grains [5, 8].

Al-Mg isotopes: Twelve grains exhibit extremely large ^{26}Mg excesses from the decay of short-lived ^{26}Al ($t_{1/2} = 7.2\times10^5$ a). The inferred $^{26}Al/^{27}Al$ ratios for these grains range from 0.01 to 0.6 (Figure 3). Such high $^{26}Al/^{27}Al$ ratios have not been observed in any presolar graphite grain but are found in SiC-X grains [9]. The largest $^{26}Al/^{27}Al$ ratio previously inferred in Murchison KE3 graphites is ~ 0.146 and Orgueil graphites (before this study) is ~ 0.33. All the grains with high $^{26}Al/^{27}Al$ ratios have ^{18}O excesses and ten have ^{28}Si excesses. All six grains with low $^{14}N/^{15}N$ ratios have large $^{26}Al/^{27}Al$ ratios. All these isotopic signatures are consistent with a SN origin for these grains – high $^{26}Al/^{27}Al$ ratios are found in the He/N zone that had undergone H burning, the He/C zone produces ^{18}O and ^{15}N excesses, and ^{28}Si is synthesized in the Si/S/O zones deep within the SN.

K and Ca isotopes: While most grains were found to have normal $^{41}K/^{39}K$ ratios (0.072), six grains have $^{41}K/^{39}K$ ratios ranging from 0.074 – 0.106. These ^{41}K excesses can be attributed to the decay of ^{41}Ca ($t_{1/2} = 1.03\times10^5$ a) because the intrinsic concentration of K in graphite grains is expected to be very low [10]. The inferred $^{41}Ca/^{40}Ca$ ratios for these grains range from 0.0003 to 0.01 (Figure 4). The values at the higher end of this range can be obtained from the He/C, C/O and O-rich zones of a Type II SN where ample neutrons are available for the production of ^{41}Ca [8]. All grains with high $^{41}Ca/^{40}Ca$ ratios have ^{18}O excesses and high $^{26}Al/^{27}Al$ ratios. Three of them are also enriched in ^{28}Si. Four graphite grains were found to have high $\delta^{42,43,44}Ca/^{40}Ca$ values that appear to be correlated. This trend is consistent with the increase in abundance of all the Ca isotopes at the bottom of the He/C zone of a SN. This region also has a large abundance of ^{15}N, which is consistent with the low $^{14}N/^{15}N$ ratios observed in two of the grains with large $\delta^{42,43,44}Ca$ values. Additionally, two grains that were found to have solar $\delta^{42,43}Ca/^{40}Ca$ values contain large ^{44}Ca excesses. Calcium-44 excesses due to slow neutron capture are expected to produce even larger $^{42,43}Ca$ excesses and hence we conclude that these excesses are most likely due to the decay of ^{44}Ti ($t_{1/2}$ = 58 a). The inferred initial $^{44}Ti/^{48}Ti$ ratios in these grains are 0.0027±0.0001 and 0.0032±0.0002. Much higher values have been observed in SiC-X grains and KE3 graphites from Murchison. Both these grains have ^{28}Si excesses that originate from the same region as ^{44}Ti – the Si/S zone in the interior of a SN. In order for these isotopic signatures to be present in graphite grains, material from the inner zones must be mixed with the C-rich He/C zone.

^{13}C enriched grains: Six graphite grains from this fraction have $^{12}C/^{13}C$ ratios less than 30. Four (g-8, 19, 14, 20) of these have excesses in ^{15}N, ^{18}O, ^{28}Si and/or large $^{26}Al/^{27}Al$ and $^{41}Ca/^{40}Ca$ ratios (Figures 1-5). Grain g-8 also shows evidence for ^{44}Ti. While all the excesses mentioned above can be obtained from different layers of a type II SN and mixed into the C-rich zones, it is very difficult to maintain the condition of C>O, required to condense graphite grains, combined with very low $^{12}C/^{13}C$ ratios. Some high-density graphites from Orgueil [6] have extremely high Ca and Ti isotopic ratios (explained by pure nucleosynthetic He-shell components of AGB stars) in conjunction with low $^{12}C/^{13}C$ ratios. Born-again AGB stars were suggested as a possible source for such grains. We did not obtain similar large Ca isotopic ratios in the ^{13}C-enriched, low-density graphite population and are, hence, still unable to explain the origin of these grains.

Conclusions: Isotopic anomalies observed in this study are much larger than those previously observed in the same graphite density fraction due to reduced contamination on the mount. Our isotopic data confirm that most low-density graphite grains from Orgueil originate from Type II SNe. We expect to obtain Ti data for these grains in the immediate future. Some questions still remain unanswered: Why do grains with high ^{18}O excesses possess close-to-solar $^{12}C/^{13}C$ ratios when these excesses are seen in zones that have almost pure ^{12}C? What is the stellar source of ^{13}C-enriched, low-density grains?

References: [1] Jadhav M. et al. (2006) *New Astron. Rev. 50*, 591–595. [2] Jadhav M. et al. (2006) *MAPS, 41*, A87. [3] Jadhav M. et al. (2008) *LPS XXXIX*, Abstract #1047. [4] Jadhav M. et al. (2007) *MAPS, 42*, A76. [5] Travaglio C. et al. (1999) *ApJ, 510*, 325-354. [6] Jadhav M., et al. (2008) *ApJ, 682*, 1479-1485. [7] Amari S. et al. (2005) *MAPS, 40*, A15. [8] Woosley S. E. and Weaver T. A. (1995) *ApJ Suppl., 101*, 181-235. [9] Nittler L. R. et al. (1995) *ApJ, 453*, L25-L28. [10] Amari S. et al. (1996) *ApJ, 470*, L101-L104.