CONFIRMATION OF EXTREME ^{54}CR ENRICHMENTS IN ORGUEIL NANO-OXIDES AND CORRELATED O-ISOTOPE MEASUREMENTS. Larry R. Nittler*, Jianhua Wang, Conel M. O'D. Alexander, Department of Terrestrial Magnetism, Carnegie Institution of Washington, Washington, DC 20015 (*e-mail: lnittler@ciw.edu).

Introduction: Endemic ^{54}Cr/^{52}Cr ratio variations in planetary materials [1-4] have been attributed to a heterogeneous distribution of highly ^{54}Cr-enriched presolar phases [5, 6]. One such carrier phase has been recently identified by NanoSIMS isotopic imaging as ≤100-nm-sized oxide (probably spinel) grains in the Orgueil (CI1) chondrite [5, 6]. In these studies, the SIMS spatial resolution (>400 nm) was much larger than the size of the grains, such that the measured anomalies (^{54}Cr/^{52}Cr up to 2.5 × the solar value) were lower limits due to contributions of signals from neighboring grains on the sample mounts. We used image simulations [6] to infer that the true ^{54}Cr enrichment of some grains was at least as high as 10 × solar and perhaps as high as 45 × solar, suggesting direct formation in one or more supernovae (SNe). Because the ^{54}Cr-rich zones of Type-II SNe are also highly enriched in ^{16}O, it would be expected that the ^{54}Cr-rich oxides would also be ^{16}O-rich, but the previous data were ambiguous due to signal overlap from adjacent grains. In this work, we have exploited the higher spatial resolution of the NanoSIMS Cs^+ beam to measure Cr isotopes as CrO^- secondary ions. We have confirmed that some Orgueil nano-oxides indeed have extreme ^{54}Cr enrichments as previously inferred from image simulations, and simultaneously determined their O-isotopic compositions. We have also identified several hundred new presolar oxide grains including a grain with the highest ^{17}O /^{16}O ratio yet observed.

Experimental: We analyzed the same Orgueil acid residue as in our previous study [6], mainly consisting of sub-μm Cr-rich spinels. O- and Cr-isotopes were measured with the Carnegie NanoSIMS 50L by rastering a ~0.5 pA (≈150 nm) Cs^+ beam over 20×20 μm^2 areas of the sample mount. Negative secondary ions of ^{16,17,18}O, AlO, ^{52}CrO, [^{54}CrO+^{54}FeO], and ^{56}FeO, as well as secondary electrons were collected in imaging mode. Typically 15-25 image planes were acquired and summed together following drift correction, for a total measurement time per 78×78 nm^2 pixel of ~0.1 sec. The contribution of ^{54}FeO to the measured signal at mass 70 was corrected for by assuming terrestrial ^{54}Fe/^{56}Fe. Images were quantitatively analyzed with the L'image software (L. R. Nittler).

Results: We analyzed 80 areas, each containing hundreds of individual sub-μm grains. Four grains with extremely high inferred ^{54}Cr/^{52}Cr ratios were unambiguously identified. Fig. 1 shows NanoSIMS ion images for one such <150 nm grain. After a ~8% correction for ^{54}Fe, this grain is inferred to have ^{54}Cr/^{52}Cr≈1, or 36 × solar (δ^{54}Cr≈37,000 ‰). Three other identified grains of similar size have δ^{54}Cr values ranging from≈10,000 ‰ to ≈30,000 ‰ (Fig. 2). All four have terrestrial O-isotopic ratios within 50–200‰ 1-σ errors. We have also identified a number of candidate grains with significant, but smaller ^{54}Cr anomalies; it is not yet known if these grains have true ^{54}Cr enrichments similar to those of the extreme grains but diluted by neighboring material, or if they represent another population of Cr-anomalous grains with more modest isotopic anomalies. Moreover, a few images revealed grains with even higher apparent ^{54}Cr values than shown in Fig. 2 (>100 × solar). However, these extreme anomalies still need to be confirmed.

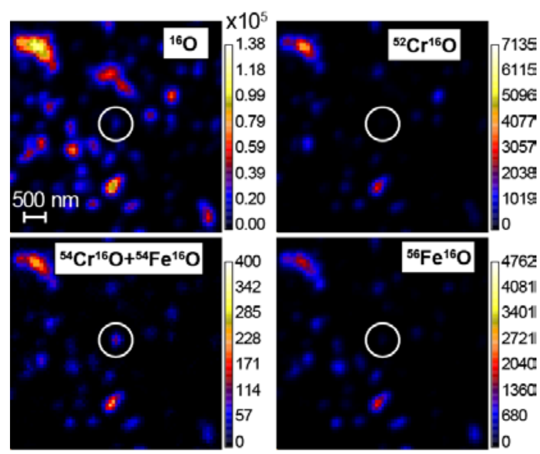

Fig. 1: Isotopic images (5×5 μm^2) of an extremely ^{54}Cr-enriched grain (circle).

Analysis of the O-isotopic images is in progress, but we have clearly identified at least 340 presolar grains with large O-isotopic anomalies (Fig. 3). Many of these are Al-rich based on measured AlO^-/O^- ratios, most likely Al_2O_3 or $MgAl_2O_4$. The grains have O-isotopic compositions spanning the ranges previously observed in presolar oxides and silicates. The lack of grains with ^{18}O/^{16}O<10^{-4} compared to previous data can be attributed to dilution of measured signals on the crowded sample mount. Three grains with extreme ^{17}O enrichments (^{17}O/^{16}O> 0.01), two grains with extreme ^{18}O enrichments (^{18}O/^{16}O≈0.015), and one grain with a very low ^{17}O /^{16}O ratio of 6×10^{-5} were found. One of the ^{17}O–rich grains has ^{17}O/^{16}O≈0.1, a factor of two higher than the most ^{17}O–rich grain previously reported. No obvious ^{54}Cr anomalies have been identified in any of the O-anomalous grains.

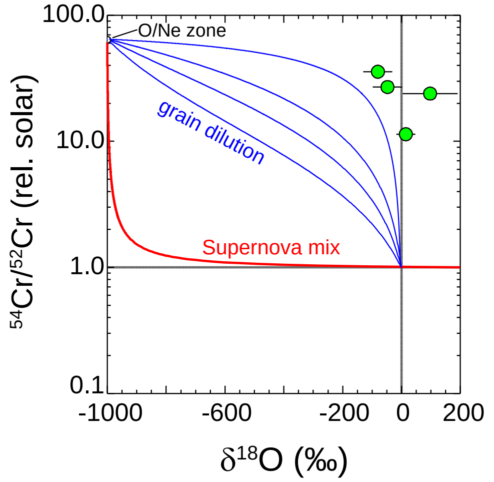

Fig.2: ^{54}Cr/^{52}Cr ratios and δ^{18}O values for 4 highly ^{54}Cr-enriched grains identified in this study (green circles). Mixing curves are described in the text.

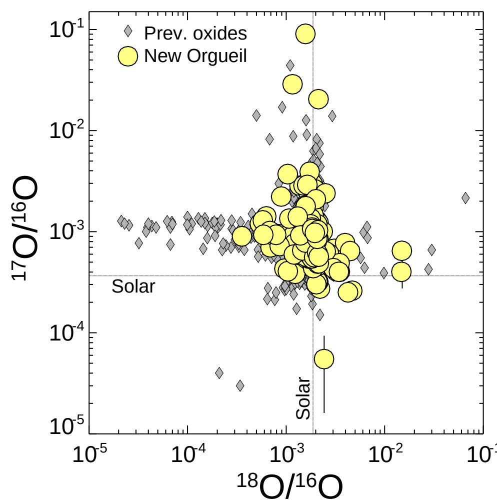

Fig.3: O-isotopic ratios of presolar oxide grains identified in this study compared to those previously reported [e.g., 7].

Discussion and Conclusions: Measurement of Cr isotopes as negative CrO secondary ions has allowed us to directly confirm that some nano-oxides in Orgueil have ^{54}Cr/^{52}Cr ratios of order tens of times higher than the solar ratio. Neutron-rich nuclei, including ^{54}Cr, are believed to originate in neutron-rich environments in both Type Ia (SNIa) and Type II SNe (SNII) [8, 9]. In fact, the grain Cr-compositions are similar to those predicted to arise from *s*-process nucleosynthesis in the O/Ne and O/C zones of SNII [10]. However, the ^{54}Cr-rich grains do not have the expected ^{16}O-rich signature for direct condensates from these zones. The red curve on Fig. 2 indicates mixing of material from the O/Ne zone of a 15M$_{\odot}$ SNII [10] with a mixture of outer-zone material that explains the average O-isotopic composition of presolar SN oxides [7]. The blue curves indicate expected compositions for dilution of an O/Ne zone composition with isotopically normal material on the sample mount due to beam overlap, for a range of assumed Cr/O ratios for the end-member grains based on ratios observed in the Orgueil ion images. Clearly, the O-isotopic compositions of the grains are not consistent with direct condensation in the O-rich SNII zones even if the true signatures are somewhat diluted by contaminating signals during the SIMS measurements. One possible explanation is that the grains originated not as oxides, but as metal or carbides that later reacted with isotopically normal O in space or on the meteorite parent body. If so, it is not possible to ascribe a SNIa or SNII origin to the grains purely on the basis of the Cr isotopes. The correlation observed between ^{54}Cr and ^{50}Ti anomalies in bulk meteorites [11] is also consistent with an origin in either type of SN. A SNII origin has been favored since seeding of the early solar system by SNII ejecta may naturally explain both the presence of short-lived radionuclides and the heterogeneous distribution of ^{54}Cr anomalies among planetary materials [5, 6]. However, ^{50}Ti anomalies in meteoritic hibonite [12] may point to a SNIa component in the early solar system and if the extremely high ^{54}Cr enrichments we have tentatively identified (see above) are borne out, these might also require such an origin.

Extreme ^{17}O enrichments cannot be explained by nucleosynthesis and mixing in low-mass AGB stars, so classical novae have been invoked to explain grains with ^{17}O/^{16}O ratios >~0.005 [13, 14]. The origin of the highly ^{17}O-depleted grain is unclear but may reflect mixing in a SN. Additional isotopic measurements (e.g., Al-Mg, Ti) of the unusual grains identified here should help better constrain their origins.

References: [1] Birck J.-L. and Allègre C. J. (1984). *GRL, 11*, 943-946. [2] Rotaru M., et al. (1992). *Nature, 358*, 465-470. [3] Trinquier A., et al. (2007). *Astrophys. J., 655*, 1179-1185.[4] Qin L., et al. (2010). *GCA, 74*, 1122-1145. [5] Dauphas N., et al. (2010). *Astrophys. J., 720*, 1577-1591. [6] Qin L., et al. (2011). *GCA, 75*, 629-644. [7] Nittler L. R., et al. (2008). *Astrophys. J., 682*, 1450-1478. [8] Hartmann D., et al. (1985). *Astrophys. J., 297*, 837-845. [9] Meyer B. S., et al. (1996). *Astrophys. J., 462*, 825. [10] Woosley S. E. and Heger A. (2007). *Physics Rep., 442*, 269-283. [11] Trinquier A., et al. (2009). *Science, 324*, 374-376. [12] Liu M.-C., et al. (2009). *Geochimica et Cosmochimica Acta, 73*, 5051-5079. [13] Vollmer C., et al. (2007). *Astrophy. J., 666*, L49-L52. [14] Gyngard F., et al. (2010). *Astrophy. J., 717*, 107-120.