DISTRIBUTION AND CARRIERS OF CR ISOTOPIC ANOMALIES IN THE INNER SOLAR SYSTEM.
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Introduction: Chromium isotopic anomalies, most notably enrichments in ^{54}Cr, are widely distributed in inner Solar System materials [1-6]. These variations have been attributed to nucleosynthetic effects, possibly carried by presolar grains. Alternatively, early irridiation by energetic particles from the young sun may also generate large ^{54}Cr enrichments in the protoplanetary dust [6]. To differentiate between the two processes, one will need to rely on the identification of the carrier (s) of the ^{54}Cr excess. We first reported the discovery of sub-micron ^{54}Cr-rich (up to 300 ‰) oxide grains in an Orgueil acid residue [7]. Continued searches have identified additional grains with more extreme ^{54}Cr enrichments, reaching at least as high as 1500 ‰ [8, 9]. We found no resolved O isotope anomalies for ^{54}Cr-rich grains. Auger spectroscopic measurements indicated that they are likely to be Cr-bearing spinels.

Methods: The organic-rich CsF/HCl Orgueil dissolution residue was ashed in an O-plasma to remove C. The remaining mineral grains were dispersed as a liquid suspension onto a gold substrate. SEM examination of the mount showed that the sample consists mostly of sub-micron Cr-rich oxide grains (most likely Cr spinels with varying amounts of Mg, Fe and Al) and a small amount of SiC. Isotopic measurements were made with the Carnegie NanoSIMS 50L ion microprobe in imaging mode. The density of grains on the mount is very high, allowing 100s to 1000s of grains to be analyzed in each image, but with the drawback that isotopic compositions of single grains can be strongly influenced by dilution of signals from surrounding material, particularly for the Cr measurements. Cr isotopic images (50, 52, 53, 54) of hundreds of thousands of grains in the residue were collected. Whenever O isotope images were collected, O isotopes were first measured prior to Cr isotope measurements with a ~100 nm Cs^+ beam rastered over 20×20 μm areas and simultaneous collection of negative secondary ions of $^{16,17,18}O^-$, $^{28}Si^-$, $Al^{16}O^-$ and $^{52}Cr^{16}O^-$. Cr isotopes were measured with a 300-700 nm O^- beam and simultaneous collection of positive secondary ions of $^{28}Si^+$, $^{48}Ti^+$, $^{50}Cr+^{50}Ti^+$, $^{52}Cr^+$, $^{53}Cr^+$, $^{54}Cr+^{54}Fe^+$, and $^{56}Fe^+$. Isotopic ratio images were generated and isotopic ratios for individual regions determined through image processing. Data for individual grains were normalized to the average composition in an image. ^{54}Cr and ^{50}Cr were corrected for isobaric interference from Fe and Ti, respectively, with the assumption of normal isotopic compositions for these elements.

Results: NanoSIMS imaging revealed 10 regions with positive δ^{54}Cr anomalies up to 1500 ‰. These regions are not associated with δ^{50}Cr and δ^{53}Cr anomalies, except one region where a small negative δ^{53}Cr anomaly was detected. These δ^{54}Cr anomalies are clearly resolvable with our analytical uncertainties, and cannot be caused by analytical artifacts. The anomalies are also very unlikely to reflect ^{54}Fe-rich supernovae grains. Because of dilution from surrounding normal grains, the measured anomalies are lower limits, and we estimated that the actual $^{54}Cr/^{52}Cr$ enrichments in the grains may be as high as 50 times solar. Such enhancements in $^{54}Cr/^{52}Cr$ ratio, combined with close to normal $^{50}Cr/^{52}Cr$ and $^{53}Cr/^{52}Cr$ ratios, cannot be produced by early irradiation by the young Sun, or in Type Ia supernovae, but strongly favor an origin in Type II supernovae. Oxygen isotope imaging of a subarea mapped for Cr isotopes revealed ~160 presolar oxide grains. The grains span a similar range of O isotope compositions as previously observed for presolar oxides [10] and silicates [11], and the majority of them were probably derived from asymptotic giant branch (AGB) stars. However, three grains with extreme δ^{17}O enrichments and two grains with extreme δ^{18}O enrichments may have formed in novae and supernovae, respectively [9]. For three δ^{54}Cr rich grains, O isotope data were extracted and did not show resolvable anomalies. However, negative δ^{17}O and δ^{18}O isotope anomalies are expected if they are indeed from the inner zones of Type II supernovae, and cannot be ruled out because negative anomalies are most vulnerable to dilution from surrounding minerals. SEM imaging showed that the δ^{54}Cr-rich regions are often associated with more than one grain. However in a few cases the anomalous grains were well separated from other grains - they are usually < 200 nm in size and smaller than the typical grain size in the sample mount. Data from both the NanoSIMS images and subsequent Auger analyses indicate that the grains contain Cr, O, and in some cases other elements including Al and/or Ti. They are likely to be Cr-bearing spinels.

Late SN Injection: The ^{54}Cr-rich grains, along with other SN-derived oxide/silicates grains plot on a single SN mixing line. Given the extremely complicated mixing conditions in supernova ejecta and tremendous variations observed in the isotopic compositions of SN-derived SiC grains, the unusually limited distributions of O isotopic compositions of these grains are most plausibly explained by a single supernova injection event. These supernova grains seem decoupled from other types of presolar grains, suggesting that they were injected later into the solar nebula. Despite their small sizes, the extreme ^{54}Cr enrichments of the grains observed here indicate that they could be a major contributor to ^{54}Cr variations in bulk chondrites. The variability in bulk ^{54}Cr/^{52}Cr between meteorite classes likely reflects heterogeneous distribution of the ^{54}Cr carrier phases in the already-formed solar protoplanetary disk following a single late supernova injection event.

Collatoral Isotopic Sequences of SN Injection: A key feature of our late SN injection model is the selective sampling of the SN ejecta implied by the presolar grain data (e.g., the particular mixing of O/Ne and O/C zones of SN but not others). This is in strong contrast to previous models [12, 13], which assumed bulk SN yields for calculating collateral isotope effects of late SN injection. Assuming that there is no element fractionation in the bulk dust condensed in these zones and that different types of dust survive injection into the disk in the same relative proportions, our model predicts correlations between ε^{54}Cr, ε^{46}Ti and ε^{50}Ti, in reasonably good agreement with the observations on bulk chondrites [14]. Considering the large uncertainties, this supports our contention that the bulk meteorite anomalies reflect variable amounts of material from the O/Ne and O/C zones of a Type II supernova. A correlation between ^{62}Ni/^{58}Ni ratios and ^{54}Cr/^{52}Cr ratios was reported for bulk meteorites, though the magnitude of the observed ^{62}Ni effect is some five times smaller than that of ^{54}Cr [15]. With the oversimplification of no elemental fractionation, our model predicts depletion in ^{62}Ni/^{58}Ni rather than excess. However, equilibrium condensation calculations based on a similar SN model to the one we consider predict Ti and Cr to condense into refractory oxides, but Ni into metallic alloys, in the O/Ne and O/C zones [16]. The different condensation temperatures and material properties of oxides versus metal thus provide a route to element fractionation in the injection model. This also might be the case for ^{60}Fe. The case of short-lived ^{26}Al is much more complicated, because it is produced both in the ^{54}Cr-rich zones and the outer zones which dominate the compositions of the ^{18}O-rich presolar SN grains. Detailed consequences of our model will be the focus of future work.

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