

Evidence for supernova injection into the solar nebula and the decoupling of r -process nucleosynthesis

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The isotopic composition of our Solar System reflects the blending of materials derived from numerous past nucleosynthetic events, each characterized by a distinct isotopic signature. We show that the isotopic compositions of elements spanning a large mass range in the earliest formed solids in our Solar System, calcium–aluminum-rich inclusions (CAIs), are uniform, and yet distinct from the average Solar System composition. Relative to younger objects in the Solar System, CAIs contain positive r -process anomalies in isotopes $A < 140$ and negative r -process anomalies in isotopes $A > 140$. This fundamental difference in the isotopic character of CAIs around mass 140 necessitates (i) the existence of multiple sources for r -process nucleosynthesis and (ii) the injection of supernova material into a reservoir untapped by CAIs. A scenario of late supernova injection into the protoplanetary disk is consistent with formation of our Solar System in an active star-forming region of the galaxy.

isotopic anomalies | early Solar System | H-Event | nebular disk

Our knowledge of the formation and evolution of our Solar System principally comes from (i) astrophysical observations, which provide views of other nascent stellar systems; (ii) theoretical models, which provide constraints in such systems; and (iii) direct evidence gathered from the study of meteorites that sample the earliest epoch of Solar System history. However, the details about the galactic environment in which our Solar System formed are still a matter of intense debate. Central to this effort is the study of short-lived radionuclides and nucleosynthetic anomalies found in meteorites, which constrain if, how much, and when our Solar System was injected with supernova material. This present work centers on the stable isotope signatures of the Solar System's earliest solids to unravel the earliest events that occurred after the birth of our Solar System.

The elemental and isotopic composition of our Solar System reflects a mixture of materials derived from nucleosynthetic reactions in past generations of stellar environments. Isotopes of elements heavier than nickel are produced by three principal mechanisms: the p -, s -, and r -processes (1, 2). The p -process creates isotopes through photodisintegration, proton capture, and neutrino reactions in supernovae, forming neutron-deficient nuclei. The s -process occurs from slow neutron addition in asymptotic giant branch stars, in which isotope production moves up the “valley of stability” through neutron addition to create isotopes that are either stable or β -decay to stable isotopes. Rapid neutron addition, better known as the r -process, occurs in the extremely high neutron densities found in supernovae. The r -process creates neutron-rich, radioactive isotopes that β -decay to stability, resulting in neutron-rich stable isotopes. Thus, for elements heavier than Ni, the combination of the p -, s -, and r -processes of nucleosynthesis is ultimately responsible for the isotope abundances present in all Solar System materials. Although the general outline of isotope production is understood, the details of how products from various nucleosynthetic processes are assembled into the material that formed our Solar System are not well defined.

Variations in the nonradiogenic isotope abundances of meteoritic materials have often been interpreted as nucleosynthetic signatures

resulting from the input of distinct materials produced from p -, s -, and r -processes (3–12). These isotopic compositions have been used to support arguments for nebular heterogeneity and to assess the degree of mixing in the early solar nebula, as well as to constrain the physical characteristics of potential carriers of the isotope anomalies. Previous work on the isotopic compositions of bulk chondrites and their various components (obtained through differential leaching or by mineral separations) has provided an immense amount of information. Nevertheless, the complex isotopic systematics measured for bulk chondrites preclude simple interpretations. Calcium–aluminum-rich inclusions (CAIs) are found in chondrite meteorites and are important carriers of isotopic information about the early Solar System. Unlike bulk chondrites, CAIs are not physical mixtures of various Solar and refractory pre-Solar System components. Instead, they are high-temperature primary condensates from a nebular gas. Although some CAIs have experienced complex histories of melting and evaporation (13, 14), these processes are generally considered to be closed, and consequently have not resulted in the addition of large amounts of material to CAIs after they formed. Thus, these objects represent the first solids to form in the cooling solar nebula, and as such they provide primary information about the isotopic composition and degree of mixing within the CAI-forming region near the young Sun (15).

The elements strontium (Sr), molybdenum (Mo), barium (Ba), neodymium (Nd), and samarium (Sm) each have multiple stable isotopes. The isotopes of each of these elements have variable inputs from p -, s -, and r -process nucleosynthesis, which makes these elements ideal for studying nucleosynthetic signatures in meteoritic material. Previous work on an exceedingly rare subset of isotopically anomalous CAIs known as Fractionation and Unknown Nuclear effect (FUN) inclusions has clearly demonstrated that they deviate substantially from the isotopic compositions of all other Solar System materials. In addition, the FUN inclusions have isotopic compositions that differ from one another (i.e., 16, 17). The

Significance

We present isotopic evidence that supports the injection of supernova material into the early Solar System within a small window of time around the formation of the first solids in the protoplanetary disk. This injected material was not incorporated into the first solids, but subsequently changed the elemental and isotopic composition of the nascent Solar System. The isotopic differences between the first solids and subsequently formed matter in the Solar System require the existence of multiple sources for r -process nucleosynthesis.

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rarity of FUN inclusions (only a few have been identified and only two have been extensively studied) highlights the uniqueness of these samples and underscores their atypical provenance. Although these unique samples are important for understanding conditions in the early Solar System, FUN inclusions represent only a vanishingly small fraction of refractory inclusions, and therefore are not the focus of this work, which centers on the most abundant type of CAIs (i.e., those devoid of “FUN” effects), simply referred to hereafter as “CAIs.”

Previous studies of CAIs have primarily focused on the isotopic compositions of single elements and demonstrate that they differ from terrestrial compositions. The observed anomalies in single elements have been modeled using addition or subtraction of various nucleosynthetic components. However, unlike isotopic investigations of FUN inclusions (e.g., 16, 17), these studies have not determined and compared the isotopic compositions of multiple elements spanning a large mass range in the same sample set. This integrated study presents the Sr, Mo, Ba, Nd, and Sm isotope compositions determined on multiple individual CAIs from the Allende meteorite to provide a framework to better understand the origin of the vast majority of samples in the CAI population. This sample set represents a range of petrographic and geochemical types from coarse- to fine-grained and from group II to nongroup II designations (18).

Results and Discussion

The Sr, Mo, Ba, Nd, and Sm measurements are presented in Fig. 1 and demonstrate that the isotopic compositions of each of these elements in Allende CAIs are (i) distinct from their respective terrestrial standards, (ii) identical between CAIs at the level of analytical precision, and (iii) in agreement with other studies of Allende CAIs for the three previously reported elements—that is, Sr (19–21), Mo (10), and Ba (3, 9). Taken in whole, these data necessitate that CAIs were formed in a homogeneous reservoir that was isotopically distinct from the “average” Solar System, given by terrestrial isotopic compositions. This reservoir is characterized by resolvable relative excesses in ^{84}Sr , ^{92}Mo , ^{94}Mo , ^{95}Mo , ^{97}Mo , ^{100}Mo , ^{135}Ba , and ^{137}Ba , combined with resolvable relative deficits in ^{145}Nd , ^{148}Nd , ^{150}Nd , ^{144}Sm , ^{147}Sm , ^{149}Sm , ^{152}Sm , and ^{154}Sm . From these analyses it is apparent that excesses in *r*-process isotopes are present for masses below 140, whereas deficits in *r*-process isotopes are present above mass 140 (as displayed in Fig. 2). All isotopic ratios measured in this investigation, along with standards, are given in *SI Appendix*.

Isotopic anomalies for a number of elements have been observed in an assortment of Solar System materials, and a variety of mechanisms have been proposed to account for the isotopic deviations. Below we discuss some of the possibilities that could

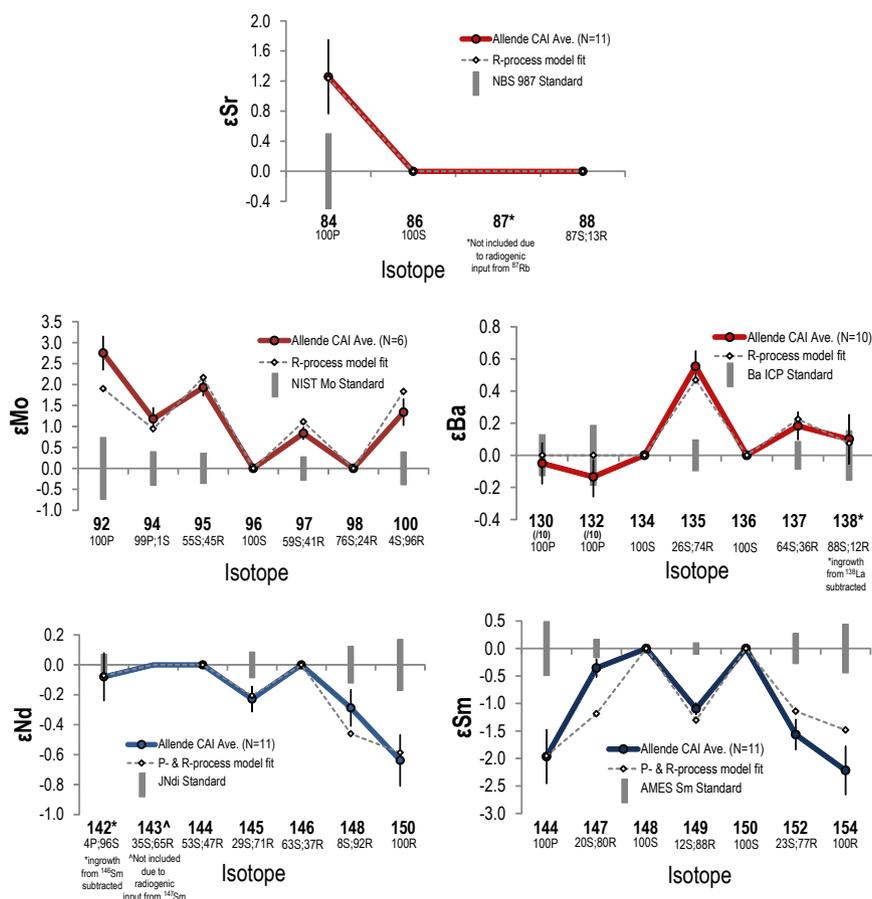


Fig. 1. The average isotopic compositions of Sr, Mo, Ba, Nd, and Sm (expressed in standard ϵ notation, or parts per 10,000 relative to the terrestrial standard for each element) in multiple Allende CAIs. The uncertainties shown for each isotope are $2 \times$ the SD based on the data for all CAIs measured. The uncertainty shown for the terrestrial standards (gray bars) represents the 2SD of the long-term reproducibility of that standard during this study. The contributions of the *p*-, *s*-, and *r*-processes for each isotope are given below the isotope number. In the case of Ba, the data (and associated uncertainties) for ^{130}Ba and ^{132}Ba are divided by 10 to fit on the same scale. Internal normalization is used for each element to correct for mass-dependent fractionation. The data and normalizations for individual samples and standards are given in *SI Appendix*.

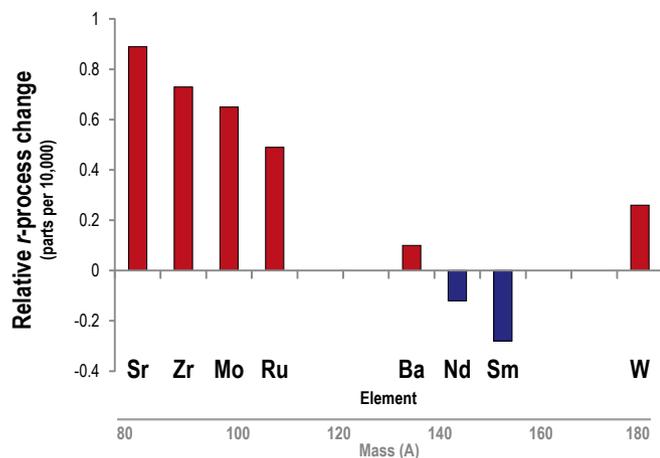


Fig. 2. Mass influence on the magnitude of the *r*-process anomaly. The relative change in *r*-process anomaly between elements measured in CAIs and terrestrial standards is plotted by a function of mass. Data for Sr, Mo, Ba, Nd, and Sm come from this study. Data from common CAIs for the elements Zr, Ru, and W are taken from refs. 41, 42, and 26, respectively.

cause the observed isotopic differences between CAIs and the average Solar System.

The capture of neutrons has the potential to measurably alter the isotopic composition of materials. The probability of neutron capture is proportional to the neutron cross-section of an individual isotope. For example, the likelihood of neutron capture is very high for ^{149}Sm , which has a large neutron cross-section of $\sim 41,500$ barns. Consequently, several studies—particularly of lunar samples—have observed deficits in ^{149}Sm (and corresponding excesses in ^{150}Sm) and attributed the anomalies to neutron capture (22). However, applying numerous normalization schemes to our Sm data demonstrates that the observed isotopic anomalies in Sm are inconsistent with neutron capture (SI Appendix). Moreover, as the neutron capture cross-section of ^{149}Sm is ~ 100 times larger than any other isotope analyzed for this study, neutron capture effects cannot produce the anomalies observed in other isotope systems.

A second possible cause for apparent isotopic anomalies is the incomplete digestion of all phases in CAIs. For example, presolar silicon carbide (SiC) grains are highly refractory, μm -sized particles that are notoriously difficult to dissolve and have been shown to have isotopic compositions that are markedly different from the bulk Solar System (23). Thus, if present in CAIs, the incomplete digestion of SiC grains could in principal produce isotopic anomalies that are similar to those observed in CAIs. However, the isotopic compositions of CAIs determined in this and other studies are uniform, despite sample digestion protocols that involve vastly different techniques. Furthermore, the similarity of isotopic signatures in all CAIs would require that the initial fraction of the undissolved material be the same in group II, nongroup II, coarse-, and fine-grained CAIs. Furthermore, the proportion of undissolved SiC grains must be exactly the same in all types of CAIs examined in several independent studies. Finally, no refractory presolar grains have been reported in CAIs. Therefore, it is highly improbable that incomplete digestion of presolar phases in CAIs is the cause of the observed isotopic anomalies.

Theoretical work by Fujii et al. (24) and Moynier et al. (25) suggests that isotopic fractionation driven by the nuclear field shift may account for some isotopic anomalies measured in CAIs. When applied to this dataset, some of the measured isotopic patterns do indeed match those predicted for fractionation resulting from nuclear field shift effects. However, large mismatches also exist in

many isotopes, such as ^{100}Mo , ^{138}Ba , ^{145}Nd , ^{149}Sm , and ^{154}Sm (see SI Appendix for more information). Whereas the nuclear field shift has the potential to cause non-mass-dependent fractionation in CAIs, the inability of this mechanism to account for the entire range of isotopic abundances observed in the CAI dataset excludes this as the primary source of isotopic variation in Allende CAIs. Furthermore, the isotopic homogeneity of these CAIs requires any fractionation caused by the nuclear field shift to affect each sample equally, regardless of CAI type or group. This scenario is very unlikely given the variety of CAIs measured in this and other studies.

To explain anomalies found in various isotope systems in presolar grains, FUN inclusions, or CAIs, previous studies have invoked excesses or deficits in the products of one or more nucleosynthetic processes in the materials studied. For example, apparent excesses in the *r*-process isotopes of Mo were reported in CAIs (10) that were attributed to a contribution of material that was enriched in *r*-process components. Simple mixing models of pure *p*-, *s*-, and *r*-process components reproduce the isotopic compositions of individual elements quite well. In fact, models using addition or subtraction of nucleosynthetic components match the isotopic data for all of the individual elements measured in our study as well (Fig. 1). The good fit of these models to individual elements provides strong evidence that the isotopic patterns are caused by nucleosynthetic processes. However, these basic modeling exercises cannot account for the isotopic compositions of the entire range of elements measured in our sample set using the same proportions of nucleosynthetic components. This is illustrated by the fact that individual CAIs with apparent *p*- or *r*-process excesses in Sr and *r*-process excesses in Ba and Mo also contain *r*-process deficits for Nd and *p*- and *r*-process deficits for Sm in the same sample (Fig. 1). [As Sr only has four stable isotopes and there is further complication from the radiogenic ingrowth of ^{87}Sr from ^{87}Rb , the exact location of the nucleosynthetic anomaly is unknown, and can be on either ^{84}Sr (*p*-process only) or ^{88}Sr (minor *r*-process). See Moynier et al. (19) for details.] Consequently, a model that reproduces Sr, Ba, and Mo isotopic compositions of CAIs by adding a larger proportion of *r*-process components produces gross mismatches for the modeled and measured isotopic compositions of Nd and Sm.

The distinct isotopic signatures in CAIs indicate that they originated from a different reservoir than other Solar System materials that contain the less refractory material, such as bulk chondritic meteorites or bulk Earth (that are taken to represent the average Solar System). These reservoirs could have been separated by (i) space, in which different regions of the solar nebula had distinct compositions, and/or (ii) time, in which a temporal evolution in the isotopic composition of the solar nebula occurred. If spatially distinct reservoirs were responsible for the different isotopic compositions of the CAI-forming region and the bulk Solar System, their dissimilar *r*-process signatures may hint at their origin and constrain a specific kind of *r*-process material that is involved in CAI formation. When the relative changes in *r*-process abundance between CAIs and terrestrial standards for a given element are plotted by mass (Fig. 2), a trend is evident in which the magnitude of the *r*-process anomalies decreases with increasing atomic mass (A). There is a change from *r*-process excesses in isotopes lighter than mass 140 to *r*-process deficits in isotopes greater than mass 140. This marked change at $A \sim 140$ suggests a fundamental difference between the creation mechanism of *r*-process isotopes $A < 140$ and isotopes $A > 140$. Additionally, another similar change may occur in heavier elements ($A \sim 180$), as shown by apparent *r*-process excesses reported in W (26). These data provide physical evidence for the decoupling of *r*-process nucleosynthesis—in which the production of the *r*-process isotope is controlled by mass.

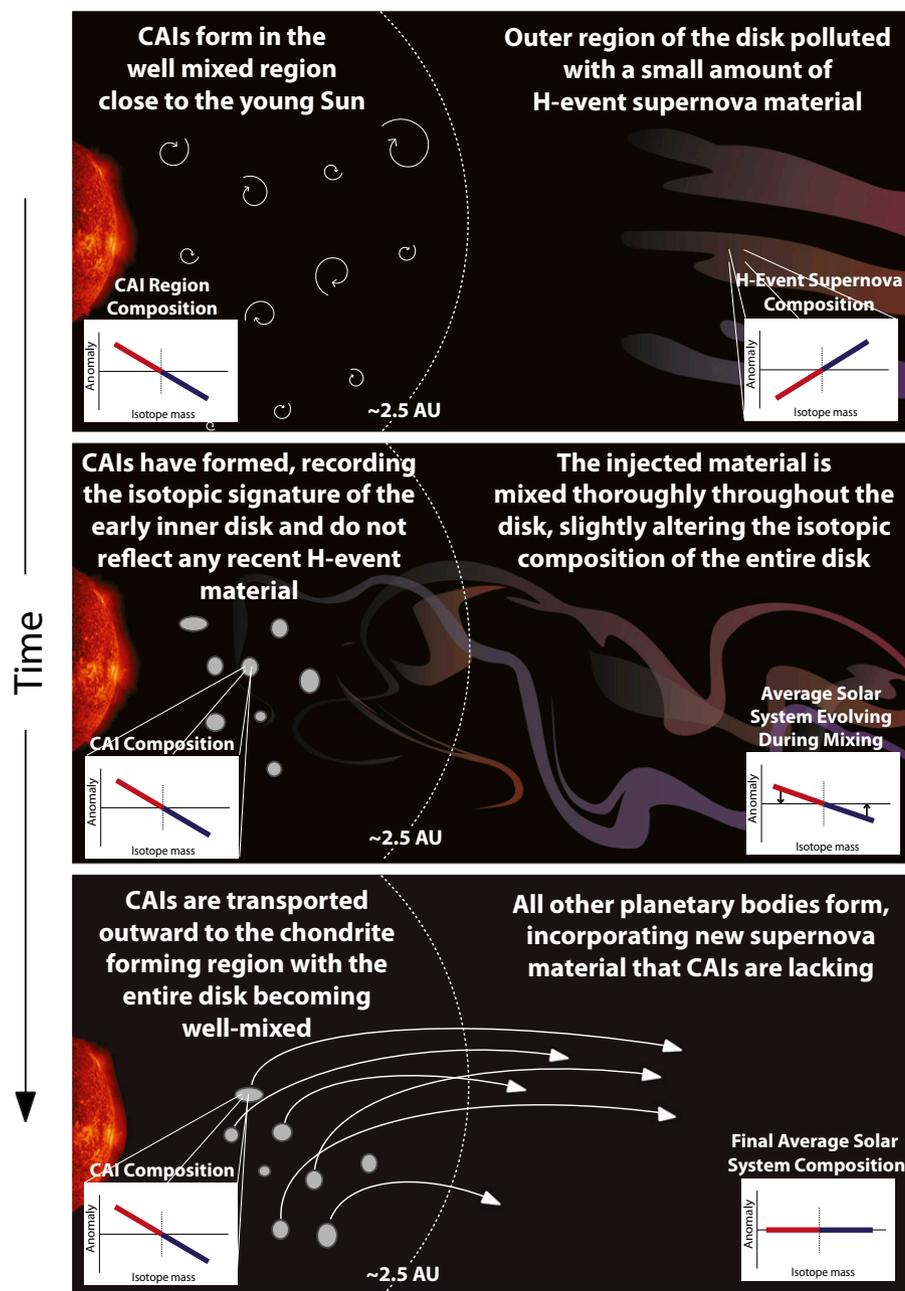


Fig. 3. Supernova injection after CAI formation. A schematic of the very early Solar System explaining how the isotopic signatures of CAIs and the average Solar System differ. The approximated isotopic signatures of various regions and materials are shown graphically to demonstrate how the initial CAI composition is “locked in” before subsequent mixing of late injected H-event supernova material, resulting in what is now considered the average Solar System isotopic composition.

Multiple sources for *r*-process nucleosynthesis have been proposed based on observed variations in the initial abundances of short-lived *r*-process isotopes ^{129}I , ^{182}Hf , and ^{244}Pu in early Solar System materials and the observation of *r*-process enrichments in halo stars (27–32). Based on discrepancies in the predicted initial abundances of these isotopes, type II supernovae have been suggested to be categorized by the masses of the *r*-process nuclei that they produce: lighter *r*-process isotopes ($A \leq 140$) are produced in L-event supernovae, whereas heavier *r*-process isotopes ($A > 140$) are created by H-event supernovae (27, 28). Additionally, a third type of event, termed an A-event, may be responsible for the highest mass *r*-nuclei (29, 30). Type II supernovae are not only categorized by the isotope masses they produce, but also by the frequency at which they occur in the galaxy. To account for the observed initial ^{129}I and ^{182}Hf isotopic abundances inherited into our solar nebula, H-event supernovae must occur ~ 10 times more frequently than L-event supernovae.

The least common A-event produces the heaviest short-lived radionuclides, such as ^{244}Pu and ^{247}Cm . These predictions are consistent not only with model calculations of element production in supernovae (32), but also with various studies that have observed a decoupling of the *r*-process at $Z \sim 56$ ($A \sim 140$) in ancient stars, which require three distinct *r*-process production sites (33–35). The variations in the abundances of *r*-process isotopes with increasing mass number observed in CAIs are consistent with this model of multiple type II supernovae contributions to the solar nebula.

Our dataset not only provides direct stable isotope evidence of *r*-process decoupling, but constrains the timing, lateral extent of mixing, and source of supernova material added to the early solar nebula. A physical mixing model can be produced that accounts for the apparent *r*-process excesses in Mo and Ba and apparent *r*-process deficits in Nd and Sm by considering the isotopic production mechanisms outlined above in the context of

what is known regarding the formation of CAIs (Fig. 3). Because H-event supernova material is relatively depleted in *r*-process isotopes $A \leq 140$ (e.g., Mo, Ba) and relatively enriched in *r*-process isotopes $140 \leq A \leq 200$ (e.g., Nd, Sm) (29), incomplete mixing of various supernova-derived material could, in principle, produce the observed isotopic variations. CAIs are thought to have formed in the innermost portion of the accreting nebular disk (15) in a short time frame, possibly as short as 20–50 thousand years (36–38). Therefore, these CAIs would have preserved a snapshot of the isotopic composition from the well-mixed CAI-forming region close to the young Sun. The addition of a small amount of H-event material to the protoplanetary disk that did not interact with this CAI-forming region, due to the separation of these regions by either space or time, would slightly alter the isotopic composition of the entire disk. A disk produced in this way would result in primitive chondrites (that contain contributions from refractory CAIs, as well as less refractory components such as chondrules and matrix that formed in either spatially or temporally distinct regions of the solar nebula) having bulk compositions approximating that of the average Solar System. At some point before the accretion of primitive chondrite parent bodies, CAIs were transported outward to the chondrite-forming region at ≥ 2.5 AU (15, 39). At this point the disk was well mixed, leaving CAIs (with a slightly different isotopic signature of the inner disk) as isotopically anomalous interlopers in the chondrite-forming region.

To preserve isotopically distinct reservoirs, the estimated timescale for mixing in the young protoplanetary disk places a strong time constraint on the addition of H-event material to the early Solar System. Mixing models demonstrate that homogenization of the nebular disk occurs on timescales of $\sim 100,000$ y (40). As such, the addition of the H-event supernova material must have occurred no earlier than 100,000 y before CAI formation and before the condensation of the less refractory phases that make up the bulk of primitive chondritic meteorites that represent average Solar System composition.

The observed isotopic compositions of CAIs could also be produced if CAIs formed in a region that was relatively enriched in L-event material compared with the rest of the Solar System (as opposed to lacking H-event material from a supernova). However, this scenario would require a more lengthy isolation of the CAI reservoir from the rest of the Solar System. This scenario is less likely given the relatively short mixing timescales predicted by early Solar System models of the nebular disk (40).

It is noted that this proposed model does not account for the isotopic signatures of FUN inclusions. If FUN inclusions originated in sources separate from those of common CAIs, these sources would have only existed for a brief period when FUN inclusions were formed. These transient sources must have been

very poorly mixed and presumably incorporated material with isotopic compositions that were different from the reservoir from which common CAIs formed.

The abundances of short-lived radionuclides in the early Solar System can be used to understand the balance between the creation of such nuclides in stars and their decay in the interstellar medium. This balance of creation and decay greatly influences our understanding of the dynamic evolution of the galaxy and the nucleosynthetic history of matter (30). Because the stable (and much more abundant) isotopic compositions in CAIs differ from the terrestrial standard compositions by only tens of parts per million, the short-lived radionuclides associated with the proposed H-event material would be undetectable with our current level of precision. Whereas data from our study cannot be directly used to refine production ratios of any short-lived radionuclides, they do provide evidence of the decoupling of *r*-process nucleosynthesis, indicating that current models of radionuclide production must be viewed with caution.

Methods Summary

Aliquots from previously dissolved CAIs of the Allende meteorite [from which U was separated, as previously described (18)] were processed for Sr, Mo, Ba, Nd, and Sm isotope measurements. This sample set includes three group II CAIs (CAI 166, 167, and 175) and eight nongroup II CAIs (CAI 164, 165, 168, 170, 171, 172, 173, and 174) (18). For this study, the elements of interest were separated from the rest of the CAI matrix using previously established procedures for each element, with references as follows: Sr (16), Mo (10), Ba, Nd, and Sm (6). Complete separation of Nd and Sm from one another (as well as separation from other isobaric interferences) was accomplished using multiple passes through methalactic acid column chemistry.

The Mo isotope compositions of samples and standards were measured on a ThermoNeptune multicollector inductively coupled mass spectrometer housed in the Isotope Cosmochemistry and Geochronology Laboratory at Arizona State University. The Sr, Ba, Nd, and Sm isotopic compositions of the samples and standards were measured on a ThermoScientific Triton thermal ionization mass spectrometer at Lawrence Livermore National Laboratory. Internal normalization was used to correct for instrumental mass bias for all isotope systems. All samples were run interspersed with terrestrial standards; the external reproducibility ($2 \times$ the SD, 2SD) of the standards was recorded as the lowest possible reported uncertainty. Isobaric interferences for each element were monitored and corrected for during runs of all samples and standards. Details for all elements measured are given in *SI Appendix*.

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