¹²⁹I and ²⁴⁷Cm in meteorites constrain the last astrophysical source of solar r-process elements

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The composition of the early Solar System can be inferred from meteorites. Many elements heavier than iron were formed by the rapid neutron capture process (r-process), but the astrophysical sources where this occurred remain poorly understood. We demonstrate that the near-identical half-lives (\approx 15.6 million years) of the radioactive r-process nuclei iodine-129 and curium-247 preserve their ratio, irrespective of the time between production and incorporation into the Solar System. We constrain the last r-process source by comparing the measured meteoritic ratio $^{129}I/^{247}Cm = 438 \pm 184$ with nucleosynthesis calculations based on neutron star merger and magneto-rotational supernova simulations. Moderately neutron-rich conditions, often found in merger disk ejecta simulations, are most consistent with the meteoritic value. Uncertain nuclear physics data limit our confidence in this conclusion.

he rapid neutron capture process (rprocess) is the source of half of the naturally occurring elements heavier than iron (1), including iodine, europium, gold, platinum, and the actinides. However, the astrophysical sites where r-process elements were synthesized and the physical conditions at these sites are not well constrained.

The gravitational wave event GW170817 (2), the identification of its electromagnetic counterpart, and the inference of lanthanide elements in the ejecta (3) have shown that neutron star mergers can synthesize at least some r-process elements. GW170817 provided only limited information on the nucleosynthesis process, as only one specific element (strontium) has been identified in its spectrum (4). More detailed isotopic information for r-process nucleosynthesis is recorded in the composition of the Solar System. Analysis of primitive meteorites has produced abundance determinations for all stable isotopes (5), whereas abundances derived from stellar spectra typically provide elemental abundances only.

The Solar System's stable isotopes include contributions from multiple nucleosynthetic events (supernovae, compact binary mergers, etc.) that occurred at any time between the birth of the Milky Way and the formation of the Sun. This evolution is difficult to model but can be simplified by considering radioactive isotopes with half-lives of several million years (Myr). Analysis of meteorites has shown that such isotopes were present at the formation time of the first solids [the

calcium-aluminum-rich inclusions (CAIs)] in the early Solar System (6). Because those radioactive isotopes have all decayed over the lifetime of the Solar System, their initial abundances are inferred from excesses of the daughter isotopes they decay into. Radioactive isotopes reflect a smaller number of nucleosynthesis events than stable isotopes. specifically the events that occurred shortly before the formation of the Sun. We consider the early Solar System abundances of two radioactive isotopes with half-lives of 15.7 and 15.6 Myr, respectively: ¹²⁹I and the heavier actinide isotope, ²⁴⁷Cm. We adopt abundances of these isotopes (Table 1) from previously published analyses of meteorites (7-9), where they are reported as ratios with reference isotopes 129I/127I and 247Cm/235U.

The process of comparing these isotopic ratios directly with predictions from simulations and determining the nucleosynthetic sources that enrich interstellar gas with heavy elements is highly uncertain. The abundance ratio ¹²⁹I/¹²⁷I has a stable isotope in the denominator, the abundance of which depends on the complete galactic enrichment history before the formation of the Solar System. This ratio is therefore affected by uncertainties in the star formation history, the amount of interstellar gas in the Milky Way, and the amount of ¹²⁷I removed from the interstellar gas by galactic outflows (10). The 247 Cm/ 235 U ratio is less affected by those uncertainties because ²³⁵U has a half-life of 704 Myr, which is short relative to the ~8 to 9 billion years of galactic enrichment before the formation of the Sun. The ²⁴⁷Cm/²³⁵U ratio is still affected by the uncertain time interval between the synthesis of these elements and their incorporation into the early Solar System. This delay is ~100 to 200 Myr for r-process isotopes (11), during which ²⁴⁷Cm and ²³⁵U decay exponentially. Because their half-lives differ by a factor of 50, the $^{247}\mathrm{Cm}/^{235}\mathrm{U}$ abundance ratio diverges from its original value before being locked into the Solar System.

Enrichment of the interstellar gas from which the Solar System formed was not continuous but stochastic (12). It is therefore unknown how many enrichment events are recorded in the isotopic ratios derived from meteorites. Because the radioactive abundances from each event decayed for an unknown amount of time, the relative contributions are even more uncertain.

Using the ¹²⁹I/²⁴⁷Cm abundance ratio by passes those uncertainties because of the combination of two properties. First, ¹²⁹I and ²⁴⁷Cm have the same half-life, within uncertainties, so their ratio is not strongly affected by decay over time. Second, both isotopes are short-lived compared with the average time elapsed between r-process events, so their ratio probably reflects only one event (supplementary

Table 1. Early Solar System isotopic ratios involving radioactive nuclei produced by the

r-process. Column 5 provides the early Solar System ratio of each isotope listed in column 1 relative to that in column 3, with the half-lives of these isotopes (28–30) given in columns 2 and 4, respectively. All uncertainties are 2σ (11).

Short-lived radionuclide	Half-life (Myr)	Reference isotope	Half-life (Myr)	Early Solar System ratio	References
¹²⁹	15.7 ± 0.8	¹²⁷	Stable	$(1.28 \pm 0.03) \times 10^{-4}$	(7)
²⁴⁷ Cm	15.6 ± 1.0	²³⁵ U	704 ± 2	$(5.6 \pm 0.3) \times 10^{-5}$	(8, 9)
129	15.7 ± 0.8	²⁴⁷ Cm	15.6 ± 1.0	438 ± 184	See text

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text). Figure 1 shows a simulation of how these isotope ratios vary over time. 129I/247Cm always stays close to its production ratio, whereas ¹²⁹I/ ¹²⁷I and ²⁴⁷Cm/²³⁵U vary by orders of magnitude. Different astrophysical sources could have synthesized a range of ¹²⁹I/²⁴⁷Cm abundance ratios throughout the history of the Galaxy, but only one event is likely recorded in meteorites for these isotopes. We determine the ¹²⁹I/²⁴⁷Cm ratio in the early Solar System (Table 1) using the reported ¹²⁹I/¹²⁷I and $^{247}\mathrm{Cm}/^{235}\mathrm{U}$ ratios together with the $^{127}\mathrm{I}/^{235}\mathrm{U}$ ratio of 189 (5). We find ${}^{129}\text{I}/{}^{247}\text{Cm} = 438 \pm 184$, and we interpret this value as reflecting the nucleosynthesis of the last r-process event that polluted the presolar nebula.

This value relies on our adoption of solar abundances commonly used in astronomy (5). However, alternative measurements have reported an iodine abundance that is an order of magnitude lower (13), which would affect our conclusions. Adopting the lower value would make iodine less abundant than neighboring isotopes. Our nucleosynthesis calculations (see below) do not predict this feature because they generally show smoother abundance trends between neighboring species, which is more consistent with the higher abundance measurement (5). The meteoritic measurements (13) could be affected by heterogeneities on scales larger than the samples that were analyzed (the nugget effect) and by possible losses of noble gases produced from halogens such as iodine through the irradiation technique adopted for the measurements (supplementary text). We therefore prefer to adopt the higher value of the iodine abundance (5)(supplementary text).

We performed theoretical nucleosynthesis calculations to determine the $^{129}\mathrm{I}/^{247}\mathrm{Cm}$ abundance ratios that would be produced in the physical conditions that occur in previous hydrodynamic simulations of potential r-process sites: neutron star-neutron star (NS-NS) mergers, neutron star-black hole (NS-BH) mergers, and core-collapse supernovae (SNe) driven by strong magnetic fields and fast rotation [magneto-rotational supernovae (MR SNe)] (14). In NS-NS and NS-BH mergers, matter is ejected in two wavs: (i) dynamical ejecta (15, 16) that are driven by tidal forces and shocks that occur promptly during the merger and (ii) disk ejecta (17) that are driven by heating that unbinds matter from the disk that forms around the compact central remnant left after the merger, which is either a neutron star or a black hole. Table S1 lists details of the seven simulations we considered. Because r-process nucleosynthesis predictions are affected by large uncertainties from nuclear physics (18-20), we repeated our calculations with three different sets of nuclear reaction rates and three different models for the distribution of fission fragments (11). This generated nine nucleosynthetic model predictions that were applied to each of the seven hydrodynamic simulations, for a total of 63 calculations shown in Fig. 2.

In Fig. 2, we compare our predicted ¹²⁹I/ ²⁴⁷Cm ratios using different nuclear physics input with the meteoritic ratio. The uncertainties on the meteoritic ratio include both the uncertainty in the derivation of the early Solar System ratio (Table 1) and the uncertainty in the half-lives of ¹²⁹I and ²⁴⁷Cm. We include the latter to account for the slight

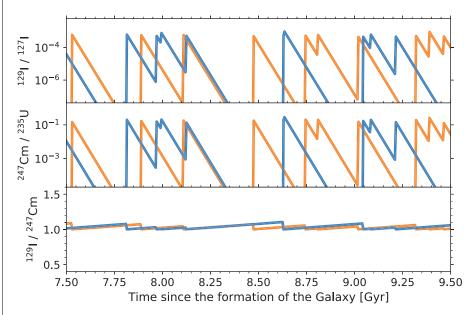


Fig. 1. Simulated evolution of the abundance ratios ¹²⁹I/¹²⁷I, ²⁴⁷Cm/²³⁵U, and ¹²⁹I/²⁴⁷Cm in a parcel of Milky Way interstellar gas. The time window shown encompasses the time when the Sun formed. Each peak is produced by an additional r-process event. The blue and orange lines show two arbitrary Monte Carlo realizations for the temporal distribution of those events (*27*). Each event is assumed to eject the same mass of ¹²⁹I, ¹²⁷I, ²⁴⁷Cm, and ²³⁵U, such that the production ratio is equal to 1 for all three isotopic ratios. Gyr, billion years.

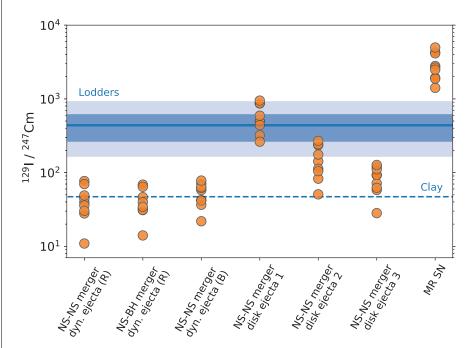


Fig. 2. ¹²⁹**I**/²⁴⁷**Cm abundance ratios predicted by theoretical r-process models.** The dots in each model denote the use of different nuclear physics inputs (*11*). The horizontal blue solid line shows the meteoritic ratio, and the shaded blue bands are its 1σ and 2σ uncertainties (*11*). The horizontal blue dashed line shows the meteoritic ratio when adopting alternative measurements (*13*). dyn., dynamical; R, Rosswog; B, Bovard.

ratio variation that could have occurred during the time elapsed between the last r-process event and the condensation of the first solids in the early Solar System (*II*). Because ¹²⁹I and ²⁴⁷Cm have substantially different atomic numbers, their relative abundances strongly depend on the physical conditions in which the r-process nucleosynthesis occurs. The predicted ratios shown in Fig. 2 vary by more than two orders of magnitude.

For the magneto-rotational supernova (MR SN) ejecta (14), the abundance ratio is always >1000 because most of the ejecta are not sufficiently neutron rich to produce enough actinides. Although other MR SN simulations may generate different results, models with alternative neutrino transport predict even lower production of actinides (21). MR SNe are expected to have occurred more often in the early Universe, because of higher stellar rotation (22), which makes MR SNe more likely to enrich very old stars than the Solar System. Collapsars are also a possible r-process site; these occur during the late evolution of some MR SNe, when a black hole surrounded by an accretion disk forms. However, their capacity to synthesize actinides (including ²⁴⁷Cm) is debated and ranges from substantial production (23) to no production (24, 25).

For the NS-NS and NS-BH merger simulations, dynamical ejecta are dominated by very neutron-rich conditions—producing more actinides (such as 247 Cm) relative to lighter nuclei (129 I, in this case)—compared with the other r-process scenarios (see also fig. S1). As a result, the dynamical ejecta 129 I/ 247 Cm ratios are all <100, which is below the 2σ uncertainty of the meteoritic ratio. Merger simulations predict the presence of very neutron-rich material (*15, 16*); however, the exact contribution of such conditions to the total ejecta is still unclear. Simulations of dynamical ejecta show a broad range of neutron richness (*26*).

The three NS-NS merger accretion-disk ejecta simulations give different results (Fig. 2). NS-NS disk 1 is consistent with the meteoritic value, NS-NS disk 2 partly overlaps with the 2σ uncertainty, whereas NS-NS disk 3 is below the 2σ uncertainty and therefore not compatible. Although these disk simulations represent a disk forming around an NS-NS remnant, NS-BH disk models can produce similar abundances (*17*).

We considered the combination of both dynamical and disk ejecta from a single binary merger (supplementary text). We found that the maximum contribution of dynamical ejecta is ~50% (in mass fraction) to remain within the 2σ uncertainty of the meteoritic ratio. ¹²⁹I and ²⁴⁷Cm in the early Solar System were likely synthesized by only one r-process event, but if two events contributed, the meteoritic ratio could be matched by a combination of dynamical ejecta with MR SN ejecta (see Fig. 2). However, such a mixture has an occurrence probability of ${<}10\%$ (supplementary text).

To test the sensitivity of these results to the input data, we performed 56 additional nucleosynthesis calculations on the dynamical ejecta (15) using a different nucleosynthesis code and a wider variety of input nuclear physics models (11). Most of these models predict ¹²⁹I/²⁴⁷Cm ratios <100 (tables S2 and S3), which is consistent with the results presented in Fig. 2. In 4 of the 56 cases, very neutron-rich dynamical ejecta reach the meteoritic ratio. The large range in predictions is due to the nuclear physics uncertainties. Our additional calculations support our conclusion that enrichment of the presolar nebula by very neutron-rich ejecta is often inconsistent with the meteoritic data. This result applies to the last r-process event that polluted the presolar nebula with radioactive isotopes, not to the collective contribution of all previous events that built up the stable r-process solar composition.

We have shown that the $^{129}I/^{247}$ Cm abundance ratio can constrain the ejecta composition of the last r-process event that polluted the presolar nebula. This ratio is highly sensitive to the physical conditions in which ^{129}I and 247 Cm were synthesized. Our results suggest that moderately neutron-rich conditions are generally most consistent with the meteoritic value. However, such conclusions are limited by solar abundance determinations and current uncertainties in the available hydrodynamical and nucleosynthesis models.

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used in PRISM. B.V. calculated the probability distributions shown in figs. S4 and S5. B.S. ran the calculations shown in fig. S3. A.A., R.S., M.P., and B.W. participated in the revising process. T.M.S. participated in the interpretation of meteoritic abundances and in the revising process. T.R. participated in the development of the nuclear reaction rates used in WINNET and in the revising process. M.L. calculated the early Solar System 1²⁹/²⁴/²⁴/Cm ratio shown in Table 1, helped develop the concept, and participated in the writing and revising process. **Competing interests:** We declare no conflicts of interest. **Data and materials availability:** The code used to calculate the isotopic ratios shown in Fig. 1 is available at https://github.com/AndresYague/ Stochastic_RadioNuclides (3J). The WINNET nucleosynthesis output and sampling code to reproduce Fig. 2 and figs. S1 and S2 are available on Zenodo (32). The code to reproduce fig. S3 is available at https://github.com/AndresYague/delta_stat_tau_ code/tree/v1.0.0 (33). The Monte Carlo code used to calculate the distributions shown in figs. S4 and S5 is available at https:// github.com/AndresYague/IodineCurium_project_distributions (34). The PRISM nucleosynthesis output necessary to reproduce tables S2 and S3 is available on Zenodo (35); this dataset is released under Los Alamos National Laboratory report no. LA-UR-21-20444. The Monte Carlo code to generate table S6 is available at https:// github.com/AndresYague/IodineCurium_project_oneEvent (36). The trajectories of the dynamical ejecta (R) simulations were taken from https://compact-merger.astro.su.se/downloads_fluid_ trajectories.html; the dynamical ejecta (B) were taken from (16); disk ejecta 1, 2, and 3 were taken from (17); and the MR SN ejecta were taken from (14). The WINNET code was developed by C. Winteler, F. Thielemann, O. Korobkin, M. Eichler, D. Martin, J. Bliss, M. Reichert, and A. Arcones; we do not have their permission to distribute it. The PRISM code is security restricted and unavailable for public release; contact M.R.M. for details.

SUPPLEMENTARY MATERIALS

science.sciencemag.org/content/371/6532/945/suppl/DC1 Materials and Methods Supplementary Text Figs. S1 to S5 Tables S1 to S6 References (*37–104*)

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¹²⁹I and ²⁴⁷Cm in meteorites constrain the last astrophysical source of solar r-process elements

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The origin of r-process elements

Theoretical models predict that the synthesis of heavy elements by the rapid neutron capture process (r-process) occurs in extreme astrophysical environments such as neutron star mergers or some types of supernovae. Testing those predictions by comparing them with the isotopic record has been difficult. Côté *et al.* examined two r-process isotopes, iodine-129 and curium-247, both of which have half-lives of 15.6 million years. Therefore, their ratio remains constant even long after the nucleosynthesis event. The ratio of those isotopes at the time of Solar System formation is recorded in meteorites. Comparing this value with nuclear astrophysics calculations shows that the most likely source was moderately neutron-rich material ejected from a binary neutron star merger.

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