LETTERS
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Short-lived ^{244}Pu points to compact binary mergers as sites for heavy r-process nucleosynthesis

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The origin of heavy elements produced through rapid neutron capture ('r-process') by seed nuclei is one of the current nucleosynthesis mysteries[1-3]. Core collapse supernovae (cc-SNe; ref. 4) and compact binary mergers are considered as possible sites[5-7]. The first produces small amounts of material at a high event rate whereas the latter produces large amounts in rare events. Radioactive elements with the right lifetime can break the degeneracy between high-rate/low-yield and low-rate/high-yield scenarios. Among radioactive elements, most interesting is ^{244}Pu (half-life of 81 million years), for which both the current accumulation of live ^{244}Pu particles accreted via interstellar particles in the Earth's deep-sea floor[8] and the Early Solar System (ESS) abundances have been measured[9]. Interestingly, the estimated ^{244}Pu abundance in the current interstellar medium inferred from deep-sea measurements is significantly lower than that corresponding to the ESS measurements. Here we show that both the current and ESS abundances of ^{244}Pu are naturally explained within the low-rate/high-yield scenario. The inferred event rate remarkably agrees with compact binary merger rates estimated from Galactic neutron star binaries[10] and from short gamma-ray bursts[11]. Furthermore, the ejected mass of r-process elements per event agrees with both theoretical[12-14] and observational[15-17] macronova/kilonova estimates.

Using the Solar abundance[19] as the mean value for stars in the Galactic disk, the total mass of heavy r-process elements ($A \geq 90$) in the Galaxy is $M_{\mathrm{tot},A\geq 90} \approx 5 \times 10^3 M_{\odot}$. These elements show a consistent abundance pattern in metal-poor stars[18], suggesting that they are produced in a single kind of event. The total mass yields a relation between the Galactic event rate, R, and the heavy r-process mass produced in each event $M_{\mathrm{ej},A\geq 90}$ (see Fig. 1):

$$\langle R\rangle \approx 50\,\mathrm{Myr}^{-1}\left(\frac{M_{\mathrm{ej},A\geq 90}}{0.01 M_{\odot}}\right)^{-1} \quad (1)$$

Here $\langle R\rangle$ is the rate averaged over the age of the Galaxy and it is not necessarily the same as the present-day event rate R_0. For sources related to the death of massive stars, the event rate should follow the star formation rate, which at present is lower than the average value[20]. For compact binary mergers, the event rate follows the star formation rate with some delay. The event rate of short gamma-ray bursts (SGRBs) (ref. 11) that probably arise from compact binary mergers[6], increases with the cosmological redshift z at least up to $z \approx 0.8$. In both cases, R_0 may be smaller than $\langle R\rangle$ by a factor of up to approximately five.

Using the total mass alone we cannot distinguish between the high-rate/low-yield and the low-rate/high-yield sources. Measurements of abundances of short-lived radioactive r-process nuclides can, however, remove this degeneracy, as these abundances reflect the r-process production history on timescales comparable to their lifetimes (modulo the Galactic mixing timescale). Among the various radioactive nuclides, ^{244}Pu seems most suitable: it is produced only via the r-process; the half-life of ^{244}Pu, 81 Million years (Myr), is sufficiently short compared to the Hubble time, although long enough to allow for significant accumulation; and both current and ESS ($\sim$4.6 Gyr before present; BP) ^{244}Pu abundance in the interstellar medium (ISM) have been measured.

The inner Solar System continuously accretes interstellar dust grains[21] containing recently produced live radioactive ^{244}Pu. Wallner *et al.*[8] (see also Paul *et al.*[22]) measured the accumulation of ^{244}Pu in a deep-sea crust sample during the past 25 Myr and estimated the ^{244}Pu flux on the Earth's orbit as $250^{+590}_{-205}\,\mathrm{cm}^{-2}\,\mathrm{Myr}^{-1}$, where the upper and lower values correspond to 2σ limits. The corresponding mean number density of ^{244}Pu in the ISM is $6 \times 10^{-17}\,\mathrm{cm}^{-3}$ and the 2σ upper limit is $2 \times 10^{-16}\,\mathrm{cm}^{-3}$ (see Supplementary Methods). These values are significantly lower than the number density in the ISM derived from the ESS ^{244}Pu abundance: $n_{\mathrm{Pu}} \approx ({}^{244}\mathrm{Pu}/{}^{238}\mathrm{U})_{\mathrm{ESS}} Y_{\mathrm{U,ESS}} n_{\mathrm{ISM}} \sim 6 \times 10^{-15}\,\mathrm{cm}^{-3}$. The relative abundance ratio of $({}^{244}\mathrm{Pu}/{}^{238}\mathrm{U})_{\mathrm{ESS}} \approx 0.008$ is estimated from fissiogenic xenon in ESS relics[9] (for example, chondritic meteorites). $Y_{\mathrm{U,\,ESS}} = 7.3 \times 10^{-13}$ is the number abundance of ^{238}U relative to hydrogen inferred from meteorites[23] (corrected from the present for ^{238}U decay) and n_{ISM} is the mean number density of the ISM, which is typically approximately $1\,\mathrm{cm}^{-3}$.

The abundance of a radioactive nuclide at a given location around the solar circle, $r_{\odot}$, depends on the event rate density, $\mathcal{R} \equiv \rho_*(r_{\odot}, 0)R/M_* \approx 0.0015 R\,\mathrm{Myr}^{-1}\,\mathrm{kpc}^{-3}$, where $\rho_*(r, z)$ is the stellar mass density in the disk, r and z are the Galactic radius and height above the Galactic plane, and M_* is the total stellar mass in the disk[24]. The abundance depends also on the mixing rate. The most relevant mixing process is turbulent diffusion (see Supplementary Methods) whose diffusion coefficient satisfies $D = \alpha v_{\mathrm{t}} H \approx \alpha\,\mathrm{kpc}^2/\mathrm{Gyr}\,(v_{\mathrm{t}}/7\,\mathrm{km\,s}^{-1})(H/0.2\,\mathrm{kpc})$, where v_{t} is the typical turbulence velocity of the ISM, H is the ISM scale height, and α is the mixing length parameter. Defining τ_{mix} as the mean time between injection events at a given location which satisfies $(4\pi\mathcal{R}\tau_{\mathrm{mix}}/3)^{-1/3} \equiv 2(D\tau_{\mathrm{mix}})^{1/2}$, we derive:

$$\tau_{\mathrm{mix}} \approx 300\,\mathrm{Myr}\,(R/10\,\mathrm{Myr})^{-2/5}(\alpha/0.1)^{-3/5} \times (v_{\mathrm{t}}/7\,\mathrm{km/s})^{-3/5}(H/0.2\,\mathrm{kpc})^{-3/5} \quad (2)$$

The median number density (the median rather than the average reflects the density that a typical observer measures) of a short-lived radioactive nuclide with a mean-life τ_i is:

$$\langle n_i\rangle_m \approx n_{\mathrm{eq},i} \exp\left(-\frac{\tau_{\mathrm{mix}}}{2\tau_i}\right) \quad (3)$$

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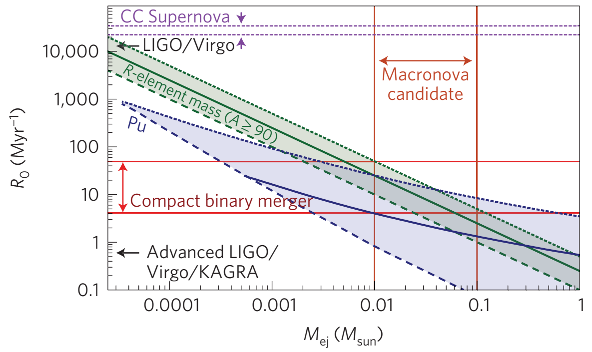

Figure 1 | The heavy r-process event rate versus the ejected mass. The diagonal green region expresses the degeneracy between high rate/low yield and low rate/high yield corresponding to the total mass of (stable) heavy ($A \geq 90$) r-process elements in the Galaxy, with $R_0 = \langle R \rangle$, $0.5\langle R \rangle$, and $0.2\langle R \rangle$ (see equation (1)). The allowed region inferred from the ^{244}Pu abundance in the deep-sea crust[8] and the ESS (refs 9,23) is shown as a blue band. The blue solid (dotted) line corresponds to the current ISM ^{244}Pu density being the median (2σ) value. The region above the dashed blue curve is the allowed region consistent with the ESS measurement (within 2σ fluctuations and taking into account that the rate at 4.6 Gyr BP can be higher than R_0 by up to a factor of five). The current event rate estimated from binary neutron stars[10] and SGRBs (ref. 11) are shown as the region between the horizontal red lines. For SGRBs, we take an (unknown) jet beaming factor in the range of 10–70. The region between the horizontal dotted purple lines corresponds to the cc-SNe event rate[29]. Macronova mass estimates[12–17] are between the vertical dark orange lines. The upper and lower horizontal arrows, respectively, show the LIGO/Virgo upper limit of the merger rate[30] and the expected capability of the advanced gravitational-wave detectors with 5 yr observations. The overlap region of the ^{244}Pu measurements and the total amount of heavy r-process elements is consistent with that of the compact binary merger scenario.

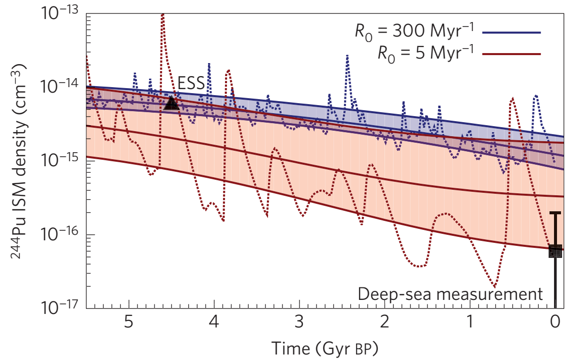

Figure 2 | Time evolution of ^{244}Pu number densities in the ISM on the solar circle. The solid red (blue) lines represent the median number density and $\pm 1\sigma$ fluctuations for $R_0 = 5$ Myr^{-1} ($R_0 = 300$ Myr^{-1}). The lower square with an error bar shows the ^{244}Pu density with 2σ limits inferred from the deep-sea measurement[8]. The triangle at 4.6 Gyr BP shows the value at the time of the ESS (refs 9,23). The production rate of ^{244}Pu follows the time evolution of the SGRB rate[11] for $R_0 = 5$ Myr^{-1} and the cosmic star formation history[20] for $R_0 = 300$ Myr^{-1}. Also shown is a Monte Carlo simulation the time sequence of ^{244}Pu number densities at a given location on the solar circle for $R_0 = 5$ Myr^{-1} (dotted red line) and $R_0 = 300$ Myr^{-1} (dotted blue line).

where $n_{eq,i} \approx N_i \mathcal{R} \tau_i$ is the equilibrium value and N_i is the total number of the nuclide i ejected by each event. For $\tau_i \gg \tau_{mix}$, a typical observer measures $n_{eq,i}$. For $\tau_i \ll \tau_{mix}$, a typical observer measures a number density much lower than $n_{eq,i}$ and one needs a larger yield to reach an observed value, interpreted here as the median number density. Figure 1 depicts the needed rate and yield so that the current ^{244}Pu number density is the median value for typical values of α, v_t and H (see equation (2)). This relation (blue area in Fig. 1) becomes flatter than $R \propto M_{ej}^{-1}$ (equation (1), green band in Fig. 1) for decreasing event rates breaking the rate-yield degeneracy.

To take into account the large fluctuation in the measured number density averaged over timescales shorter than τ_{mix}, we simulate the history of the ^{244}Pu abundance in the ISM around the solar circle over the past 7 Gyr. We take into account the radioactive decay, the turbulent diffusion process and the time evolution of the production rate. We consider here a characteristic low-rate/high-yield case of $R_0 = 5$ Myr^{-1} following the SGRB rate[11] evolution and a high-rate/low-yield case of $R_0 = 300$ Myr^{-1} following the cosmic star formation history[20] (Fig. 2). As expected the fluctuations of the low-rate/high-yield case are much larger than those of the high-rate/low-yield case. For both cases, the estimated range of number densities around 4.6 Gyr BP are consistent with the ESS values and they decrease with time following the decreasing event rate. Whereas for $R_0 = 5$ Myr^{-1} the simulated values are also consistent with the current deep-sea measurements, for $R_0 = 300$ Myr^{-1} the decline is insufficient even when taking the fluctuations into account. Figure 1 depicts upper and lower bounds on the event rate which consistently explain the ^{244}Pu abundance of the ESS and the current ISM. The sources must satisfy $R_0 \leq 90$ Myr^{-1} and $M_{ej} \geq 0.001$ $M_\odot$. Although these limits vary slightly with different assumed parameters (see Supplementary Methods), the qualitative result that we reach is robust and independent of these choices. We conclude that unless an unidentified process suppresses the present amount of ^{244}Pu that reaches Earth, the heavy r-process sources are dominantly low-rate/high-yield ones. The rarity of r-processing events also consistently explains the large scatter in r-elements/Fe abundance of metal-poor stars[25–28].

These results are compared with astronomical observations concerning the possible sources. The low rate clearly rules out cc-SNe. The current ^{244}Pu abundance should be larger by a factor of 5–100 to be compatible with a dominant cc-SNe source (see Supplementary Methods). Turning to compact binary mergers, Fig. 1 depicts also the merger rate estimated from known Galactic binary neutron stars[10] and from the current SGRB rate[11], as well as the ejected mass of r-process elements estimated from macronova candidates associated with GRB 130603B (refs 15,16) and with GRB 060614 (ref. 17). Remarkably, the rates and masses estimated here are fully consistent with those observations. In fact most of the overlap between the allowed ^{244}Pu region and the overall r-process production range is just in this part of the astrophysical parameter phase space describing compact binary mergers and macronova ejection estimates. This result is independent of the choice of the efficiency and diffusion parameters.

Compact binary mergers, which we can conclude are the sources of heavy r-process nucleosynthesis, are also the prime candidates of sources for the gravitational-wave detectors, advanced LIGO/Virgo and KAGRA. Our estimates provide an upper limit to the expected detection rate (assuming a detection horizon distance of 200 Mpc): $R_{GW} \leq 30$ yr^{-1}. The estimated ejected mass in each event is significant, implying that macronovae[31–33] and radio flares[34] associated with the gravitational-wave merger events will be detectable with follow-up observations.

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Author contributions
K.H. performed the calculations and produced the figures of the paper. T.P. contributed to all aspects of astrophysical discussions. M.P. contributed to the interpretation and discussion of the experimental data of the ^{244}Pu abundance. All authors contributed to the writing of the paper.

Additional information
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Competing financial interests
The authors declare no competing financial interests.