Future radioisotope measurements to clarify the origin of deep-ocean $^{244}\text{Pu}$

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$^{244}\text{Pu}$ has been discovered in deep-ocean deposits spanning the past 10 Myr, a period that includes two $^{60}\text{Fe}$ pulses from nearby supernovae. $^{244}\text{Pu}$ is among the heaviest $r$-process products, and we consider whether the $^{244}\text{Pu}$ was created in the supernovae, which is disfavored by model calculations, or in an earlier kilonova that seeded $^{244}\text{Pu}$ in the nearby interstellar medium, which was subsequently swept up by the supernova debris. We propose probing these possibilities by measuring other $r$-process radioisotopes such as $^{129}\text{I}$ and $^{182}\text{Hf}$ in deep-ocean deposits and in lunar regolith.

Measurements of live radioactive isotopes can provide insights into recent astrophysical explosions such as core-collapse supernovae (SNe) within $\mathcal{O}(100)$ pc of Earth [1] that are expected to occur every few million years, clarifying the possibility of rarer events within $\mathcal{O}(10)$ pc that might have caused mass extinctions in the past [2, 3]. Many experiments over the past two decades have detected pulses of live $^{60}\text{Fe}$ in deep-ocean deposits [4–9] from between 2 and 3 My ago (Mya), very likely due to a nearby core-collapse SN. There have also been hints of earlier deep-ocean $^{60}\text{Fe}$ deposition, as well as measurements of $^{60}\text{Fe}$ in the lunar regolith [10], in cosmic rays [11] and in Antarctic snow [12].

In addition to these measurements, there have been some tantalizing hints of deep-ocean $^{244}\text{Pu}$ [13–16]. These are interesting because the $^{244}\text{Pu}$ would have been produced by the astrophysical $r$-process, whose likely sites include neutron-star mergers (kilonovae, KNe), and possibly some atypical SNe. Motivated by these hints, in a recent paper we studied possible signatures of SN and KN $r$-process events, analyzing the potential implications of $^{244}\text{Pu}$ detection, estimating the strengths of other $r$-process radioisotope signatures, and discussing how they could help distinguish between potential sites [17].

A major advance in studies of live astrophysical radioisotopes has recently been made by Wallner et al. [18], who have discovered a second $^{60}\text{Fe}$ pulse $\sim 7$ Mya, as well as $^{244}\text{Pu}$ in deep-ocean ferromagnetic crusts from periods that include both the $^{60}\text{Fe}$ pulses. These results deepen greatly our picture of recent near-Earth explo-
sions, and widen dramatically the scope of their implications. Broadly, the second $^{60}\text{Fe}$ pulse shows that there were multiple explosions, as one would expect from massive stars that are highly clustered [19]. And the detection of $^{244}\text{Pu}$ not only is the second firmly detected radioisotope in this epoch, but also demands an $r$-process source and so probes the astrophysical site of the $r$ process. In this paper we study the interpretation and potential implications of these new experimental results.

As has been shown in Fry et al. [20], ordinary non-$r$-process nucleosynthesis in core-collapse SNe provides the only plausible source of $^{60}\text{Fe}$ observed in the two pulses. Wallner et al. [18] concur, making this a starting point of their analysis. The question then becomes: could either or both of these SNe also have produced the $^{244}\text{Pu}$, or is a separate event required, presumably a KN?

The two $^{60}\text{Fe}$ pulses require two recent SNe. The $^{244}\text{Pu}$ data were not sampled as finely in time as the $^{60}\text{Fe}$ data: $^{244}\text{Pu}$ was measured in three separate time windows, including a surface layer that probes anthropogenic contamination. The two deeper layers each overlap with a $^{60}\text{Fe}$ pulse, as indicated by the yellow band in Fig. 1. The data show $^{60}\text{Fe}$ to be much more abundant than $^{244}\text{Pu}$ in both pulses.

SNe may produce the $r$-process via a $\nu$-driven wind or via magnetohydrodynamic (MHD) effects, but both struggle to make actinides, see [17] and references therein. If SNe are confirmed as robust sources of actinides such as $^{244}\text{Pu}$, the available models must have major omis-
sions. We thus discussed in Wang et al. [17] a modified
neutrino wind scenario forced to produce actinides and a high magnetic field MHD model, denoted by $\nu^*$ (SA) and SB, respectively, which we constrained using data on the metal-poor star HD160617. We show in Fig. 1 results from these models, both without and including ordinary (non-$\nu$-process) SN $^{60}$Fe production. Our calculations are made using the nuclear reaction network code Portable Routines for Integrated nucleoSynthesis Modeling (PRISM) [21, 22], as implemented in Wang et al. [17], with baseline nuclear data from [23] and [24] (FRDM+QRPA), and variations in the masses [25] (HFB), $\beta$-decay rates [26] (MKT), and fission yields [27]. The non-$\nu$-process SN $^{60}$Fe yields are for an explosion at 100 pc with $M_{ej,60} \sim 10^{-4.5} M_\odot$ with an uncertainty discussed in the Supplemental Materials.

Neutron star mergers that lead to KN explosions are much rarer than SNe, but estimates of the KN rate in the Galaxy are compatible with a KN explosion $\mathcal{O}(300)$ pc away that occurred $\mathcal{O}(30)$ Mya. Accordingly, we also show in Fig. 1 results from two scenarios invoking a KN explosion 10 or 20 Mya, one a combination of calculations of dynamical ejecta and a disk $\nu^*$-driven wind (KA) constrained to fit data on HD160617, and the other a modified scenario (KB) that fits data on the actinide-boost star J0954+5246: both models are described in Wang et al. [17]. The KN $^{60}$Fe/$^{244}$Pu ratios span a large range $^{60}$Fe$/^{244}$Pu$_{KN} \sim 10^{-5}$ to $10^{-2}$ when accounting for model uncertainties, but in the absence of an additional SN $^{60}$Fe source $^{244}$Pu is orders of magnitude more abundant than $^{60}$Fe in both models. This is because, whereas SNe expel $^{60}$Fe produced in multiple sites within the event and its progenitor star, the outflows from a neutron star merger are expected to be sufficiently neutron-rich to progress robustly beyond the iron peak in the bulk of the ejecta.

We show in Fig 2 the uncertainties in these calculations found [17] using the nuclear data variations described above. We see again that either of the SN models SA or SB could accommodate the (similar) $^{60}$Fe/$^{244}$Pu ratios reported by [18] in the periods around 3 and 7 Mya. On the other hand, both the KN models KA and KB still predict much smaller $^{60}$Fe/$^{244}$Pu ratios, even when the uncertainties are taken into account. We therefore conclude that the $^{60}$Fe pulses and $^{244}$Pu detection cannot be due to KN explosions alone, at least as described by the models considered here.

We consider first the data of Wallner et al. [18] on the $^{60}$Fe pulse from $\sim 3$ Mya. The timing of this signal is consistent with that measured previously in $^{60}$Fe deposits in deep-ocean sediments and crusts [4–9], though this peak is somewhat broader. A model in which $^{60}$Fe from a SN 100 Mpc away is transported to Earth in dust via ‘pinball’ trajectories that are deflected and trapped by a magnetic field within the SN remnant is compatible with a pulse of the observed size and duration $\sim 1$ Myr [28], and the pulse width indicated by the Wallner et al. [18] measurements could also reflect smearing in the crust they study. Accordingly, we assume that this pulse was produced by a single SN, and assume that the $^{244}$Pu from $\leq 4.57$ Mya measured by [18] is associated with this SN. We emphasize that observations with finer timing resolution would be needed to confirm this association, but note that many of our comments below would apply also if it were due to two or more SNe.

As discussed above, the additional $^{60}$Fe peak discovered by Wallner et al. [18], see also Fig. 1 of Fitoussi et al. [6], is likely due to another SN that occurred $\sim 7$ Mya, also some $\sim 100$ pc away. We assume that all the $^{244}$Pu from 4.57 to 9 Mya measured by Wallner et al. [18] is associated with this SN explosion, while emphasizing that observations with finer timing resolution would be needed to confirm this association. Under this assumption, the $^{244}$Pu/$^{60}$Fe ratios in the ejecta of the two SNe $\sim 3$ and $\sim 7$ Mya are comparable within a factor $\sim 2$ and indistinguishable in Fig. 1.

This is intriguing, since simulations indicate that only very specific types of SN can make such $^{244}$Pu [17], in which case seeing two of them looks like a remarkable coincidence. If such an interpretation were correct, it would suggest not only that many or most SNe are $\nu$-process sites, but also that their production extends all the way to the actinides. If this could be established, standard $\nu$-driven wind and MHD models must have major omissions. However, actinide production is possible in the forced neutrino wind or MHD models $\nu^*$ (SA) and SB discussed in Wang et al. [17].

As seen in Fig. 1, the artificially-enhanced SA model
overproduces $^{244}\text{Pu}$ relative to $^{60}\text{Fe}$! This could be brought into line with the data either by (1) including non-$r$-process $^{60}\text{Fe}$ production, or (2) with a smaller adjustment of the neutrino-driven wind, though this would still be incompatible with state-of-the-art SN models. The $^{60}\text{Fe}/^{244}\text{Pu}$ ratio from the MHD SN model SB is compatible with the data, with or without non-$r$-process $^{60}\text{Fe}$ production.

However, serious potential issues for scenarios with actinide production in many or most SNe are provided by measurements of the $r$-process abundances in metal-poor stars. (a) It is known that $r$-process/Fe ratios (estimated using Eu/Fe as a proxy) vary wildly, with most stars showing low values and only a minority showing high values. The obvious interpretation is that Fe and $r$-process production are decoupled. If SNe do indeed make the $r$-process, one possibility would be that (core-collapse) supernova Fe production is highly variable. However, there are observational constraints on this from observations of SN light curves powered by $^{56}\text{Ni}$ decay, so it seems more likely that the variations in $r$-process/Fe ratios are due to variations in $r$-process production. Another issue is that (b) searches for $r$-process species in metal-poor dwarf galaxies found them only in $\sim 10\%$. This strongly suggests that $r$-process events are much rarer than SNe. An alternative hypothesis is that the $r$-process material is ejected preferentially from the dwarf galaxies, e.g., in jets, but in this case jets would have to be features of most SNe, which is not supported by observations.

Motivated by these considerations, we proposed in Wang et al. [17] that $^{244}\text{Pu}$ signals could arise via a two-step process in which material deposited previously in the interstellar medium (ISM) by an earlier KN was then swept up by subsequent SN explosions [29]. This hypothesis is consistent with the data shown in Fig. 1, and could explain naturally the similarity between the $^{244}\text{Pu}/^{60}\text{Fe}$ ratios in the periods covering the two $^{60}\text{Fe}$ pulses found by Wallner et al. [18] (see the Suppemental Material).

Any $r$-process mechanism that produces $^{244}\text{Pu}$ also produces many other radioisotopes, not only other actinides such as $^{230}\text{U}$, $^{237}\text{Np}$, and $^{247}\text{Cm}$, but also many other radioisotopes with masses intermediate between $^{244}\text{Pu}$ and $^{60}\text{Fe}$. Hence their abundances would in general exhibit pulses coincident with the two SN $^{60}\text{Fe}$ pulses. This feature would be independent of the $r$-process location, whether a recent, nearby SN or an earlier, more distant KN. However, the relative abundances of the peaks of different $r$-process isotopes would be affected by their lifetimes, which would help distinguish scenarios in which the $r$-process occurred at different times in the past.

We have calculated the relative abundances of live $r$-process radioisotopes produced by the forced $r$-wind (SA) and the MHD (SB) models for the SNe that occurred 3 Mya and 7 Mya discussed above, as well as the two scenarios for a KN explosion 10 or 20 Mya. Figure 3 compares the yields of selected live $r$-process radioisotopes predicted by our SN and KN models with direct deposition (one-step), and calculations for additional live $r$-process radioisotopes are tabulated in the Supplemental Material. We see that, if the $^{244}\text{Pu}$ measured by Wallner et al. [18] was produced by a SN, one could hope to see accompanying $^{93}\text{Zr}$, $^{129}\text{I}$ and $^{182}\text{Hf}$.

Also shown in Fig. 3 are the $^{93}\text{Zr}$, $^{129}\text{I}$ and $^{182}\text{Hf}/^{244}\text{Pu}$
ratios calculated in the two-step KN scenarios KA and KB, assuming that the KN occurred 10 Mya followed by a non-ν-process SN 3 Mya. We see that in both models and after both time lapses the \(^{129}\text{I}/^{244}\text{Pu}\) ratio exceeds unity, whereas the \(^{182}\text{Hf}/^{244}\text{Pu}\) ratios is smaller. The non-ν-process SN \(^{53}\text{Zr}\) yields are \(M_{\text{ej},53} \sim 10^{-7.7} M_\odot\) with an uncertainty discussed in the Supplemental Material.

It is a common feature of all the SN and KN models studied above that the best prospects for discovering a second live \(\nu\)-process radioisotope (in addition to \(^{244}\text{Pu}\)) may be offered by \(^{129}\text{I}\). The \(^{129}\text{I}/^{244}\text{Pu}\) ratio calculated in the two-step KN scenarios KA and KB, assuming that the KN occurred 10 Mya followed by a non-\(\nu\)-process SN 3 Mya. We see that in both models and after both time lapses the \(^{129}\text{I}/^{244}\text{Pu}\) ratio exceeds unity, whereas the \(^{182}\text{Hf}/^{244}\text{Pu}\) ratios is smaller. The non-\(\nu\)-process SN \(^{53}\text{Zr}\) yields are \(M_{\text{ej},53} \sim 10^{-7.7} M_\odot\) with an uncertainty discussed in the Supplemental Material.

The relatively short \(^{244}\text{Pu}\) half-life of 2.6 My would make searches for earlier \(^{244}\text{Pu}\) pulses even more challenging. On the other hand, indirect evidence for earlier SNe could come from pulses of swept-up \(^{244}\text{Pu}\) in earlier deep-ocean deposits, in view of its much longer half-life \(\sim 80\) My. Ref. [16] reported the results of a search for \(^{244}\text{Pu}\) extending over the past 25 My, finding one event from \(> 12\) Mya. This event might just be background, but if not would correspond to a rate of deposition similar to that between 5 and 12 Mya. The more sensitive \(^{244}\text{Pu}\) results of Wallner et al. [18] extend back to 9 Mya, and it would clearly be interesting to extend the search for an earlier \(^{244}\text{Pu}\) signal and any possible time structure. A prime \(\nu\)-process candidate for corroborating any another \(^{244}\text{Pu}\) peak would again be \(^{129}\text{I}\), in view of the production rates found in both SN and KN models and its favourable half-life \(\sim 16\) My.

We close with some remarks about searches for live \(\nu\)-process radioisotopes in samples of lunar regolith. We recall that Finiani et al. [10] have reported the discovery of \(^{60}\text{Fe}\) in several Apollo samples. The data of Wallner et al. [18] suggest that this \(^{60}\text{Fe}\) is likely to have originated from a combination of the \(^{60}\text{Fe}\) pulses from 3 and 7 Mya, mainly the more recent pulse, in view of its greater size and younger age. Confirmation of this \(^{60}\text{Fe}\) signal, e.g., in the sample returned recently by the Chang’e-5 mission [36] or that from a future Artemis mission [37], would require analyzing a modest sample of \(\lesssim 100\) mg of lunar material [38]. The relative abundance of \(^{244}\text{Pu}\) reported by [18] is \(\sim 5 \times 10^{-5}\), suggesting that several kg of lunar regolith might be needed to discover a \(^{244}\text{Pu}\) signal. On the other hand, a much smaller sample might be sufficient to detect an \(^{129}\text{I}\) signal, since the \(^{129}\text{I}/^{244}\text{Pu}\) ratio is predicted to lie in the range \(\mathcal{O}(10)\) (for the KN models) through \(\mathcal{O}(100)\) (for SN model SA) to \(\mathcal{O}(10^6)\) (for SN model SB).

The AMS sensitivity for \(^{129}\text{I}\) already reaches the needed level, but the challenge is to avoid contamination from anthropogenic sources. \(^{129}\text{I}\) has already been measured in a Fe-Mn crust [39], showing a dropoff with depth consistent with a background source such as natural uranium fission. The Fe-Mn crust is not independently dated, but the abundance levels appear inconsistent with the SB
model while allowing room for some SA models and the KA and KB models. We note that Nishizumi et al. [40] used accelerator mass spectrometry to measure $^{129}I$ in the lunar regolith, and Nishizumi et al. [41] measured it in lunar rock, finding very similar abundances. These data foreshadow the power of new radioisotope measurements on both terrestrial and lunar samples, particularly $^{129}I$ and $^{182}Hf$, which can probe the nature of the recent explosions and of the r-process generally.

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The standard neutron capture mechanism. If the $^{244}\text{Pu}$ was produced by a KN 10 or 20 Mya, the best prospects for detection are for $^{129}\text{I}$, followed by $^{236}\text{U}$. There may also be some prospects for detection of $^{107}\text{Pd}$, for $^{182}\text{Hf}$ in model KA, and for $^{93}\text{Zr}$ in model KA if the explosion occurred 10 Mya.

In the main text and in Figure 3 we highlight representative isotopes in each of the three regions of the r-process abundance pattern: namely $^{93}\text{Zr}$, $^{129}\text{I}$, and $^{182}\text{Hf}$. We anticipate $^{182}\text{Hf}$ to be a clear marker of prompt supernova production, as it is present in potentially detectable levels for the SA and SB models but not in the two-step kilonova scenarios. $^{93}\text{Zr}$ can be produced in an alpha-rich freezeout of mildly neutron-rich supernova ejecta without an accompanying main r-process, so its detection could be a probe of this additional nucleosynthetic source. Our predictions show $^{129}\text{I}$ should be detectable alongside $^{244}\text{Pu}$ in any scenario, with the measured ratio offering possibly the strongest evidence to discriminate between scenarios. The other radioisotopes in the Table could provide similar insights into the r-process, if they could be measured.

Our predictions for radioisotope ratios to $^{244}\text{Pu}$ in Figs. 1 and 3 include results that combine r-process yields with those from ordinary (non-r-process) supernova explosions. This requires absolute mass yields (not just ratios) of $^{244}\text{Pu}$ and $^{60}\text{Fe}$, which are implemented as follows. We adopt the [17] estimate for the total kilonova r-process yields: $M_{\text{ej},r}(\text{KA, KB}) = (1.76 \times 10^{-2}, 7.00 \times 10^{-3}) M_\odot$. The $^{244}\text{Pu}$ mass yield is then $M_{\text{ej},244} = A_{244}Y_{244}M_{\text{ej},r}$.

Gamma-ray line observations give an observational indication of the mean $^{60}\text{Fe}$ yield. Given a Galactic steady-state $^{60}\text{Fe}$ mass $M_{60,ss} = 2.85 M_\odot$ [42], and a core-collapse supernova rate $\dot{R}_{\text{SN}} = 1.7 \text{events/century}$ [43], the mean $^{60}\text{Fe}$ yield is $M_{\text{ej},60} = M_{60,ss}/\dot{R}_{\text{SN}} = 4.5 \times 10^{-5} M_\odot$ where $\dot{R}_{\text{SN}}$ is the $^{60}\text{Fe}$ lifetime; the uncertainty in this mean yield is at least a factor of 2. Supernova nucleosynthesis calculations suggest that $^{60}\text{Fe}$ yields span a wide range, varying both sensitively and non-monotonically with progenitor mass. Yields in ref. [44] lie in the range $(4 \times 10^{-6}, 3 \times 10^{-4}) M_\odot$, which includes the calculations of [45] and model w of [46]. We thus adopt a $^{60}\text{Fe}$ yield of $M_{\text{ej},60} = 10^{-4.5 \pm 1} M_\odot$. Similarly, the yields of $^{93}\text{Zr}$ from ordinary (non-r-process) supernova explosions are in the range $(1.4 \times 10^{-9}, 2.4 \times 10^{-7}) M_\odot$ from [45] and model w of [46], thus a $^{93}\text{Zr}$ yield of $M_{\text{ej},93} = 10^{-7.5 \pm 1} M_\odot$ is adopted.

The measured $^{60}\text{Fe}/^{244}\text{Pu} = F_{60}/F_{244}$ ratio reflects the fluence ratio. For $^{244}\text{Pu}$ we use the observed interstellar $^{244}\text{Pu}$ flux to determine the fluence $F_{244} = \Phi_{\text{interstellar}} \Delta t$, where $\Delta t$ is the timespan of the mean...
TABLE I. r-process isotope ratios in forced SN models for explosions 3/7 Mya, corresponding to the known $^{60}$Fe pulses, and in KN models for explosions 10/20 Mya, bracketing the formation of the Local Bubble.

| Radioisotope   | Supernova Models | Kilonova Models |
|---------------|-----------------|----------------|
|               | SA 3 Mya 7 Mya  | KA 10 Mya 20 Mya |
|               | SB 3 Mya 7 Mya  | KB 10 Mya 20 Mya |
| $^{60}$Fe/$^{244}$Pu | 9.2 x 10^{-2} 3.2 x 10^{-2} | 3.7 x 10^{-4} 3.0 x 10^{-5} |
|               | 5.3 x 10^{3} 1.8 x 10^{3} | 1.7 x 10^{-6} 1.4 x 10^{-7} |
| $^{93}$Zr/$^{244}$Pu | 5.2 0.93 | 0.24 3.6 x 10^{-3} |
|               | 8.2 x 10^{4} 1.5 x 10^{4} | 7.7 x 10^{-3} 1.2 x 10^{-4} |
| $^{107}$Pd/$^{244}$Pu | 52 35 | 3.7 1.4 |
|               | 1.3 x 10^{5} 8.6 x 10^{4} | 0.34 0.13 |
| $^{129}$I/$^{244}$Pu | 3.2 x 10^{2} 2.8 x 10^{2} | 69 49 |
|               | 1.7 x 10^{6} 1.5 x 10^{6} | 14 10 |
| $^{135}$Cs/$^{244}$Pu | 5.4 0.68 | 8.7 x 10^{-3} 5.5 x 10^{-5} |
|               | 1.2 x 10^{5} 1.5 x 10^{4} | 3.7 x 10^{-2} 2.4 x 10^{-4} |
| $^{182}$Hf/$^{244}$Pu | 3.1 2.3 | 0.43 0.22 |
|               | 4.4 x 10^{3} 3.3 x 10^{3} | 6.9 x 10^{-2} 3.5 x 10^{-2} |
| $^{236}$U/$^{244}$Pu | 1.8 1.7 | 1.8 1.5 |
|               | 9.5 8.7 | 1.0 0.92 |
| $^{237}$Np/$^{244}$Pu | 0.66 0.18 | 8.2 x 10^{-2} 3.7 x 10^{-3} |
|               | 1.6 0.43 | 5.6 x 10^{-2} 2.5 x 10^{-3} |
| $^{247}$Cm/$^{244}$Pu | 0.50 0.43 | 0.38 0.27 |
|               | 0.45 0.39 | 0.35 0.25 |

The r-process contribution to $^{60}$Fe then follows as $F_{60,r} = f_{Fe}/f_{Pu} (^{60}$Fe/$^{244}$Pu)$_rF_{244}$, where ($^{60}$Fe/$^{244}$Pu)$_r$ is the model ratio, and we take the ratio of dust fractions to be $f_{Fe}/f_{Pu} = 1$. For the ordinary (non-r-process) supernova $^{60}$Fe fluence, we use $F_{non-r,60} = f_{Fe}M_{ej,60}/16\pi A_{60}m_{d}r_{SN}^2$, where we take the dust fraction $f_{Fe} = 0.1$, and use $r_{SN} = 100$ pc.