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Limits on supernova-associated 60 Fe/ 26 Al nucleosynthesis ratios from AMS measurements of deep-sea sediments

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We searched for the presence of 26 Al in deep-sea sediments as a signature of supernova influx. Our data show an exponential dependence of 26 Al with the sample age that is fully compatible with radioactive decay of terrigenic 26 Al. The same set of samples demonstrated a clear supernova 60 Fe signal between 1.7 and 3.2 Myr ago. Combining our 26 Al data with the recently reported 60 Fe data results in a lower limit of $0.18^{+0.15}_{-0.08}$ for the local interstellar 60 Fe/ 26 Al isotope ratio. It compares to most of the ratios deduced from nucleosynthesis models and is within the range of the observed average galactic 60 Fe/ 26 Al flux ratio of (0.15 ± 0.05) .

The radionuclides ²⁶Al ($t_{1/2}=0.717\pm0.017$ Myr [1]) ³⁵ and ⁶⁰Fe ($t_{1/2}=2.61\pm0.04$ Myr [2–4]) are key isotopes for ³⁶ understanding nucleosynthesis in our galaxy. Both were ³⁷ present in the early solar system, as evidenced by an excess of their decay-products in meteorites [5, 6]. Today, ³⁹ the decay of live ²⁶Al and ⁶⁰Fe is observed in the interstellar medium (ISM) through their associated characteristic ⁴¹ γ -rays [7, 8].

Significant amounts (3-10%) of ²⁶Al and ⁶⁰Fe are ⁴³ 9 freshly synthesized and ejected into the ISM by (su-44 10 per) asymptotic giant branch – (S)AGB – stars in the 45 11 mass range of \sim 5-9 M_{\odot} [9, 10]. However, the major frac- ⁴⁶ 12 tion is thought to be released by massive stars $(\geq 9 M_{\odot})_{47}$ 13 that explode as core-collapse supernovae (CCSNe) $[11-_{48}]$ 14 13]. Electron-capture (EC)SNe (\sim 7-11 M_{\odot}) produce 49 15 ⁶⁰Fe, but negligible ²⁶Al during explosive nucleosynthesis 50 16 [10, 14]. Additionally, stellar winds of Wolf-Rayet (WR) 51 17 stars with masses $>40 \,\mathrm{M}_{\odot}$, which also end their lives as $_{52}$ 18 SNe, have been proposed as major sources for the galac- 53 19 tic 26 Al inventory [15, 16], while they are not believed to $_{54}$ 20 contribute significant amounts of ⁶⁰Fe [13, 17]. Hence, ⁵⁵ 21 the observed ISM distribution of 26 Al and 60 Fe combines ${}_{56}$ 22 a mixture of different stellar sources, with a galactic av- 57 23 eraged ⁶⁰Fe/²⁶Al γ -ray flux ratio of (0.15 \pm 0.05) [8]. 24 58

Freshly produced radionuclides from supernova (SN) 59 25 explosions can be transported over large interstellar dis- 60 26 tances. Material ejected from nearby SNe can enter our 61 27 solar system and cross the Earth's orbit, potentially leav- 62 28 ing traces of the ejecta in terrestrial archives [18, 19]. 63 29 Indeed, the radionuclide ⁶⁰Fe has been identified in ter- 64 30 restrial [20–24] and lunar samples [25]. The detection of 65 31 SN-associated ⁶⁰Fe deposited about 2-3 Myr ago provides ⁶⁶ 32 an opportunity to determine the specific SN-associated 67 33 ²⁶Al/⁶⁰Fe ratio, disentangling it from the observed galac- 68 34

tic average ratio.

Here, we present for the first time a detailed ²⁶Al time profile and combine this data with the previously measured SN-associated ⁶⁰Fe data from the same deep-sea sediments [22] to derive the ⁶⁰Fe/²⁶Al ratio associated with the recent and local SN events. First, we estimate whether SN-associated ²⁶Al is within the detection limit of accelerator mass spectrometry (AMS) using an ⁶⁰Fe/²⁶Al isotope ratio of 0.5. Furthermore, we compare our derived SN-associated ⁶⁰Fe/²⁶Al ratio to SN nucleosynthesis models and to the observed galactic averaged ⁶⁰Fe/²⁶Al γ -ray flux ratio.

We have quantified the 26 Al content of a total of 83 samples from four *Eltanin* cores from the Indian Ocean, located ~1000 km south-west of Australia; ELT 45-16, ELT 45-21, ELT 49-53, and ELT 50-02 [26, 27]. The cores were recovered in the years 1970 and 1971 from depths of ~4300 m below the sea surface at locations of 35°0.7'S - 39°58'S and 100°02'E - 104°56'E. The largest set of samples was taken from ELT 45-21 (28 samples) from depths of 398-697 cm below the ocean floor and from ELT 49-53 (45 samples) from depths of 120-517 cm, collected at 3-17 cm increments with average lengths of ~1 cm [28].

For age determination, pre-existing magnetostratigraphic data [29] was combined with radioisotopic dating values using the decay of atmospherically-produced (cosmogenic) 10 Be [22]. The resulting sediment accumulation rates were $\sim 3 \text{ mm kyr}^{-1}$. Samples ($\sim 3 \text{ g each}$) from the two largest sets cover the time range of the enhanced 60 Fe signal: ELT 45-21 between 1.8 and 2.6 Myr ago and ELT 49-53 between 1.7 and 3.2 Myr. Additionally, recent (near-surface) samples were studied. The silt and clay dominated [29] sediment samples were leached with a mild acid to extract the authigenic Al fraction and ⁶⁹ chemically treated using a procedure described elsewhere ⁷⁰ [28, 30, 31]. On average, 3 mg of Al₂O₃ was produced ⁷¹ from each sample.

Assuming an ${}^{60}\text{Fe}/{}^{26}\text{Al}$ isotope ratio of 0.5 (e.g. 72 [32]) and identical transport of ²⁶Al and ⁶⁰Fe, we can 73 estimate the SN-associated ²⁶Al deposition in terres-74 trial archives from the ⁶⁰Fe signal. Using the decay-75 corrected concentration of $(5-10) \times 10^4$ ats g⁻¹ of ⁶⁰Fe (see 76 Tab. 1 in Ref [22], corresponding to an average depo-77 sition rate of $\sim 25 \,\mathrm{ats} \,\mathrm{cm}^{-2} \,\mathrm{yr}^{-1}$), we would expect (1-78 $2) \times 10^{526} \text{Al} \text{ ats g}^{-1} \text{ sediment (or } \sim 50 \text{ ats cm}^{-2} \text{ yr}^{-1}) \text{ at}$ 79 the time of deposition. After 2.6 Myr of radioactive de-80 cay (corresponding approximately to the SN peak's cen-81 ter) $(0.9-1.7) \times 10^4 \text{ ats g}^{-1}$ are left. 82

The only method sensitive enough to measure such 83 low concentrations is AMS. The ²⁶Al content of the 84 leachate was determined via ²⁶Al/²⁷Al isotope ratio mea-85 surements at the AMS facility VERA (Vienna Environ-86 mental Research Accelerator) at the University of Vi-87 enna, Austria. The amount of stable authigenic ²⁷Al was 88 measured with inductively coupled plasma mass spec-89 trometry (ICP-MS) at the Helmholtz-Zentrum Dresden-90 Rossendorf, Germany. On average, each sample leachate 91 contained $(1.86\pm0.07)\times10^{19}$ ats g⁻¹ of ²⁷Al (see supple-92 ment). This value combined with the estimated SN-93 associated ²⁶Al value of $(0.9-1.7) \times 10^4$ ats g⁻¹ results in an isotope ²⁶Al/²⁷Al ratio of $(0.5-1) \times 10^{-15}$; a ratio very 94 95 close to the detection limit of $\sim 6 \times 10^{-16}$. With an over-96 all detection efficiency of 2×10^{-4} [33] for ²⁶Al at VERA, 97 the estimated SN-associated ²⁶Al influx would result in 98 the detection of 5-10 26 Al atoms per 3 g sample. 99

Concurrent natural ²⁶Al production on Earth makes 100 the detection of any SN-associated ²⁶Al influx above the 101 terrestrial background challenging. The main production 102 mechanisms of ²⁶Al in the Earth's atmosphere are spal-103 lation reactions via cosmic-ray particles on argon [34]. A₁₂₅ 104 mean atmospheric flux of $\sim 1280^{26}$ Al ats cm⁻² yr⁻¹ is ob-126 105 served in the Earth's atmosphere of which about 5 % orig-127 106 inate from influx of extraterrestrial matter such as mete-128 107 orites and interplanetary dust [35]. These constantly pro-129 108 duced cosmogenic radionuclides reach the deep-sea sed-130 109 iment surfaces on timescales of 100 years [36] and make₁₃₁ 110 up our baseline background above which a SN-associated₁₃₂ 111 26 Al signal has to be detected. Post-depositional *in-situ*₁₃₃ 112 production also contributes to the terrestrial ²⁶Al back-134 113 ground, albeit at lower yields [37]. 114 135

The individual samples were measured with AMS for₁₃₆ 115 several hours each until fully consumed, resulting in a₁₃₇ 116 precision of between 3 and 15%. While the modern sur-138 117 face samples yielded up to 1240 counts of ²⁶Al atoms in₁₃₉ 118 a single 3 g sample, the 3 Myr old samples yielded only₁₄₀ 119 \sim 70 counts. The measured ²⁶Al/²⁷Al ratios, contain-₁₄₁ 120 ing the terrigenic and any potential SN-associated ²⁶Al,¹⁴² 121 were found to exponentially decrease with increasing age₁₄₃ 122 (Fig. 1a). 123 144

¹²⁴ In the following, we investigate whether the data show₁₄₅

FIG. 1. (a) 26 Al/ 27 Al ratios of individual samples from 4 deep-sea sediment cores versus time, not corrected for radioactive decay. The exponential decay function derived from the measured initial (surface) ratio is displayed as a coloured line with its uncertainty range. (b) Decay-corrected 60 Fe/Fe ratios as 200 kyr-averages versus age fitted with a Gaussian distribution and showing only the fit uncertainties. (c) 26 Al/ 27 Al ratios as 200 kyr-averages versus age, not corrected for radioactive decay (logarithmic scale). The Gaussian-shaped 60 Fe signal has been translated to SN-associated 26 Al using an isotopic ratio of 60 Fe/ 26 Al = 0.02.

any SN-associated ²⁶Al influx on top of the baseline influx of ²⁶Al. We assume a constant production rate for ²⁶Al that is dominated by cosmogenic atmospheric production neglecting *in-situ* production, and no significant SN-associated influx at present time, as demonstrated by the 60 Fe data ([22], Fig. 1b). Five surface samples vielded a modern ratio of ${}^{26}\text{Al}/{}^{27}\text{Al}=(2.56\pm0.08)\times10^{-13}$, which was used to calculate an exponential function with its error (Fig. 1a). This modern ²⁶Al/²⁷Al surface ratio is in excellent agreement with the decaycorrected average of all samples between 1.7 and 3.2 Myr of $(2.60\pm0.03)\times10^{-13}$, suggesting a SN-associated ²⁶Al contribution was not detected. Obvious deviations from the derived exponential function, being beyond statistical fluctuations and occurring as clusters of neighbouring values that span up to about 0.2 Myr, might indicate unknown geological processes acting at these time scales. In contrast, the ⁶⁰Fe data indicates a SN-associated material deposition lasting more than 1 Myr ([22], Fig. 1b), which would be equally expected for SN-associated ²⁶Al influx. The data analysis does not show any long-term



¹⁴⁶ deviations from an ideal exponential decay of atmo¹⁴⁷ spheric ²⁶Al input, hence, we conclude that any SN¹⁴⁸ associated ²⁶Al influx is below statistical significance.

We assume that the SN-associated ²⁶Al is hidden within our ²⁶Al measurement uncertainty to derive limits in the SN-associated ⁶⁰Fe/²⁶Al ratio with

$$\left(\frac{{}^{60}\text{Fe}}{{}^{26}\text{Al}}\right)_{\text{SN}} \ge \frac{{}^{60}\text{Fe}}{\sigma({}^{26}\text{Al})} = \frac{\left(\frac{{}^{60}\text{Fe}}{{}^{5e}}\right)_{\text{AMS}} \times C_{\text{Fe}}}{\sigma\left[\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_{\text{AMS}} \times C_{{}^{27}\text{Al}}\right]}.$$
 (1)

The individual ²⁶Al/²⁷Al sediment data from AMS mea-152 surements within the SN-peak interval of 1.7-3.2 Myr 153 were converted to atom concentrations of ²⁶Al per gram 154 using the corresponding ${}^{27}Al$ concentrations $C_{27}Al$ cor-155 recting for radioactive decay. The uncertainties σ ⁽²⁶Al) 156 of the resulting data set increase with sediment age due 157 to lower ²⁶Al counting statistics at higher ages (Fig. 2a) 158 and pose an upper limit of SN-associated ²⁶Al influx with 159 time. Similarly, individual ⁶⁰Fe/Fe data were converted 160 to decay-corrected ⁶⁰Fe concentrations using the Fe con-161 centrations C_{Fe} (Fig. 2b, supplementary tables I-III). 162

¹⁶³ The ratio of the SN-associated ⁶⁰Fe concentration to ¹⁶⁴ the uncertainties of the ²⁶Al concentration (Fig. 2c) rep-¹⁶⁵ resents a lower limit for the SN-associated ⁶⁰Fe/²⁶Al ra-¹⁶⁶ tio. Note the ⁶⁰Fe/ σ (²⁶Al) ratios are influenced by scat-¹⁶⁷ ter in the ⁶⁰Fe concentrations; and the increasing uncer-¹⁶⁸ tainty of ²⁶Al with time causes a decrease in the ratios ¹⁶⁹ at greater ages.

We modelled a lower limit of 60 Fe/ 26 Al by fitting the 170 data using a Nadaraya-Watson kernel regression [38, 39] 171 with a confidence level of 68% (Fig. 2c , see supplemen-172 tary information). Calculating the mean of the function 173 and its uncertainty in the time-period of 1.5 Myr yields an 174 average lower limit of ${}^{60}\text{Fe}/{}^{26}\text{Al}{=}0.18^{+0.15}_{-0.08}$. Hence, considering only the lower and upper bounds of the 68 % un-175 176 certainty band yields two average data-derived lower lim-177 its for the SN-associated ⁶⁰Fe/²⁶Al ratio: A conservative 178 minimum lower limit of ${}^{60}\text{Fe}/{}^{26}\text{Al} \ge 0.18-0.08 = 0.10$ and 179 a maximum lower limit of ${}^{60}\text{Fe}/{}^{26}\text{Al} \ge 0.18 + 0.15 = 0.33$. 180 For comparison, we fitted the ²⁶Al atom concentra-181 tion uncertainties yielding an average upper limit of_{198} 182 44×10^4 ats g⁻¹ (Fig.2a) and the ⁶⁰Fe concentration of₁₉₉ 183 $7.5^{+5.5}_{-3.3} \times 10^4 \text{ ats g}^{-1}$ (Fig.2b) to calculate the lower SN-¹⁹⁹₂₀₀ associated ⁶⁰Fe/²⁶Al limit yielding a similar result of₂₀₁ 184 185 $0.17^{+0.13}_{-0.08}$ 186 We use our derived minimum and maximum lower203 187

 60 Fe/ 26 Al limits in combination with the measured SN- $_{204}$ 188 associated ⁶⁰Fe data to deduce SN-associated ²⁶Al yields₂₀₅ 189 and check these for compatibility with our initially mea-206 190 sured ²⁶Al/²⁷Al data. This approach requires the as-207 191 sumption that after ejection by a SN 60 Fe and 26 Al₂₀₈ 192 behave identically during transport and deposition on₂₀₉ 193 Earth. First, we modelled the measured and decay-210 194 corrected 60 Fe/Fe time profile with a Gaussian fit of the₂₁₁ 195 data, yielding a signal centered at 2.64 Myr with a full₂₁₂ 196



FIG. 2. ²⁶Al atom concentrations uncertainties (a) and ⁶⁰Fe atom concentrations (b) per gram of sediment corrected for background and radioactive decay, and their ratios (c). Panel (a) displays the regression line (solid blue) of the data as upper limit, (b) and (c) show the mean and uncertainty of each regression by solid blue lines. Their averages are displayed as solid (mean) and dashed (uncertainty) black lines.

width at half maximum of 1.14 Myr (Fig. 1b). Next, we converted this signal to the absolute amount of ⁶⁰Fe using concentrations of stable Fe $(1.81\pm0.03\times10^{19} \text{ ats g}^{-1} \text{ on})$ average, see supplement). Subsequently, we calculated the corresponding amount of ²⁶Al which we translated to a ${}^{26}\text{Al}/{}^{27}\text{Al}$ time profile using the measured average concentration of stable ²⁷Al $(1.86\pm0.07\times10^{19} \text{ ats g}^{-1})$. Taking the exponential decay into account, we added the $^{26}\text{Al}/^{27}\text{Al}$ time profile to the exponential function obtained from the modern surface samples (Fig. 3). The maximum lower SN-associated ²⁶Al/⁶⁰Fe limit of 0.33 results in a SN-associated $^{26}\mathrm{Al}$ signal hidden within the uncertainty of the terrestrial influx. A higher ²⁶Al SN influx, based on the minimum lower limit of 0.10 indicates a SN signal that is not entirely supported by the 200 kyr and moving averages of the measured ${}^{26}\text{Al}/{}^{27}\text{Al}$ data.



FIG. 3. 200 kyr-averages of 26 Al/ 27 Al isotope ratios, a moving average summed over 5 adjacent data points and the exponential function derived from modern samples versus sediment age. The two SN-associated 26 Al signals on top of atmospheric input (blue) correspond to the specific lower 60 Fe/ 26 Al limits derived from experimental data.

In the following, we examine the wide range of reported 213 60 Fe/ 26 Al ratios deduced from stellar nucleosynthesis 214 models for compatibility with our experimental data. 215 Different input physics (e.g. reaction rates, stellar ro-216 tation) in the nucleosynthesis models leads to ${}^{60}\text{Fe}/{}^{26}\text{Al}$ 217 production ratios for SNe that vary between 0.02 and 2 218 over the stellar initial mass range of $9-25 \,\mathrm{M_{\odot}}$ [11–13, 40– 219 43]. 220

As an example, we use the lowest reported ${}^{60}\text{Fe}/{}^{26}\text{Al}$ 221 ratio of 0.02 [13] to convert the Gaussian-shaped 60 Fe 222 time profile (Fig. 1b) to a SN-associated ²⁶Al/²⁷Al time 223 profile, which is added to the exponential function ob-224 tained from modern surface samples. The resulting sig-225 nal from the model is not observed in the measured data 226 (Fig. 1c), but shows that 26 Al would have been detected 227 if the SN ejecta reaching Earth would have carried this 228 low ${}^{60}\text{Fe}/{}^{26}\text{Al}$ ratio. 229

Thus, our experimental SN-associated ${}^{60}\text{Fe}/{}^{26}\text{Al lim}^{244}$ 230 its are in agreement with most of the CCSN-associated 231 ratios derived from stellar nucleosynthesis calculations 232 233 to have a high 60 Fe yield but negligible 26 Al production 234 during explosive nucleosynthesis. After exploding, the $\frac{1}{250}$ 235 expanding remnant of the ECSN picks up the matter₂₅₁ 236 blown out by the stellar winds of its prior SAGB phase 237 [44] that contains large amounts of 26 Al and 60 Fe ([10], 238 Fig. 4). In such a scenario, the total 26 Al/ 60 Fe isotope 239 ratio becomes 240 255

$$\left(\frac{{}^{60}\text{Fe}}{{}^{26}\text{Al}}\right)_{\text{final}} = \frac{{}^{60}\text{Fe}_{\text{SAGB}} + {}^{60}\text{Fe}_{\text{ECSN}}}{{}^{26}\text{Al}_{\text{SAGB}}} \times \frac{26}{60}. \qquad (2)_{257}^{256}$$



FIG. 4. $^{60}\mathrm{Fe}/^{26}\mathrm{Al}$ nucleosynthesis isotope ratios [10–13, 40–43] versus initial stellar mass. We display ratios of (S)AGB stars and SNe that contribute $^{26}\mathrm{Al}$ and $^{60}\mathrm{Fe}$ to the ISM as well as the galactic average γ -flux ratio. The shaded blue areas indicate the possible SN-associated $^{60}\mathrm{Fe}/^{26}\mathrm{Al}$ ratios derived from our measured $^{26}\mathrm{Al}$ data. Abbreviations denote rotating (rot) and non-rotating (non-rot) stellar models [42]. Z9.6 and W18 refer to pre-SN evolution models in the mass ranges of 9-12 M_{\odot} and 12.5-25.2 M_{\odot} , respectively [43].

of $^{60}\rm{Fe}$ and $3.064\times10^{-6}\,\rm{M}_{\odot}$ of $^{26}\rm{Al}$. If this star explodes as ECSN an additional $^{60}\rm{Fe}$ SN contribution, with a yield ranging from $3.61\times10^{-5}\,\rm{M}_{\odot}$ to $1.3\times10^{-4}\,\rm{M}_{\odot}$ [14], may increase the original $^{60}\rm{Fe}/^{26}\rm{Al}$ SAGB ratio to 5.5-18.8. Our results agree with previous studies that suggested ECSNe as primary candidates for the origin of the $^{60}\rm{Fe}$ signal 2-3 Myr ago [45, 46]. However, our sediment data is also consistent with nucleosynthesis models for more massive stars.

The modelled SN-associated 60 Fe/ 26 Al nucleosynthesis ratios are usually integrated over the IMF (initial mass function, mass distribution of stars in a stellar cluster at birth) to obtain an average galactic steady-state ratio. This ratio is, for some SN nucleosynthesis models, higher than the 60 Fe/ 26 Al γ -flux ratio observed in the ISM of (0.15 \pm 0.05) [8, 12, 40, 43]. It has been suggested that this difference could be bridged by additional sources, such as the stellar winds of WR-stars that add a significant

fraction of ²⁶Al to the ISM [16, 47]. Our experimental₃₁₆ 262 lower limits for ⁶⁰Fe/²⁶Al map recent specific local SN³¹⁷ 263 events within our solar environment, as opposed to the $^{\scriptscriptstyle 318}$ 264 steady-state conditions of the ISM. Since our data $\mathrm{set}^{^{319}}$ 265 provides lower limits, it does not exclude, for example, $_{320}^{320}$ 266 an additional WR-source enriching the galactic mixture 267 with ²⁶Al and thus lowering the observed γ -flux ratio₃₂₃ 268 compared to the SN-associated ${}^{60}\text{Fe}/{}^{26}\text{Al}$ ratios. 269

We note, that all statements made so far assume that³²⁵ 270 ⁶⁰Fe and ²⁶Al are transported equally to the solar system³²⁶ 271 within dust particles. It is in fact not clear whether their³²⁷ 272 isotopic ratio is conserved during the journey e.g. due_{329}^{328} 273 to different dust survival rates within the SN remnant $\frac{1}{330}$ 274 [46, 48] and non-isotropic clumpy ejecta [49]. However,331 275 the ⁶⁰Fe signal is advocated to originate from a series of₃₃₂ 276 nearby SN explosions forming the Local Superbubble in³³³ 277 which our solar system is embedded [24, 45, 50]. The³³⁴ 278 combined ejecta from a number of SNe could average out³³⁵ 279 some of the inhomogeneities between 26 Al and 60 Fe. 280 337

Under the assumptions made, we can conclude that the astrophysical scenarios proposed to explain the SN- $_{339}$ associated 60 Fe signal (ECSNe, CCSNe) are consistent with our results derived from 26 Al measurements. The³⁴¹ non-detection of 26 Al provides a constraint on the SN- 342 associated 60 Fe/ 26 Al isotope ratio in the solar environ- 343 ment in the recent past.

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