



1 **Isotopic composition of aerosol iron from anthropogenic sources:**
2 **implications for source apportionment of aerosol iron**

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24



25 **Abstract**

26 Aerosol iron (Fe) significantly impacts human health, atmospheric chemistry and marine
27 biogeochemistry. The stable isotope ratio of Fe, typically reported as $\delta^{56}\text{Fe}$, has emerged as a
28 promising method for source apportionment of total and soluble aerosol Fe. However, the $\delta^{56}\text{Fe}$
29 endmember values remain poorly constrained for aerosol Fe from various non-dust sources,
30 impeding the application of Fe isotopes in atmospheric research. This work measured isotopic
31 compositions for aerosol Fe from desert dust and several anthropogenic sources. The average
32 $\delta^{56}\text{Fe}$ was determined to be $+0.14 \pm 0.10\text{‰}$ for the seven dust samples we examined, in good
33 agreement with previous work. We found that different anthropogenic aerosols exhibit a wide
34 range of Fe isotopic composition. Compared to desert dust, the average $\delta^{56}\text{Fe}$ was found to be
35 higher for power plant coal fly ash ($+0.26 \pm 0.18\text{‰}$, $n=28$), slightly lower for steelwork fly ash
36 ($-0.07 \pm 0.41\text{‰}$, $n=18$), and considerably lower for biofuel burning aerosol ($-0.28 \pm 0.39\text{‰}$,
37 $n=11$). In addition, the average $\delta^{56}\text{Fe}$ was determined to be $+0.20 \pm 0.12\text{‰}$ ($n=2$) for municipal
38 incineration fly ash, $+0.38 \pm 0.13\text{‰}$ ($n=1$) for heavy oil bottom ash, and $+0.08 \pm 0.13\text{‰}$ for
39 certificated urban particulate matter sample ($n=1$). We suggest that not all the anthropogenic
40 aerosol Fe is isotopically lighter than natural dust Fe, in contrast to what is conventionally
41 assumed. Our findings also imply that Fe isotope-based source apportionment must account
42 for the $\delta^{56}\text{Fe}$ endmember variability both among and within different anthropogenic aerosols.
43 Overall, our work substantially improves our ability to constrain $\delta^{56}\text{Fe}$ endmember values from
44 various anthropogenic sources.

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46



47 **1 Introduction**

48 Atmospheric aerosol deposition is the primary external source of dissolved iron (Fe) in
49 the surface waters of the open ocean, influencing marine primary productivity and the global
50 carbon cycle (Boyd and Ellwood, 2010; Moore et al., 2013; Tagliabue et al., 2017). Aerosol Fe
51 also has important impacts on human health and atmospheric chemical processes (Zuo et al.,
52 2022; Al-Abadleh, 2024). On the global scale, natural dust aerosol emitted from arid and semi-
53 arid regions is the major source of total aerosol Fe (Jickells et al., 2005; Boyd et al., 2007). On
54 the other hand, although the contribution of anthropogenic sources to total aerosol Fe is rather
55 small, they may contribute substantially to soluble aerosol Fe as the solubility of anthropogenic
56 aerosol Fe can be much higher than natural dust Fe (Ito et al., 2019; Rathod et al., 2020; Chen
57 et al., 2024). Currently, it remains a great challenge to accurately distinguish and quantify the
58 contributions of different sources to total and soluble aerosol Fe (Mahowald et al., 2018; Zhang
59 et al., 2025).

60 As an emerging technique, Fe isotopes demonstrate unique advantages in source
61 apportionment of aerosol Fe, as Fe in natural dust and anthropogenic aerosols may have distinct
62 isotopic signatures (Fitzsimmons and Conway, 2023), typically reported as $\delta^{56}\text{Fe}$. Natural dust
63 aerosols from different regions exhibit relatively homogeneous Fe isotopic composition with
64 $\delta^{56}\text{Fe}$ values around +0.1‰ (Wang et al., 2022; Wei et al., 2024; Zhang et al., 2025), being very
65 similar to that for the upper continental crust (UCC) (Poitrasson, 2006). Field observations
66 usually attributed lighter Fe (i.e. $\delta^{56}\text{Fe}$ lower than +0.1‰) they observed to the influence of
67 anthropogenic emissions, and employed the two-component mixing model to quantitatively
68 resolve the contribution of natural dust and anthropogenic sources to total and soluble aerosol
69 Fe (Zhang et al., 2025). For instance, over the North Atlantic, the average $\delta^{56}\text{Fe}$ was found to
70 be $-1.15 \pm 0.24\text{‰}$ for air masses from Europe and North America, much lower than that
71 ($+0.09 \pm 0.02\text{‰}$) for air masses originating from the Saharan region (Conway et al., 2019), and



72 fossil fuel combustion was estimated to contribute 50-100% to soluble aerosol Fe observed in
73 European and North American air masses. Another study ([Kurusu et al., 2024](#)) found $\delta^{56}\text{Fe}$ to
74 be as low as -0.5‰ and -1.9‰ for total and soluble Fe in total suspended particles (TSP)
75 collected over the subarctic North Pacific, and suggested that combustion contributed up to 13%
76 and 45% of total and soluble Fe.

77 The $\delta^{56}\text{Fe}$ endmember values directly affect the result of Fe isotope-based source
78 apportionment, but remain poorly constrained for anthropogenic aerosols. First, in most
79 previous studies, a given study usually assigned a single $\delta^{56}\text{Fe}$ endmember value (usually set
80 to the lowest $\delta^{56}\text{Fe}$ value observed) to anthropogenic aerosol Fe ([Zhang et al., 2025](#)); in other
81 words, they did not take into account the potential difference in $\delta^{56}\text{Fe}$ endmember values for
82 aerosol Fe from different anthropogenic sources (e.g., fossil fuel combustion, steelwork, and
83 biomass burning). Furthermore, $\delta^{56}\text{Fe}$ endmember values adopted for anthropogenic aerosol
84 Fe showed large variations across different studies, ranging from -4.7‰ to -1.6‰ ([Conway et](#)
85 [al., 2019](#); [Kurusu et al., 2021](#); [Hsieh and Ho, 2024](#); [Kurusu et al., 2024](#); [Bunnell et al., 2025](#);
86 [Shuai et al., 2025](#)). Lastly, a few studies ([Labatut et al., 2014](#); [Bunnell et al., 2025](#); [Camin et](#)
87 [al., 2025](#)) found $\delta^{56}\text{Fe}$ values significantly higher than +0.1‰ for tropospheric aerosols, and
88 the sources and/or mechanisms which could explain the heavier Fe observed in tropospheric
89 aerosols are still under debate.

90 The available measurements at present are not sufficient to provide reliable constraints on
91 $\delta^{56}\text{Fe}$ endmember values for anthropogenic aerosols. Two recent studies ([Wang et al., 2022](#);
92 [Wei et al., 2024](#)) compiled aerosol Fe isotope data and then utilized the MixSIAR model to
93 quantify aerosol Fe sources. The $\delta^{56}\text{Fe}$ endmember values adopted for anthropogenic emission
94 display large inconsistency, for example, being -1.6 ± 0.1 ‰ for fossil fuel combustion emission
95 in one study ([Wang et al., 2022](#)), and $+0.4 \pm 0.2$ ‰ for coal combustion emission in the other
96 study ([Wei et al., 2024](#)). As discussed in a recent review paper ([Zhang et al., 2025](#)), only a



97 limited number of studies have measured the isotopic composition of aerosol Fe from different
98 anthropogenic sources, and these studies only examined a very small number of anthropogenic
99 sources and a limited number of samples for each source. Moreover, the $\delta^{56}\text{Fe}$ endmember
100 values of biomass burning aerosol, which may be an important source for soluble aerosol Fe
101 (Mead et al., 2013; Li et al., 2025), have not been determined.

102 The lack of reliable $\delta^{56}\text{Fe}$ endmember values for anthropogenic aerosols limits the
103 application of Fe isotopes in atmospheric aerosol research. This study measured the isotopic
104 composition of aerosol Fe from several important anthropogenic emissions, including coal fly
105 ash, steelwork fly ash, biofuel burning aerosols, municipal incineration fly ash, heavy oil
106 bottom ash, and certificated urban particulate matter sample. In contrast to what is
107 conventionally assumed, we found that not all the anthropogenic aerosol Fe is isotopically
108 lighter than natural dust Fe. Moreover, large variabilities in Fe isotopic compositions were
109 revealed among and within different anthropogenic aerosols. Our measured $\delta^{56}\text{Fe}$ endmember
110 values can be important for quantitative isotope-based source apportionment of aerosol Fe.

111 2 Materials and Methods

112 2.1 Sample information

113 This study measured the Fe isotope compositions for seven types of particulate samples,
114 consisting of desert dust, power plant coal fly ash, steelwork fly ash, biofuel burning aerosols,
115 municipal waste fly ash, oil bottom ash, and urban particulate matter.

116 2.1.1 Desert dust, power plant coal fly ash, and steelwork fly ash

117 Seven desert dust samples were examined (Table 3), comprising Saharan dust, Arizona
118 Test Dust (ATD, nominal 0-3 μm fraction), Luochuan loess (LC loess), Taklimakan dust (TK
119 dust), Gobi dust (GB dust), Qinghai dust (QH dust), and Tibetan Plateau dust (Tibet dust).
120 Saharan dust was collected at the Cape Verde Islands (Chen et al., 2020), ATD was purchased
121 from Powder Technology, Inc., USA, and the last five dust samples were collected from



122 different regions in China.

123 Among the 28 power plant coal fly ash samples we studied (Table S3), 26 samples were
124 provided by large coal-fired power plants from 25 provinces in China (Li et al., 2025), one
125 sample was the certified reference material (GBW08401) provided by the Research Center for
126 Eco-Environmental Sciences, Chinese Academy of Sciences (Li et al., 2022), and one sample
127 (CFA-FDU) was provided by Fudan University (Wu et al., 2023).

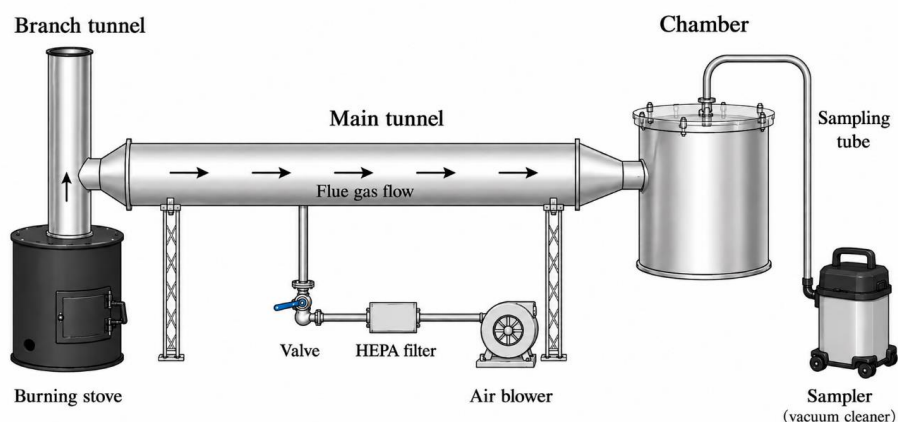
128 The 21 steelwork fly ash samples (Table S4) were provided by large and medium-sized
129 steelwork plants located in different regions in China (Li et al., 2025). These samples were
130 emitted from several major steelwork processes, including coking (n=3), sintering (n=5), blast
131 furnace (n=2), converter (n=2), post-steelmaking (n=1), and casting (n=1). Samples could be
132 provided by the same plant or obtained from the same/similar steelwork processes, while there
133 were no two samples obtained from the same process in the same plant. Among the 21 samples,
134 12 samples were collected from baghouses or electrostatic precipitators, 2 blast furnace fly ash
135 samples were obtained from gravity settlers, and the other 7 samples were collected from
136 chimney outlets. All the samples were dried in an oven (65 °C) for 24 hours, and stored in
137 sealed vessels for further analysis.

138 **2.1.2 Biofuel burning aerosols**

139 We employed the device depicted in Figure 1 to generate and collect biofuel burning
140 aerosols, and more details can be found elsewhere (Li et al., 2025). It should be noted that
141 biofuel burning experiments this work conducted are designed to simulate domestic biofuel
142 burning, and may not be representative of wildfires (Hamilton et al., 2022). As shown in Figure
143 1, biofuel was burned in a commercial cook stove, and the smoke generated from biofuel
144 burning passed through a horizontal metal chimney (200 cm in length, 30 cm in inner diameter)
145 and then entered a chamber (50 cm in height, 45 cm in diameter). Air in the chamber was then
146 drawn through a filter bag using a vacuum cleaner to collect aerosol particles. Being different



147 from our previous work (Li et al., 2025) which used a medium volume aerosol sampler to
148 collect PM_{2.5} samples, the present study employed a vacuum cleaner to collect aerosol particles
149 emitted from biofuel burning (i.e. no size selection was applied), and the purpose was to obtain
150 a sufficient amount of Fe-containing particles for Fe isotopic analysis. Biofuel examined in this
151 work contained no apparent soil particles and was burned in a stove, and the influence of Fe
152 contained in soil particles was minimized.



153

154 **Figure 1.** Schematic diagram of the device used in our study to generate and collect biofuel
155 burning aerosols. This figure was created with the assistance of ChatGPT.

156

157 Prior to each sample collection, a new filter bag was used for the vacuum cleaner and the
158 sampling tubing was cleaned with ultrapure water, in order to avoid cross-contamination
159 between samples. Each sample was collected over a period of 6-12 h, with final sample mass
160 of 2-5 g. After sampling, filter bags loaded with particles were placed in a desiccator at room
161 temperature for 48 hours to remove moisture, and then stored in sealed vessels for further
162 analysis.

163 This work examined 11 types of biofuel commonly found in China, consisting of 5 crop
164 straws (wheat, maize, peanut, soybean, and rice) and 6 woods (Chinese fir, birch, pine, lychee,



165 apple, and pear). One aerosol sample was collected for each biofuel, and thus in total 11 biofuel
166 burning aerosol samples were collected.

167 **2.1.3 Municipal waste fly ash, oil bottom ash, and urban particulate matter**

168 Moreover, this study measured the Fe isotope compositions of another 4 anthropogenic
169 samples. They were a municipal waste fly ash sample (MWFA-FDU) provided by Fudan
170 University (Ding et al., 2019), a certified reference material for waste incineration fly ash
171 (BCR-615) from the Institute for Reference Materials and Measurements (IRMM), a heavy oil
172 bottom ash sample provided by Fudan University (Fu et al., 2012), and a certificated reference
173 material for urban particulate matter (NIST 1648a) from the National Institute of Standards and
174 Technology (NIST).

175 **2.2 Sample pretreatment and chemical purification**

176 Sample pretreatment (mainly digestion) and chemical purification were conducted in a
177 Class 100 ultra-clean laboratory at Guangzhou Institute of Geochemistry, Chinese Academy of
178 Sciences. A brief overview is given below, and further details are provided in the Supplement
179 (Sect. S1).

180 **2.2.1 Sample digestion**

181 Due to low Fe content and high organic matter content, biofuel burning aerosol samples
182 were first heated in a muffle furnace at 900 °C for at least 90 min to remove organic matters
183 they contained; after being cooled down to room temperature, for each sample, 0.5 g of the
184 sample was weighed and then used for subsequent analysis. All the other samples were heated
185 in a muffle furnace at 600°C for at least 60 min; after being cooled down, for each sample, 10-
186 35 mg was weighed and then used for further analysis.

187 As detailed in the Supplement (Sect. S1.2), except for steelwork fly ash and biofuel
188 burning aerosol, samples examined in this work were digested by a sequential protocol at
189 120 °C, using 1) HNO₃-HF, 2) H₂O₂-HNO₃, 3) Aqua Regia, and 4) HCl, respectively. Steelwork



fly ash and biofuel burning aerosol samples contained substantial amounts of carbonaceous or refractory materials, requiring additional pre-digestion to remove carbonaceous materials and microwave-assisted digestion to dissolve refractory materials; as a result, these samples were digested using the following sequential digestion protocol, namely 1) pre-digestion in HNO_3 - H_2O_2 at room temperature, 2) microwave-assisted digestion in HNO_3 -HF, 3) heated dissolution in Aqua Regia, and 4) heated dissolution in HCl. After digestion, the digestates were diluted with 2.5 mol/L HCl and then subjected to centrifugation, and the resulting solutions were used for the subsequent chemical purification.

2.2.2 Chemical purification

Chemical purification, which minimizes matrix effects and enhances the accuracy of Fe isotope analysis, is an essential step for high-precision Fe isotope analysis. In this study, chemical purification of Fe was achieved using a single column filled with AGMP-50 cation exchange resin (Bio-Rad, USA), as detailed in the Supplement (Sect. S1.3). In brief, the resin column was sequentially cleaned with 6 mol/L HNO_3 (10 mL), 6 mol/L HCl (15 mL), and ultrapure water (5 mL), respectively. It was then conditioned with 0.2 mol/L HCl (1 mL), and after that 100 μL sample solution was loaded into the resin column. Subsequently, HCl-HF (both at 0.2 mol/L, 3 mL) and HCl-HF solution (which contained 0.2 and 0.5 mol/L HCl and HF, 1 mL) were sequentially added into the resin column to elute matrix elements. Finally, 7 mL HCl-HF solution (which contained 0.2 and 0.5 mol/L HCl and HF) was added into the resin column to elute Fe, and the Fe-containing fraction was collected into a clean PFA beaker.

The Fe-containing fraction was then evaporated to dryness, dissolved with 200 μL concentrated HNO_3 , and evaporated again to dryness at 120 °C. The obtained residue was then dissolved in 2% HNO_3 for Fe isotope measurement. For the purification procedure employed in this study, the Fe recovery was >98% and the procedural blank was <6 ng (Zhu et al., 2020). This procedural blank was negligible when compared to the amount of Fe our samples



215 contained (>300 ng).

216 **2.3 Multi-Collector Inductively Coupled Plasma Mass Spectrometry**

217 A multi-collector inductively coupled plasma mass spectrometer (Neptune Plus MC-ICP-
218 MS, Thermo Fisher Scientific) was employed to measure the Fe isotopic compositions of our
219 samples. The sample and standard solutions were diluted to the same Fe concentration (2 µg/g)
220 using 2% HNO₃ (v/v), in order to minimize the measurement errors arising from differences in
221 solution matrix and Fe concentration (Malinovsky et al., 2003; Dauphas et al., 2009). Since
222 argon (Ar) was used as the carrier gas and the solutions to be analyzed contained HNO₃,
223 polyatomic ions (such as ⁴⁰Ar¹⁴N⁺, ⁴⁰Ar¹⁶O⁺, ⁴⁰Ar¹⁶OH⁺, and ⁴⁰Ar¹⁸O⁺) could be generated in
224 high-temperature plasma and would cause isobaric interferences on ⁵⁴Fe, ⁵⁶Fe, ⁵⁷Fe, and ⁵⁸Fe
225 (Weyer and Schwieters, 2003). To effectively eliminate these isobaric interferences, Fe isotopic
226 measurements were conducted in the pseudo-high-resolution mode (resolution > 14,000) (Zhu
227 et al., 2020).

228 The standard-sample bracketing (SSB) method was employed to correct for instrumental
229 mass discrimination during the analysis. In addition, the impact of ⁵⁴Cr on ⁵⁴Fe was corrected
230 by monitoring the ⁵³Cr signals and using the natural ⁵³Cr/⁵⁴Cr ratio (Weyer and Schwieters,
231 2003). Fe isotopic compositions are typically reported in δ values, given by Eq. (1):

$$232 \quad \delta^x\text{Fe}(\text{‰}) = \left[\frac{(^x\text{Fe}/^{54}\text{Fe})_{\text{sample}}}{(^x\text{Fe}/^{54}\text{Fe})_{\text{standard}}} - 1 \right] \times 1000 \quad (1)$$

233 where x is 56, 57, or 58. To be consistent with the majority of previous studies (Fitzsimmons
234 and Conway, 2023), Fe isotopic compositions are reported here as δ⁵⁶Fe (and δ⁵⁷Fe) values
235 relative to IRMM-014, thereby facilitating comparison among different studies.

236 **3 Results and discussion**

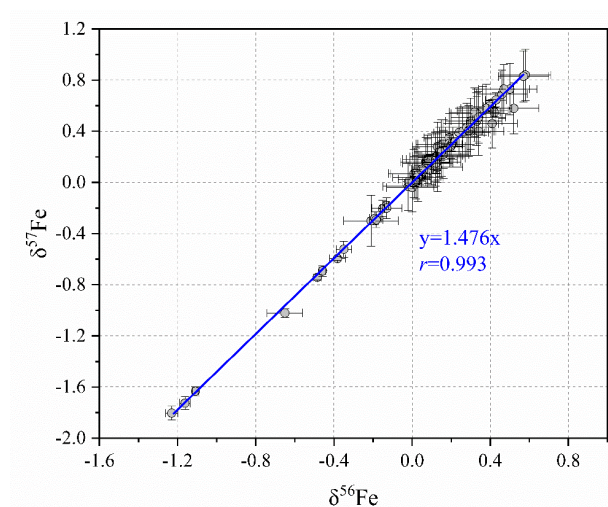
237 **3.1 Measurement precision and accuracy**

238 Figure 1 plots all the δ⁵⁶Fe values measured in our work versus δ⁵⁷Fe, suggesting that
239 δ⁵⁷Fe are very well correlated with δ⁵⁶Fe which ranged from -1.23‰ to +0.58‰. In addition,



240 the slope was found to be 1.476 ± 0.014 (1σ), being very close to the expected value (1.475)

241 (Beard et al., 2003b; Kubik et al., 2021; Zhang et al., 2025).



242

243 **Figure 2.** Comparison of $\delta^{56}\text{Fe}$ versus $\delta^{57}\text{Fe}$ values measured in our work. All the individual
244 measurements (84 in total, including measurements of standards) conducted in this work are
245 presented in this figure.

246

247 As shown in Table S2, for the nine standards/samples for which replicate measurements
248 were conducted, the range of $\delta^{56}\text{Fe}$ values determined by replicate measurements, defined as
249 the difference between the maximum and minimum values, did not exceed 0.08‰. Such
250 variations were not larger than the uncertainties associated with individual measurements,
251 demonstrating the reproducibility and precision of our $\delta^{56}\text{Fe}$ measurements. Moreover, Table 1
252 shows that our measured $\delta^{56}\text{Fe}$ values agreed very well with those reported in literature for the
253 four standards this work our study examined (BRC-2, BHVO-2, SARM4 and GIG-Fe),
254 supporting the accuracy of our $\delta^{56}\text{Fe}$ measurements.

255

256 **Table 1.** Comparison of $\delta^{56}\text{Fe}$ values measured in our work for standards with those reported



in previous studies. In this table, n denotes the number of replicate measurements we performed.

Sample	$\delta^{56}\text{Fe}$ (‰)	Note
BHVO-2	+0.12±0.13 (n=1)	This work
	+0.13±0.05	(Du et al., 2017)
	+0.11±0.03	(Zhao et al., 2017)
	+0.13±0.03	(Zhu et al., 2020)
BCR-2	+0.10±0.08 (n=2)	This work
	+0.11±0.08	(Du et al., 2017)
	+0.11±0.03	(Liu et al., 2014)
	+0.13±0.02	(Zhu et al., 2020)
SARM4	+0.07±0.08 (n=2)	This work
	+0.09±0.03	(Zhu et al., 2020)
GIG-Fe	+0.71±0.12 (n=1)	This work
	+0.71±0.02	(Zhu et al., 2020)

3.2 Isotopic composition of natural desert dust Fe

In this study, $\delta^{56}\text{Fe}$ was measured to be +0.01±0.13‰ (n=1) for Saharan dust. As shown in Table 2, the average $\delta^{56}\text{Fe}$ were measured to be +0.09±0.13‰ and +0.08±0.10‰ for fine (<2.5 μm) and coarse (>2.5 μm) Saharan dust aerosol (n=14) (Mead et al., 2013), and Conway et al. (2019) reported an average $\delta^{56}\text{Fe}$ of +0.12±0.03‰ (n=13) for Saharan dust aerosol. The Fe isotope composition of Saharan dust measured in our work agree well with these reported in previous work (Mead et al., 2013; Conway et al., 2019). The average $\delta^{56}\text{Fe}$ was determined to be +0.04±0.07‰ (n=3) for ATD in this study (Table 2), and it was measured previously to be +0.13±0.04‰ (Mead et al., 2013), -0.04±0.10‰ (Li et al., 2022) and +0.06±0.04‰ (Kurusu et al., 2026). The $\delta^{56}\text{Fe}$ value measured in our work is consistent with those reported in previous studies (Mead et al., 2013; Li et al., 2022; Kurusu et al., 2026), when the overall analytical uncertainties (about ±0.05‰) are taken into account.

Table 2. Comparison of $\delta^{56}\text{Fe}$ values measured in our work for desert dust with those reported in previous studies. In this table, n denotes the number of replicate measurements we performed.



Sample	$\delta^{56}\text{Fe}$ (‰)	Note
Saharan dust	$+0.01 \pm 0.13$ (n=1)	This work
	$+0.09 \pm 0.13$ (<2.5 μm)	(Mead et al., 2013)
	$+0.08 \pm 0.10$ (>2.5 μm)	(Mead et al., 2013)
	$+0.12 \pm 0.03$	(Conway et al., 2019)
Arizona test dust	$+0.04 \pm 0.07$ (n=3)	This work
	$+0.13 \pm 0.04$	(Mead et al., 2013)
	-0.04 ± 0.10	(Li et al., 2022)
	$+0.06 \pm 0.04$	(Kurusu et al., 2026)
Luochuan loess	$+0.14 \pm 0.03$ (n=2)	This work
Loess	$+0.13 \pm 0.06$	(Beard et al., 2003a)
Loess and paleosol	$+0.09 \pm 0.03$	(Gong et al., 2017)
Taklamakan dust	$+0.14 \pm 0.13$ (n=1)	This work
Gobi dust	$+0.30 \pm 0.13$ (n=1)	This work
Qinghai dust	$+0.19 \pm 0.13$ (n=1)	This work
Tibet dust	$+0.16 \pm 0.13$ (n=1)	This work
TLF dust	$+0.21 \pm 0.05$ (n=1)	(Li et al., 2022)

274

275 Our work found the Luochuan loess in China to have an average $\delta^{56}\text{Fe}$ of $+0.14 \pm 0.03\text{‰}$
276 (n=2). The average $\delta^{56}\text{Fe}$ was determined to be $+0.13 \pm 0.06\text{‰}$ for loess samples (n=10) from
277 different regions of the world (Beard et al., 2003a) and $+0.09 \pm 0.03\text{‰}$ for loess and paleosol
278 samples (n=32) from the Chinese Loess Plateau (Gong et al., 2017). Our measured $\delta^{56}\text{Fe}$ of
279 loess agrees well with previous studies (Beard et al., 2003a; Gong et al., 2017). Among the
280 four Chinese dust samples we examined (Table 2), $\delta^{56}\text{Fe}$ were measured to be $+0.14 \pm 0.13\text{‰}$,
281 $+0.19 \pm 0.13\text{‰}$, and $+0.16 \pm 0.13\text{‰}$ for Taklamakan dust, Qinghai dust, and Tibet dust, in good
282 agreement with that for TLF dust ($+0.21 \pm 0.05\text{‰}$) reported in our previous work (Li et al.,
283 2022). Gobi dust was found in our work to have a $\delta^{56}\text{Fe}$ value of $+0.30 \pm 0.13\text{‰}$, which is
284 broadly comparable with those for other dust samples.



In summary, the average and median $\delta^{56}\text{Fe}$ were found to be $+0.14 \pm 0.10\text{‰}$ (1σ) and $+0.14\text{‰}$ for the seven dust samples we studied (Table 3), and $\delta^{56}\text{Fe}$ values fell into a small range ($+0\text{‰}$ to $+0.2\text{‰}$) for most samples. Our experimental measurements support the view that the average Fe isotopic composition of desert dust is close to that of UCC ($+0.09 \pm 0.10\text{‰}$) (Beard et al., 2003b; Beard and Johnson, 2004; Poitrasson, 2006).

290

Table 3. Summary of our measured $\delta^{56}\text{Fe}$ values (in ‰) for natural desert dust, power plant coal fly ash, steelwork fly ash, biofuel burning aerosol, municipal waste fly ash, heavy oil bottom ash, and urban particulate matter. In this table, n denotes the number of samples we investigated for each sample type; for steelwork fly ash, this table does not include the 3 samples from the coking process (see Section 3.3.2 for more details).

sample type	n	range	average	median
natural desert dust	7	+0.01 to +0.30	$+0.14 \pm 0.10$	+0.14
power plant coal fly ash	28	-0.20 to +0.57	$+0.26 \pm 0.18$	+0.28
steelwork fly ash	18	-1.16 to +0.29	-0.07 ± 0.41	+0.04
biofuel burning aerosol	11	-1.23 to +0.21	-0.28 ± 0.39	-0.15
municipal waste fly ash	2	+0.08 to +0.32	$+0.20 \pm 0.12$	+0.20
heavy oil bottom ash	1	+0.38	/	/
urban particulate matter	1	+0.08	/	/

296

3.3 Isotopic composition of anthropogenic and combustion Fe

3.3.1 Power plant coal fly ash

For the 28 power plant coal fly ash samples we investigated, $\delta^{56}\text{Fe}$ ranged from -0.20‰ to $+0.57\text{‰}$ (Table S3), showing considerable variation, and the median and average values were found to be $+0.28\text{‰}$ and $+0.26 \pm 0.18\text{‰}$ (1σ) (Table 3). The three coal fly ash samples Mead et al. (2013) investigated had $\delta^{56}\text{Fe}$ values of $+0.22 \pm 0.10\text{‰}$, $+0.22 \pm 0.06\text{‰}$, and $+0.61 \pm 0.08\text{‰}$, with the average being $+0.35 \pm 0.23\text{‰}$. Li et al. (2022) found $\delta^{56}\text{Fe}$ to be



304 $+0.05 \pm 0.08\text{‰}$, $+0.20 \pm 0.07\text{‰}$, and $+0.75 \pm 0.01\text{‰}$ for one Chinese and two American coal fly
305 ash samples, with an average value of $+0.33 \pm 0.37\text{‰}$. Our measured $\delta^{56}\text{Fe}$ values have large
306 overlaps with these reported by the two previous studies (Mead et al., 2013; Li et al., 2022),
307 although the average $\delta^{56}\text{Fe}$ we reported is slightly lower.

308 As shown in Table 3, our work suggests that the $\delta^{56}\text{Fe}$ values have substantial overlaps
309 for power plant coal fly ash (-0.20‰ to $+0.57\text{‰}$) and desert dust ($+0.01\text{‰}$ to $+0.30\text{‰}$), while
310 the average $\delta^{56}\text{Fe}$ is higher for coal fly ash ($+0.26 \pm 0.18\text{‰}$) than desert dust ($+0.14 \pm 0.10\text{‰}$).
311 This implies that coal fly ash Fe could be isotopically heavier (at least not lighter) than desert
312 dust Fe.

313 3.3.2 Steelwork fly ash

314 The 21 steelwork fly ash samples we studied showed large variation in $\delta^{56}\text{Fe}$ (-1.16‰ to
315 $+0.43\text{‰}$) (Table S4), with average and median values being $+0.00 \pm 0.42\text{‰}$ (1σ) and $+0.09\text{‰}$.
316 Among the 21 samples, $\delta^{56}\text{Fe}$ were measured to be $+0.39\text{‰}$, $+0.42\text{‰}$, and $+0.43\text{‰}$ for the three
317 coking fly ash samples, with an average of $+0.42 \pm 0.04\text{‰}$ (1σ), being much higher than the
318 other 18 steelwork fly ash samples. This difference can be attributed to the fact that coking
319 process (during which coal is pyrolyzed in an oxygen-free environment to produce coke) uses
320 coal as the main raw material (Keboletse et al., 2021), and thus Fe composition of the generated
321 fly ash is different from that produced in other steelwork processes. In fact, the average $\delta^{56}\text{Fe}$
322 ($+0.42 \pm 0.04\text{‰}$, 1σ) we determined for coking fly ash is rather close to that for power plant coal
323 fly ash ($+0.26 \pm 0.18\text{‰}$, 1σ), implying that they originated from the same raw material (i.e. coal).

324 For the other 18 steelwork fly ash samples we examined, $\delta^{56}\text{Fe}$ ranged from -1.16‰ to
325 $+0.29\text{‰}$, and the median and average values were determined to be $+0.04\text{‰}$ and $-0.07 \pm 0.41\text{‰}$
326 (1σ) (Table 3). The average $\delta^{56}\text{Fe}$ value was reported to be $+0.08 \pm 0.24\text{‰}$ ($n=10$) (Flament et
327 al., 2008) and $-0.12 \pm 0.08\text{‰}$ ($n=4$) for steelwork fly ash (Maters et al., 2022), aligning fairly
328 well with our result. Compared to desert dust, although a few samples were enriched in



329 isotopically heavier Fe, most steelwork fly ash in general shows slightly lower or similar $\delta^{56}\text{Fe}$
330 values; therefore, aerosol Fe emitted by steelworks may be a potential source of isotopically
331 light Fe observed in the tropospheric aerosols.

332 3.3.3 Biofuel burning aerosol

333 This study determined the Fe isotopic composition of 11 biofuel burning aerosol samples
334 (Table S5). As mentioned in Section 2.1.2, biofuel burning experiments conducted in this work
335 are designed to simulate domestic biofuel burning, and may not be representative of wildfires,
336 which can also entrain soil particles into the atmosphere (Hamilton et al., 2022; Bunnell et al.,
337 2025). The lowest and highest $\delta^{56}\text{Fe}$ values (-1.23‰ versus $+0.21\text{‰}$) were found for pear wood
338 and soybean straw burning aerosol, respectively. As shown in Table 3, the median and average
339 $\delta^{56}\text{Fe}$ were determined to be -0.15‰ and $-0.28 \pm 0.39\text{‰}$ (1σ), lower than that of desert dust
340 ($+0.14 \pm 0.10\text{‰}$). Most biofuel burning aerosol samples we examined exhibited $\delta^{56}\text{Fe}$ below
341 0‰ , showing significant enrichment of isotopically lighter Fe. In contrast, a previous study
342 (Kurusu and Takahashi, 2019) found average $\delta^{56}\text{Fe}$ to be $+0.09 \pm 0.03\text{‰}$ for reed residual ash
343 (and $+0.08 \pm 0.10\text{‰}$ for reed), being very similar to the UCC and thus indicating no obvious
344 enrichment of lighter Fe.

345 Plants use Strategy I or II to absorb Fe from soil (Kobayashi and Nishizawa, 2012). It was
346 suggested that compared to the soil where plants are grown, Strategy I plants are usually
347 enriched in isotopically lighter Fe while Strategy II plants are relatively enriched in the heavier
348 Fe (Guelke and Von Blanckenburg, 2007). Eight Strategy I plants, namely peanut, soybean,
349 birch, lychee wood, apple wood, pear wood, fir, and pine, were examined in our work, and the
350 average $\delta^{56}\text{Fe}$ was determined to be $-0.22 \pm 0.47\text{‰}$ (1σ , $n=8$) for the corresponding biofuel
351 burning aerosols. We also investigated three Strategy II plants, namely wheat, corn, and rice
352 straw, and the average $\delta^{56}\text{Fe}$ was determined to be $-0.33 \pm 0.28\text{‰}$ (1σ , $n=3$) for the
353 corresponding biofuel burning aerosol. Based on the data our work obtained, our work suggests



354 that $\delta^{56}\text{Fe}$ values of biofuel burning aerosols generated from Strategy I plants are not
355 significantly different from those for Strategy II plants ($p>0.05$). Our result seems to deviate
356 from what was reported by [Guelke and Von Blanckenburg \(2007\)](#), probably because biofuel
357 we studied were grown from different soils which may have different Fe isotopic compositions
358 ([Johnson et al., 2020](#)). In addition, the number of biofuel burning samples analyzed in our study
359 is relatively limited, and further measurements are required to confirm our finding.

360 **3.3.4 Other samples**

361 For the two municipal incineration fly ash samples we examined, $\delta^{56}\text{Fe}$ were measured to
362 be $+0.32\pm 0.06\text{‰}$ and $+0.08\pm 0.13\text{‰}$ (Table 3), respectively, with an average value of
363 $+0.20\pm 0.12\text{‰}$ (1σ). [Kurisu et al. \(2016b\)](#) found $\delta^{56}\text{Fe}$ to be $-0.08\pm 0.09\text{‰}$ and $-0.10\pm 0.03\text{‰}$ for
364 municipal incineration bottom and fly ash from Japan, slightly lower than our result. [Li et al.](#)
365 [\(2022\)](#) found $\delta^{56}\text{Fe}$ to be $+0.10\pm 0.08\text{‰}$ for a European municipal incineration fly ash sample
366 (BCR-176R), close to that obtained in our current study. Overall, available studies suggest
367 relatively large variations in $\delta^{56}\text{Fe}$ for municipal incineration fly ash, ranging from -0.10‰ to
368 $+0.32\text{‰}$.

369 The $\delta^{56}\text{Fe}$ value was measured to be $+0.38\pm 0.13\text{‰}$ for the heavy oil bottom ash sample
370 we examined (Table 3), similar to that for oil fly ash ($+0.30\pm 0.17\text{‰}$, $n=4$) reported by [Mead et](#)
371 [al. \(2013\)](#). NIST 1648a and NIST 1649a are both certificated urban particulate matter provided
372 by NIST. The $\delta^{56}\text{Fe}$ was measured by our work to be $+0.08\pm 0.13\text{‰}$ for NIST 1648a (Table 3),
373 in good agreement with those reported for NIST 1649a ($+0.00\pm 0.03\text{‰}$ and $+0.01\pm 0.12\text{‰}$,
374 respectively) by two previous studies ([Beard et al., 2003a](#); [Mead et al., 2013](#)).

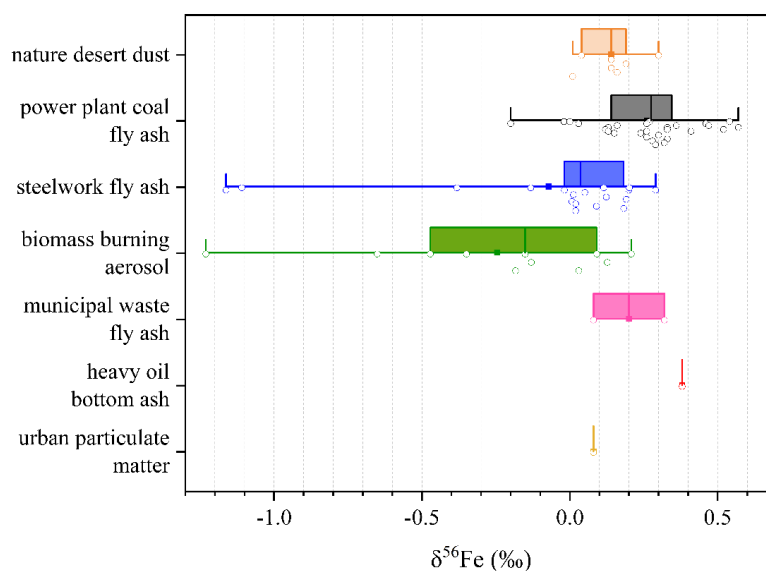
375 **3.4 Discussion**

376 **3.4.1 Atmospheric implications**

377 Figure 3 summarizes the Fe isotopic composition of natural desert dust and several
378 important anthropogenic particles our work investigated. [Mead et al. \(2013\)](#) proposed biomass



burning as the most likely source of isotopically light Fe observed in the troposphere, since plant matter is the sole material known to contain light Fe. Nevertheless, at that time no measurement of $\delta^{56}\text{Fe}$ endmember values was available. Here we found that $\delta^{56}\text{Fe}$ values ranged from -1.23‰ to +0.21‰ (average: $-0.28 \pm 0.39\text{‰}$, 1σ) for the 11 biofuel burning aerosol samples examined in this work, lower than natural desert dust. Thus, our work suggests that biofuel burning aerosol could be an potentially important source of light Fe in the troposphere, providing experimental results to support what Mead et al. (2013) proposed. The lowest $\delta^{56}\text{Fe}$ we reported for domestic biofuel burning was -1.23‰, while $\delta^{56}\text{Fe}$ could be lower than -3‰ for soluble Fe in tropospheric aerosols (Kurusu et al., 2016a; Hsieh and Ho, 2024). This indicates that biofuel burning alone cannot explain the very light Fe observed for tropospheric aerosols, i.e. other sources/processes are needed.



390

Figure 3. Summary of $\delta^{56}\text{Fe}$ values measured in our work for natural desert dust (n=7), power plant coal fly ash (n=28), steelwork fly ash (n=18, and the three samples from the coking process were not included), biofuel burning aerosol (n=11), municipal waste fly ash (n=2),



394 heavy oil bottom ash (n=1), and urban particulate matter (n=1).

395

396 In addition to domestic biofuel burning, there are other types of biomass burning, such as
397 wildfire and open burning of agricultural straws. Our study represents domestic biofuel burning
398 well. On the other hand, soil and biomass Fe both contribute substantially to aerosol Fe emitted
399 by wildfires ([Kurusu and Takahashi, 2019](#); [Hamilton et al., 2022](#)), and therefore wildfire aerosol
400 Fe may have $\delta^{56}\text{Fe}$ endmember values different from that we report for domestic biofuel
401 burning aerosol.

402 The 28 power plant coal fly ash samples we examined had $\delta^{56}\text{Fe}$ in the range of -0.20‰
403 to +0.57‰ (average: $+0.26 \pm 0.18\text{‰}$, 1σ), indicating that power plant coal fly ash is usually
404 enriched in heavier Fe. As a result, we suggest that not all the anthropogenic aerosol Fe is
405 isotopically lighter than desert dust Fe, in contrast to what previous studies usually assumed.
406 Tropospheric aerosol Fe typically exhibits similar or lower $\delta^{56}\text{Fe}$ values relative to desert dust;
407 nevertheless, higher values have been reported in a few studies ([Labatut et al., 2014](#); [Bunnell](#)
408 [et al., 2025](#); [Camin et al., 2025](#)), the causes of which are still under debate. Based on $\delta^{56}\text{Fe}$
409 endmember values our work reported, we suggest that coal-fired power plant emission can
410 potentially (at least partly) explain the higher $\delta^{56}\text{Fe}$ values observed for aerosol Fe in the
411 troposphere.

412 The average $\delta^{56}\text{Fe}$ was $-0.07 \pm 0.41\text{‰}$ (1σ) for the 18 steelwork fly ash samples we
413 examined (excluding the three samples collected from the coking process). Compared to desert
414 dust, most steelwork fly ash samples had slightly lower or similar $\delta^{56}\text{Fe}$ (ranging from -1.16‰
415 to +0.29‰) (Figure 3), and thus may contribute to lighter Fe observed for tropospheric aerosols.

416 Compared to desert dust, municipal incineration fly ash, heavy oil combustion bottom ash,
417 and urban particulate matter our work examined had similar or higher $\delta^{56}\text{Fe}$ (Figure 3), but the
418 small sample sizes preclude us from drawing further conclusions.



419 3.4.2 Recommendations for future work

420 Biofuel burning aerosol samples this work used were aerosol particles emitted by biofuel
421 burning in a commercial stove, thereby being of direct atmospheric relevance. Instead of
422 aerosol particles, fly ash samples were used to represent particulate matters emitted by coal-
423 fired power plants and steelwork. Their volume mean diameters were mostly a few tens of μm
424 (Li et al., 2025), considerably larger than these for tropospheric aerosols (typically a few μm
425 or less). Kurisu et al. (2016b) found $\delta^{56}\text{Fe}$ to be much lower for TSP than fly ash collected in
426 an incinerator, possibly due to enrichment of lighter Fe in aerosol particles caused by the
427 evaporation-condensation process in combustion. This hypothesis was further supported by a
428 following study (Kurisu et al., 2019), which found that $\delta^{56}\text{Fe}$ was much lower in the fine mode
429 ($<1.3\mu\text{m}$) than the coarse mode ($>1.3\mu\text{m}$) for aerosol particles collected at a site which was ~ 4
430 km from a steel plant. If the hypothesis proposed by Kurisu et al. (2016b) is correct, then the
431 actual $\delta^{56}\text{Fe}$ endmember values for coal-fired power plant and steelwork emission could be
432 lower than these reported in our work.

433 It is noted that an earlier study (Flament et al., 2008) did not support what was proposed
434 by Kurisu et al. (2016b). In fact, Flament et al. (2008) observed no significant Fe fractionation
435 during steelwork processes, and found the average $\delta^{56}\text{Fe}$ of total Fe to be $+0.14\pm 0.11\%$ (1σ ,
436 $n=6$) for aerosol particles collected at a site which was ~ 5 km from a major steelwork plant,
437 being similar to (or even slightly larger than) that for desert dust Fe. Further measurements of
438 $\delta^{56}\text{Fe}$ values of aerosol particles emitted by anthropogenic sources, especially size-resolved
439 aerosol particles, can be very valuable.

440 4 Conclusion

441 Aerosol Fe, emitted from natural and anthropogenic sources, has remarkable impacts on
442 atmospheric chemistry, human health and marine biogeochemistry, while quantitative source
443 apportionment of total and soluble aerosol Fe is still a big challenge. In the two decades Fe



isotopes have been increasingly utilized for source apportionment of aerosol Fe, showing great advantages. The $\delta^{56}\text{Fe}$ endmember values for anthropogenic aerosol Fe are critical for using Fe isotopes for apportionment of aerosol Fe, but are poorly constrained for anthropogenic emissions due to the lack of experimental measurements.

In this work, we measured and reported isotopic composition of aerosol Fe from several important anthropogenic sources. For the seven desert dust samples we investigated, $\delta^{56}\text{Fe}$ values ranged from +0.01 to +0.30‰ with an average value of $+0.14 \pm 0.10$ ‰ (1 σ), supporting that Fe isotopic composition of desert dust is rather homogeneous and close to that of crustal materials ($+0.09 \pm 0.10$ ‰). Compared to desert dust, the average $\delta^{56}\text{Fe}$ was determined to be $+0.26 \pm 0.18$ ‰ (1 σ) for power plant coal fly ash (n=28). This implies that not all the anthropogenic Fe is isotopically lighter than desert dust Fe, and that coal-fired power plant emission can be one potential reason to explain the higher $\delta^{56}\text{Fe}$ values observed for aerosol Fe in the troposphere. The average $\delta^{56}\text{Fe}$ was found in our work to be -0.07 ± 0.41 ‰ (1 σ) for steelwork fly ash (n=18), being slightly lower than desert dust Fe; it was reported to be -0.28 ± 0.39 ‰ (1 σ) for biofuel burning aerosol (n=11), considerably lower for desert dust. As a result, one may conclude that emission from steelwork and biofuel burning both could be important sources for the lighter Fe observed in the troposphere.

Based on Fe isotopic data, previous studies usually utilized the two-component mixing model for aerosol Fe source apportionment (Conway et al., 2019; Hsieh and Ho, 2024; Kurisu et al., 2024; Shuai et al., 2025; Kurisu et al., 2026). The two-component mixing model assigned a single $\delta^{56}\text{Fe}$ endmember value to the anthropogenic source. Our work reveals large differences in the average $\delta^{56}\text{Fe}$ values among power plant coal fly ash, steelwork fly ash, and biofuel burning aerosol; furthermore, substantial variations in $\delta^{56}\text{Fe}$ values are also observed among individual samples within each type of these anthropogenic sources. Compared to the simple two-component mixing model, the Bayesian MixSIAR model (Wang et al., 2022; Wei



469 [et al., 2024](#)), which can incorporate $\delta^{56}\text{Fe}$ endmember values for multiple sources and account
470 for their associated uncertainties, could offer distinct advantages for the quantification of
471 aerosol Fe sources.

472 Our work also has some caveats. In addition to domestic biofuel burning, there are other
473 types of biomass burning, such as wildfire and open burning of agricultural straws. Since
474 wildfires can entrain both soil and biomass Fe into the atmosphere ([Kurisu and Takahashi, 2019](#);
475 [Hamilton et al., 2022](#)), wildfire aerosol Fe may exhibit $\delta^{56}\text{Fe}$ endmembers which differ from
476 those for domestic biofuel burning. We also note that the lowest $\delta^{56}\text{Fe}$ endmember value
477 measured in our work was only -1.23‰ (Table 3), implying that other sources/processes are
478 needed to explain the very low $\delta^{56}\text{Fe}$ values (sometimes lower than -3‰) aerosol Fe in the
479 troposphere. Furthermore, our work only measured $\delta^{56}\text{Fe}$ endmember values of total aerosol
480 Fe from different sources. Previous studies have generally assumed that for any given source,
481 the $\delta^{56}\text{Fe}$ endmember values are identical for total and soluble Fe. This assumption, however,
482 is not necessarily valid since isotopic fractionation may occur during dissolution; therefore,
483 simultaneous measurements of isotopic composition of total and soluble Fe are warranted for
484 various sources. Finally, as discussed in Section 3.4.2, compared to aerosol particles in the
485 troposphere, power plant coal fly ash and steelwork fly ash samples we examined have
486 considerably larger diameters, and thus our result may not be fully representative. Experimental
487 measurements of $\delta^{56}\text{Fe}$ values for aerosol particles emitted by anthropogenic sources, and size-
488 resolved measurements in particular, would be very valuable in providing reliable constraint
489 on $\delta^{56}\text{Fe}$ endmember values.

490

491 **Data availability.**

492 Data used in this work can be found in the manuscript or the supplement.

493 **Author contribution.**



494 MT designed this study; YZ, GZ, RL, TZ, YC, and JM conducted experimental work; ML
495 and YY provided key samples used in this work; XM and MT secured funding resources; YZ,
496 GZ, JM, and MT analyzed the result and wrote the manuscript; all the authors reviewed and
497 approved the manuscript.

498 **Competing interests.**

499 At least one of the (co-)authors is a member of the editorial board of Atmospheric
500 Chemistry and Physics.

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