

Response to Reviewer 2 Comments

We sincerely thank the reviewer for providing valuable comments and helpful suggestions. We do feel that these comments have helped us to perform more in-depth analyses and improve the quality of this manuscript greatly after careful revision. The response to each comment is listed below. The original comments from reviewer are in normal font, followed by our responses in blue, and the *marked changes in the revised manuscript are in red and italic*. Page and line numbers refer to those in the revised manuscript (the clean version).

Reviewer #2 General comment: This manuscript presents a well-conducted study on atmospheric iron sources in the Northwest Pacific using innovative shipboard online measurements. The research addresses an important knowledge gap in understanding non-dust iron contributions to marine ecosystems. The multi-cruise approach provides a compelling dataset to examine spatiotemporal patterns. The key findings that non-dust sources dominate soluble Fe supply, and that ship emissions can be the primary source of soluble Fe in summer—are clearly demonstrated and have important implications for modeling marine primary productivity. However, the revisions suggested below are aimed at further improving the clarity, precision, and impact of the paper to meet the high standards of ACP:

Response to Reviewer #2 General comment: We sincerely thank the reviewer for this thorough and positive comment. We appreciate the recognition of the value of the multi-cruise dataset and the implications for marine primary productivity modeling. To respond, we have carefully revised the manuscript following the specific suggestions below to further improve the clarity, precision, and overall impact of the manuscript. Our point-by-point responses are presented below.

Major comments

Reviewer #2 Major comment No.1: The most significant issue in this manuscript is that the seasons of two field campaigns collected aerosol samples with identical particle size are different, while two other campaigns conducted in similar seasons collected aerosol samples with different particle sizes. This compromises the scientific validity of comparative analyses between campaigns, yet the manuscript frequently discusses results from different campaigns together without addressing this critical limitation. For example, Section 3.23 does not differentiate between $PM_{2.5}$ and PM_{10} when discussing non-dust source contributions to dissolved Fe, which is scientifically inappropriate given the distinct source characteristics of coarse and fine particles. A comprehensive review and revision of the text is required to prevent the indiscriminate mixing of

39 NWP1 and NWP2 results without explicitly acknowledging the differences in particle
40 size.

41 **Response to Reviewer #2 Major comment No.1:** We thank the reviewer for raising
42 this important concern. We fully agree that the differences in both sampling season and
43 particle size among the three campaigns should be clearly acknowledged in the
44 discussion. Based on the reviewer’s comment, we conducted a comprehensive review
45 and revision of the manuscript, including (1) clearly distinguishing the results derived
46 from PM_{2.5} and PM₁₀ samples in the discussion, and (2) explicitly showing particle size
47 information in relevant figures (Figs. 6, 9, 10, and 11).

48 (1) We revised the Results and discussion section to prevent the indiscriminate mixing
49 of NWP1 and NWP2 results. Specifically, changes have made for statements in
50 Sections 3.2.3 (Page 22, Lines 549–551; Page 23, 576–577), 3.3.1 (Page 25, Lines 628-
51 633), and 3.3.2 (Page 27, Lines 697–699; Page 28, Lines 713–715; Page 29, Lines 723–
52 729).

53 For example, the discussion of Fe sources has been revised to differentiate PM₁₀ and
54 PM_{2.5} samples:

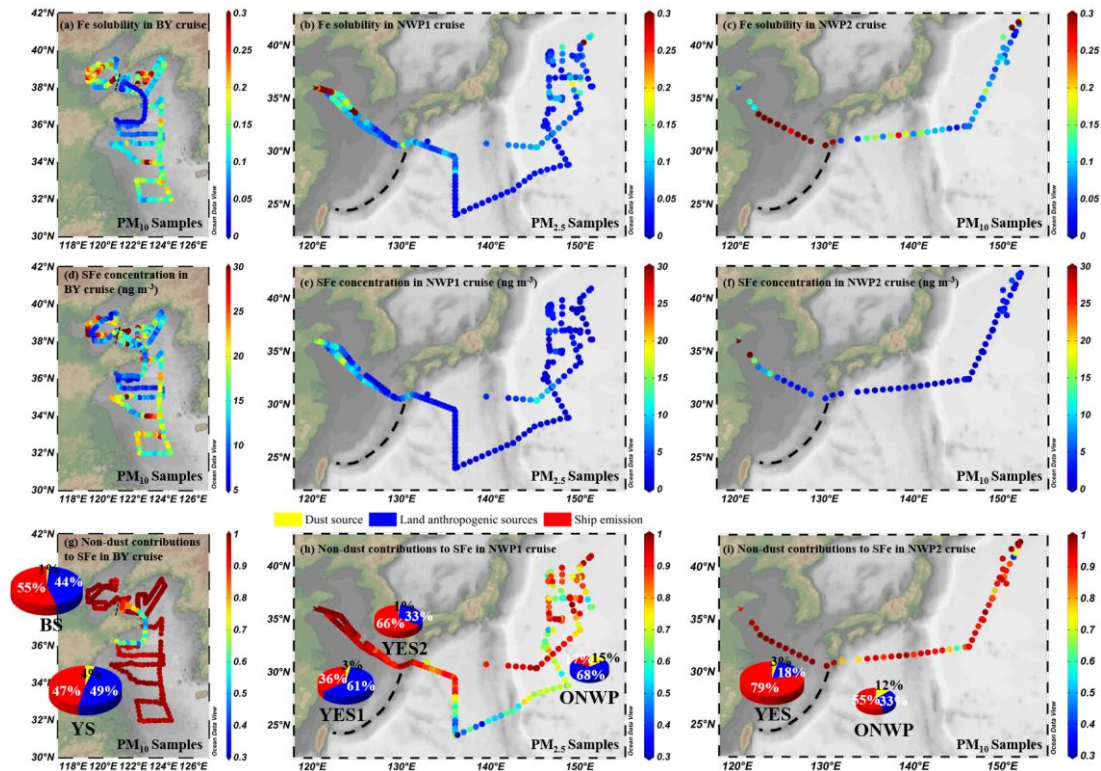
55 ➤ Page 27, Lines 697–699:

56 “Overall, dust remained the dominant source of total Fe, accounting for 59%–76% in
57 three PM₁₀-sampling cruise segments (BY, coastal NWP2, and open NWP2) and 44%–
58 70% in two PM_{2.5}-sampling cruise segments (coastal and open NWP1). For both size
59 fractions, dust contributions were higher in the open Northwest Pacific ($\geq 70\%$) than
60 in coastal sea regions.”

61 ➤ Page 28, Lines 713–715:

62 “However, non-dust sources, including land anthropogenic sources and ship emission,
63 dominated atmospheric soluble Fe consistently in this study, with contributions to
64 soluble Fe reaching 88%–97% in three PM₁₀-sampling cruise segments and 85%–98%
65 in two PM_{2.5}-sampling cruise segments.”

66
67 (2) All figures with data from multiple cruises have been revised to clearly indicate the
68 particle size fraction associated with each campaign, including Figures 6, 9, 10, and 11.
69 For instance, Figure 9 has been updated to show that PM₁₀ samples were collected
70 during the BY and NWP2 cruises and PM_{2.5} samples were collected during the NWP1
71 cruise.



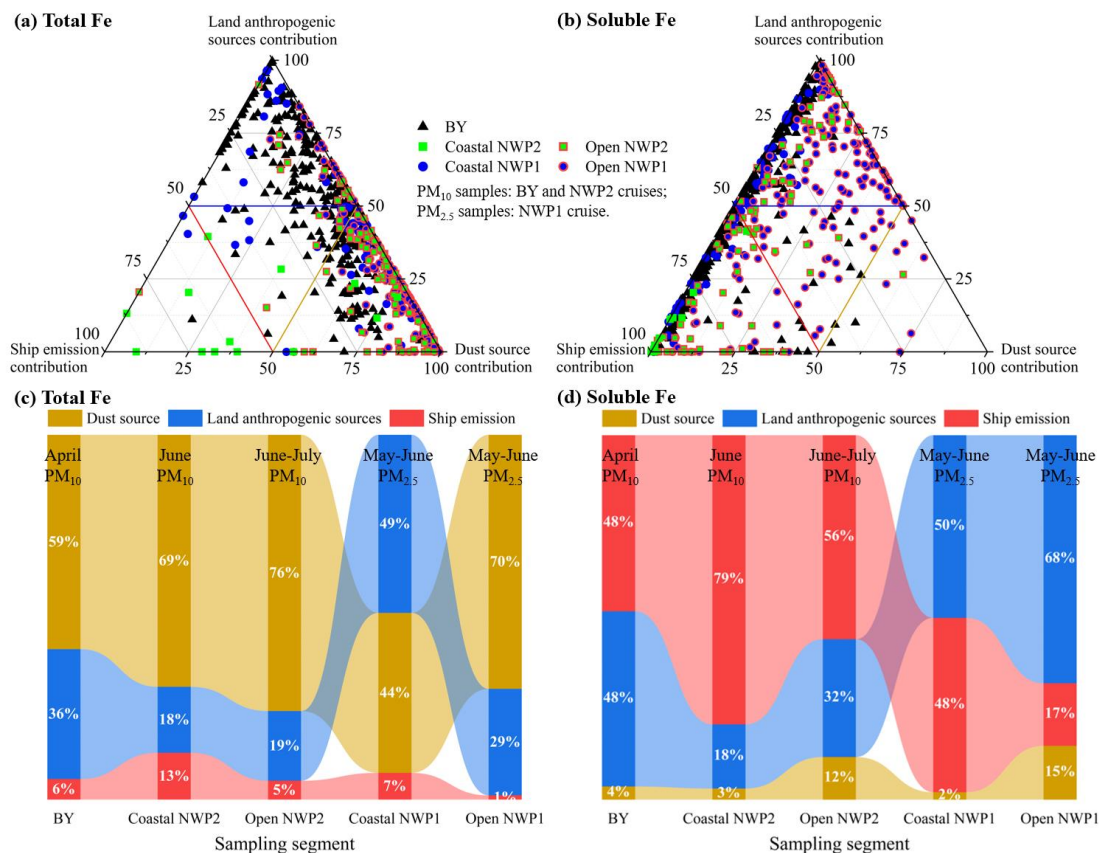
72
 73 **Figure 9.** Iron solubility, soluble Fe concentration, and soluble Fe sources in three cruises. (a–c)
 74 Coloured dots represent atmospheric Fe solubility (*dimensionless*) for the BY cruise (*PM₁₀*
 75 *samples*), NWP1 cruise (*PM_{2.5} samples*), and NWP2 cruise (*PM₁₀ samples*), respectively. (d–f)
 76 Coloured dots represent atmospheric soluble Fe concentrations (ng m^{-3}) for the three cruises. (g–i)
 77 Coloured dots represent the fractional contribution of non-dust sources to soluble Fe
 78 (*dimensionless*). Pie charts in (g–i) depict the average contributions of different sources to soluble
 79 Fe in different cruise legs.

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 82 **Reviewer #2 Major comment No.2:** Similarly, this issue manifests in Figure 11. It is
 83 unclear whether the "marginal seas" referred to in Figures 11b and 11d include both the
 84 Yellow and Bohai Seas campaigns and the coastal segments of NWP1/NWP2, and
 85 whether the data presented encompass both $\text{PM}_{2.5}$ and PM_{10} results. If this figure
 86 combines $\text{PM}_{2.5}$ and PM_{10} data, such presentation is inappropriate. Additionally, the
 87 criteria used to define "spring" and "summer" seasons in this figure require clarification.

88 **Response to Reviewer #2 Major comment No.2:** We thank the reviewer for this
 89 important comment. In the original Figure 11, the term "marginal seas" indeed included
 90 both the Yellow and Bohai Seas campaigns as well as the coastal segments of NWP1
 91 and NWP2. The data presented in original figure encompassed results from both $\text{PM}_{2.5}$
 92 and PM_{10} . Spring and summer season refer to period of March–May and June–August,
 93 respectively.

94 We agree with the reviewer that the original figure mixed $\text{PM}_{2.5}$ and PM_{10} results
 95 is misleading. Based on this comment, we have revised Figure 11 to display $\text{PM}_{2.5}$ and

96 **PM₁₀ samples separately. The updated figure provides the sampling month and particle**
 97 **size information inside the figure.**



98
 99 **Figure 11.** Source structure of total Fe and soluble Fe across different *cruises and sea areas*. (a)
 100 and (b) are ternary plots illustrating the source structure of total Fe and soluble Fe, respectively;
 101 the three vertices represent the three sources; the closer a sample point is to a given vertex, the
 102 the greater the relative contribution of that source to the sample. (c) and (d) show the average
 103 contributions of the three sources to total Fe and soluble Fe, respectively, across different
 104 *sampling segments*. *BY* refers to all samples collected during the *BY* cruise. *Coastal NWP2* (or
 105 *NWP1*) represents samples collected from coastal regions, including the Yellow Sea and East
 106 China Sea, during the *NWP2* (or *NWP1*) cruise, while *Open NWP2* (or *NWP1*) refers to samples
 107 collected from the open Northwest Pacific.

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110 **Reviewer #2 Major comment No.3:** In the abstract, the quantitative claims " Results
 111 showed non-dust emissions contributed substantially to atmospheric Fe, accounting for
 112 21%–48% of total Fe across different regions and seasons. Importantly, their
 113 contributions to soluble Fe were significantly higher, reaching 79%–98% and largely
 114 dominating the bioavailable Fe supply in the study area." are not supported by
 115 corresponding data in the Results and Discussion Sections. The same issue occurs in
 116 the Conclusions section. The source of these numerical values remains unclear, so does
 117 their specific association with either PM_{2.5} or PM₁₀.

118 **Response to Reviewer #2 Major comment No.3:** We would like to clarify here that
119 the numerical values in the abstract and conclusion sections were derived from the
120 original Figure 11, in which the contribution of non-dust emissions was calculated as
121 the sum of land anthropogenic sources and ship emission. These values included both
122 PM_{2.5} and PM₁₀ results, which may lead to confusion.

123 Therefore, in the revised manuscript, we have updated Figure 11, and revised the
124 relevant statements to clearly distinguish source contributions to Fe in PM_{2.5} and PM₁₀
125 samples. Correspondingly, the quantitative statements in both the Abstract and
126 Conclusion have been revised to show these changes.

127 (1) Abstract section (Page 1, Lines 22-25):

128 “Results showed non-dust emissions *were notable contributors* to atmospheric *total* Fe,
129 *representing 24%–41% of total Fe in PM₁₀ and 30%–56% in PM_{2.5} samples across*
130 *different cruise segments*. Importantly, their contributions to soluble Fe were
131 significantly higher, reaching *88%–97% in PM₁₀ and 85%–98% in PM_{2.5} samples.*”

132 (2) Conclusion section (Page 30, Lines 752-756):

133 “Among *non-dust sources*, land anthropogenic sources contributed substantially to both
134 total Fe and soluble Fe, *with its highest contribution being 49% to total Fe in coastal*
135 *PM_{2.5} and 68% to soluble Fe in open Northwest Pacific PM_{2.5}, both were observed*
136 *during the NWP1 cruise*. In contrast, the contribution of ship emission to total Fe was
137 relatively minor, yet it was a notable source of soluble Fe, *reaching a mean contribution*
138 *of 79% in coastal PM₁₀ samples during the NWP2 cruise.*”

139

140 Additionally, we would like to clarify that the values reported in the Abstract and
141 Conclusion are consistent with the data presented in the revised Figure 11, as well as
142 with the statements in Section 3.3.2 (Pages 27-29, Lines 696–732) in the revised
143 manuscript.

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145 **Reviewer #2 Major comment No.4:** PMF method: In Text S4, the statement "The
146 value of Q relative to Q_{expected} reached its minimum at seven factors, which suggested
147 that the mathematical diagnostics favored a seven-factor solution." requires
148 clarification. What do Q and Q_{expected} specifically refer to? Are these equivalent to Q_{true}
149 and Q_{robust} mentioned in the preceding sentence? What's the methodological basis or
150 reference for determining the optimal factor number using the minimum Q/Q_{expected}
151 value? Additionally, have the authors evaluated whether PMF solutions with 4 or 5
152 factors might yield higher physical interpretability?

153 **Response to Reviewer #2 Major comment No.4:** We thank the reviewer for this
154 comment. In the statement “The value of Q relative to Q_{expected} reached its minimum at
155 seven factors...”, the first Q refers specifically to Q_{robust}, and the second one (Q_{expected})
156 is calculated according to the following equation:

157
$$Q_{expected} = n \times m - p \times (n + m) \quad (R1)$$

158 Where n is the number of chemical species (only those marked as strong in the model
159 are included), m is the sample size, p is the number of factors.

160 The reference for determining the optimal factor number using the minimum
161 $Q_{robust}/Q_{expected}$ value is the EPA PMF 5.0 User Guide (available at:
162 https://www.epa.gov/sites/default/files/2015-02/documents/pmf_5.0_user_guide.pdf)
163 (Gary et al., 2014).

164 Regarding the PMF solutions with four or five factors mentioned by the reviewer,
165 we have further provided a more detailed comparison of the 3–5 factor solutions,
166 highlighting why the three-factor solution was ultimately selected. As shown in the
167 newly added Tables S1 and S2, the characteristic elements of each factor in the 3-5
168 factor solutions regarding PM₁₀ samples and PM_{2.5} samples are presented, respectively.

169 According to Table S1, the three factors correspond to dust, ship emission, and
170 land-based anthropogenic sources, respectively. When transitioning from a three-factor
171 to a four-factor solution, a factor characterized by only Ca emerged. While Ca may
172 originate from sea salt in addition to dust source, this factor shows only a weak
173 correlation with wind speed (correlation coefficient of 0.11), making it unlikely to
174 represent a sea spray source. Additionally, when moving from the four-factor to the
175 five-factor solution, the factor of land anthropogenic source appeared to split into two
176 factors, neither of which could be clearly attributed to a specific source. Therefore, the
177 three-factor solution was selected, as it identified distinct and interpretable sources.

178 Table S1. Characteristic elements in each factor from PMF analysis of PM₁₀ sample

	Three-factor solution	Four-factor solution	Five-factor solution
Factor 1	Ca, Ba, Fe, V, Mn ^a	Ba, Ca, Fe, V, Mn	Ba, Ca, Fe, V, Mn
	Dust source ^b , 59.3% ^c	Dust source, 56.0%	Dust source, 54.5%
Factor 2	Ni, V	Ni, V	Ni, V
	Ship emission, 5.7%	Ship emission, 3.7%	Ship emission, 3.2%
Factor 3	As, Pb, Zn, Se, Cu, Mn, Ba, Fe	As, Zn, Pb, Se, Cu, Mn, Fe, Ba	As, Se, Zn, Mn
	Land anthropogenic source, 35.0%	Land anthropogenic source, 38.5%	Coal burning? Industry? 28.1%
Factor 4		Ca	Ca
		?, 1.8%	?, 0.9%
Factor 5			Pb, Cu, Ba
			Industry? Brake and tire wear? 13.4%

179 ^a Characteristic elements in a factor are defined as those where the factor's contribution to the element exceeds
180 30%, and the elements are sorted by factor contribution;

181 ^c Emission sources are named according to the factor profiles, with "?" indicating sources that are difficult to
182 identify;

183 ^c The value represents the contribution of the factor to the target element Fe.

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185 As shown in Table S2 for PM_{2.5} samples, the characteristic elements of each factor
186 in the 3-5 factor solutions again highlight dust, ship, and land anthropogenic sources
187 form the most interpretable combination of factors.

188 Table S2. Characteristic elements in each factor from PMF analysis of PM_{2.5} sample

	Three-factor solution	Four-factor solution	Five-factor solution
Factor 1	Ca, Ba, K, Fe, Cu, Mn ^a ; Dust source ^b ; 59.8% ^c	Ba, Fe, Ca, Mn, K; Dust source; 64.1%	Ba, Fe, Ca, Mn; Dust source; 57.0%
Factor 2	V, Ni; Ship emission; 3.5%	V, Ni; Ship emission; 2.2%	V, Ni; Ship emission; 0.8%
Factor 3	As, Zn, Se, Pb, Mn, Fe; Land anthropogenic source; 36.7%	As, Zn, Se, Pb, Mn; Land anthropogenic source; 22.5%	Se, As, Pb; Coal burning? Industry?; 13.7%
Factor 4		Cu, K, Ca; ?; 11.2%	Ca, K, Cu; ?; 12.3%
Factor 5			Zn, Pb, Mn, Cu; Industry? Coal burning?; 16.2%

189 ^a Characteristic elements in a factor are defined as those where the factor's contribution to the element exceeds
190 30%, and the elements are sorted by factor contribution;

191 ^b Emission sources are named according to the factor profiles, with "?" indicating sources that are difficult to
192 identify;

193 ^c The value represents the contribution of the factor to the target element Fe.

194

195 The above information has been incorporated into the Supplementary Material
196 (**Revised Supplementary Text S5, newly added Tables S1-S2**).

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198 Reference:

199 Gary, N., Rachelle, D., Steve, B., and Song, B.: EPA positive matrix factorization (PMF)
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203 [02/documents/pmf_5.0_user_guide.pdf](https://www.epa.gov/sites/default/files/2015-02/documents/pmf_5.0_user_guide.pdf) (last accessed: September 22, 2025).

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206 **Reviewer #2 Major comment No.5:** Given that aerosols in different samples
207 experience different aging processes during atmospheric transport (i.e., variable R_j
208 values in equations 3-5 should be different in different samples), this fixed-parameter

209 approach should theoretically produce less accurate results. However, Figure 8a
210 demonstrates the reliability of the fixed iron solubility parameterization scheme
211 combined with PMF results for estimating aerosol iron solubility. Does this mean that
212 atmospheric processing may have minimal modification effects on the initial iron
213 solubility of different source-derived aerosols? Please provide further detailed
214 explanation of this issue in the article.

215 **Response to Reviewer #2 Major comment No.5:** We thank the reviewer for this
216 insightful comment. As pointed out by the reviewer, aerosols in different samples may
217 experience different degree of atmospheric aging during transport, and therefore the
218 atmospheric processing term (R_j) should vary among samples. In the case of the BY
219 cruise, however, the sampling region (Bohai Sea–Yellow Sea) covers a relatively
220 limited area spatially. As a result, the variability in atmospheric processing intensity
221 (R_j) among different samples is expected to be small. This is probably the major reason
222 why the parameterized Fe solubility estimates based on fixed source-specific Fe
223 solubility ($SFe\%_{j,r}$) values remain in generally comparable to the measured values in
224 the BY cruise (Fig. 8a).

225 This does not mean that atmospheric processing has minimal modification effects
226 on the initial iron solubility of different source-derived aerosols. In fact, the Fe
227 solubility values adopted for each source category do not represent the initial solubility
228 at emission. Instead, they correspond to the solubility of source-specific Fe measured
229 at receptor sites in oceanic area, thereby reflecting the cumulative effects of
230 atmospheric processing.

231 Following the reviewer’s suggestion, we have provided detailed explanation in the
232 revised manuscript (Page 20, Lines 514–523):

233 *“Nevertheless, the measured Fe solubility during the BY cruise showed greater*
234 *variability than the calculated values. For example, for samples BY3–BY6, the*
235 *measured Fe solubility ranged from 11.5% to 18.4%, whereas the calculated values*
236 *were in a narrower range (13.2%–14.0%). These four samples were collected from the*
237 *southern, central, and northern Yellow Sea (Fig. 1b), and the greater variability in*
238 *measured solubility likely reflects slight variations in atmospheric processing influence*
239 *(R_j) and source-specific Fe solubility ($SFe\%_{j,r}$) across the Yellow Sea. Such variations*
240 *were relatively limited at this spatial scale. Therefore, using fixed $SFe\%_{j,r}$ still yield Fe*
241 *solubility comparable to observations. At larger spatial scales (especially for global*
242 *scale), however, such variations should be considered. As previously mentioned, to*
243 *account for this, dust-derived Fe was assigned $SFe\%_{j,r}$ values of 0.5% over the Chinese*
244 *marginal seas and 1.0% over the open Northwest Pacific, while land anthropogenic*
245 *and ship-derived Fe were assigned to be constants due to insufficient observational*
246 *constraints.”*

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249 **Reviewer #2 Major comment No.6:** Additionally, numerous studies have used PMF-
250 based methods to estimate Fe solubility in aerosols from different sources. While the
251 solubility estimated by these methods shows similar trends to that of source-specific
252 samples collected directly from emission sources, there are significant discrepancies in
253 absolute values. A key reason for this discrepancy is that the sources identified by PMF
254 are not pure but may instead represent mixtures of multiple pollution sources, whose
255 individual components can exhibit drastically different solubility. For example, fly ash
256 typically exhibits very low solubility, whereas some industrial sources may have much
257 higher solubility; yet these distinct sources might be lumped into a single source factor
258 in PMF analysis. Another case in point is ship emissions: in many studies, PMF-
259 estimated solubility for ship emissions is far below 70%. Given these issues, it is
260 recommended that the authors include a discussion on the limitations and uncertainties
261 of this method.

262 **Response to Reviewer #2 Major comment No.6:** We sincerely appreciate the
263 reviewer's comment regarding the limitations and uncertainties associated with the Fe
264 solubility parameterization method.

265 As pointed out by the reviewer, source factors resolved by PMF may reflect
266 mixtures of multiple emission types with potentially distinct Fe solubilities. This issue
267 is particularly relevant for the land anthropogenic source factor identified in this study.
268 Previous source testing studies (Fu et al., 2012; Oakes et al., 2012), as well as PMF-
269 based estimation method (Li et al., 2023a; 2025a), have demonstrated substantial
270 variability in Fe solubility among different land anthropogenic sources. Consequently,
271 assigning an average Fe solubility to the land anthropogenic sources inevitably
272 introduces uncertainty, which we acknowledge as a limitation of the current approach.

273 Regarding the solubility of Fe from ship emissions, it has been reported by
274 multiple source testing experiments of heavy oil combustion, including 74.1% and 85.9%
275 (Fu et al., 2012), and 77% and 81% (Schroth et al., 2009). In addition, Li et al. (2025a)
276 reported a PMF-estimated Fe solubility of 93.1% for the ship emission factor based on
277 PM_{2.5} observations in Qingdao, China. Although lower values such as 38% have also
278 been reported (Desboeufs et al., 2001), the 70% ship-derived Fe solubility adopted in
279 this study should be acceptable.

280 Following the reviewer's suggestion, we have included discussion on the
281 limitations and uncertainties of this method in the revised manuscript (Pages 19–20,
282 Lines 491–505):

283 “Observational data on the solubility of anthropogenic Fe in marine aerosols
284 remain limited. This study referenced the solubility of combustion-derived Fe (11%)
285 *over the open Northwest Pacific* reported by Kurisu et al. (2021), setting the solubility
286 of land anthropogenic Fe at 11%. *To be noted that, land anthropogenic Fe may*
287 *originate from multiple sources, such as coal combustion, industrial emissions, and*
288 *biomass burning (Chen et al., 2021; Zhang et al., 2025), with varied solubility of Fe.*
289 *For example, PMF-based source apportionment of total and soluble Fe in PM_{2.5}*
290 *collected in Qingdao, China showed fly ash with 1% solubility, while aged industrial*

291 *emissions reached 12.5% (Li et al., 2023a). Treating these diverse sources as a single*
292 *category with one assigned solubility inevitably introduces uncertainty, representing a*
293 *limitation of this approach. Future ship-based studies integrating online metal*
294 *measurements with additional chemical tracers, such as water-soluble ions and*
295 *organic species, are needed to resolve more detailed source categories.*

296 For ship emission, which primarily originates from heavy oil combustion, the
297 relatively short-range transport from ship emission sources to the receptors in marine
298 atmosphere likely resulted in limited atmospheric aging. Therefore, we referred to
299 source test results for heavy oil combustion, including 38% (Desboeufs et al., 2001),
300 74.1% and 85.9% (Fu et al., 2012), and 77% and 81% (Schroth et al., 2009). *In addition,*
301 *Li et al. (2025a) reported a PMF-derived Fe solubility of 93.1% for ship emission based*
302 *on field observations in Qingdao, China.* Based on these evidences, a ship-derived Fe
303 solubility of 70% was assumed as a conservative value in this study.”

304

305 References

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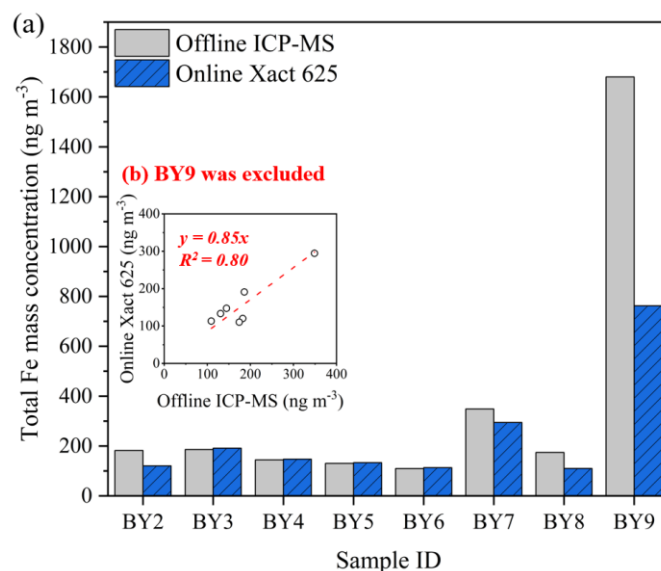
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 344 **Reviewer #2 Major comment No.7:** During the Yellow and Bohai Seas campaign,
 345 both Xact 625 measurements and filter sample analyses (ICP-MS) were employed for
 346 elements detection. How consistent are the results obtained from these two analytical
 347 methods?

348 **Response to Reviewer #2 Major comment No.7:** As suggested by the reviewer, we
 349 have added a comparison between the results of two analytical methods during the
 350 Bohai Sea and Yellow Sea (BY) campaign.

351 As shown in the newly added Fig. S2, after excluding one outlier (sample BY9),
 352 the remaining samples exhibit comparable atmospheric Fe concentrations, with an R^2
 353 of 0.80 and a slope of 0.85.



354 **Figure S2.** Comparison of atmospheric Fe concentrations measured by the online Xact 625
 355 instrument and offline filter-based inductively coupled plasma mass spectrometry (ICP-MS)
 356 analysis during the BY cruise. (a) Bar charts comparing Fe concentrations measured by the two
 357 methods for samples collected during the BY2–BY9 sampling periods. (b) Scatter plot comparing
 358 Fe concentrations measured by the two methods for samples collected during the BY2–BY8
 359 sampling periods. The BY1 filter sample is not shown because the online instrument data were
 360 incomplete during its sampling period.
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363 The only sample that exhibited a poor comparison between the online and offline
364 Fe detection methods (Figure S2) was the BY9 sample, which was collected during an
365 intense dust event (Section 3.1.1). Consequently, coarse particles experienced
366 significant losses due to inertial impaction in the sampling tubing of Xact 625, resulting
367 in substantially lower concentrations of dust-related elements in PM₁₀ measured by the
368 online Xact 625 compared with offline filter-based analyses, such as Fe (online: 762.4
369 ng m⁻³; offline: 1680 ng m⁻³) and Ca (online: 1119 ng m⁻³; offline: 1938 ng m⁻³). In
370 contrast, fine-mode anthropogenic elements showed no such discrepancy, such as Pb
371 (online: 7.88 ng m⁻³; offline: 6.18 ng m⁻³) and Cu (online: 3.54 ng m⁻³; offline: 3.57 ng
372 m⁻³).

373 This underestimation mainly affects crustal elements rather than fine-mode
374 elements and would therefore lead to an underestimation of the dust contribution in the
375 PMF analysis based on online data. However, the PMF results already indicate an
376 overwhelming dust contribution of 96% during the BY9 sampling period, supporting
377 our conclusion that Fe during this sampling period was dominated by dust source.
378 Therefore, the underestimation introduced by particle losses is considered minor and
379 will not affect the main conclusions.

380 Besides, we note that the online–offline comparison during the BY cruise was
381 affected by specific sampling conditions. During the BY cruise, the Xact 625
382 instrument was installed inside the main deck laboratory, connected to the sampling
383 inlet via a relatively long (~10 m) sampling tubing, which might result in sampling loss
384 of large particles. In contrast, during the NWP1 and NWP2 cruises, the instrument was
385 installed in a foredeck container and connected to the sampling inlet using a short,
386 vertically oriented sampling tubing of approximately 1 m, which likely minimized
387 particle losses.

388 The above explanations and discussions have been added to the Supplementary
389 Materials (**newly added Supplementary Text S1 and Figure S2**).

390

391

392 **Reviewer #2 Major comment No.8:** The authors have not contextualized their
393 findings alongside recent studies on Fe sources in Chinese coastal regions or other
394 marine environments. To strengthen the scientific significance of this research and
395 more clearly articulate its contributions, it is strongly recommended that the authors
396 undertake a comparative analysis engaging with both recent and prior literature.

397 **Response to Reviewer #2 Major comment No.8:** We thank the reviewer for this
398 insightful comment. We have expanded the discussion based on a thorough literature
399 review and have undertaken a comparative analysis to highlight consistencies and
400 differences between our work and prior studies.

401 **(1) Discussion on Fe solubility (Page 22, Lines 551-559):**

402 *“Overall, the calculated Fe solubility across the three cruises here agrees with recent*
403 *ship-based measurements of TSP in similar regions. Yang et al. (2020) reported 8%*
404 *and 15% over the Chinese marginal seas (Yellow Sea and East China Sea) and the*
405 *Northwest Pacific, respectively, while Li et al. (2025b) reported 11% ± 9.3% and 14%*
406 *± 7.7% (non-dust days) in these two regions. These results differ from global model*
407 *simulations. Using the IMPACT model, Ito et al. (2021) reported atmospheric Fe*
408 *solubility of 2%–4% over the Chinese marginal seas and 4%–6% over the open*
409 *Northwest Pacific. Hamilton et al. (2019), employing the CMA model, reported Fe*
410 *solubility of 4%–6% for both regions. Across the study domain, model-simulated Fe*
411 *solubility was consistently lower than results in our study and previous measurements,*
412 *likely reflecting the underestimation of modelled contributions from non-dust emissions*
413 *in regions strongly influenced by human activities.”*

414

415 **(2) Comparison of Fe source apportionment results with previous PMF-based** 416 **studies**

417 ➤ Ship emission contribution; Page 15, Lines 378-380:

418 *“Similar observational results regarding the contribution of ship-derived Fe were*
419 *reported by Qi et al. (2025), with ship emission contributing 10% of atmospheric Fe in*
420 *PM_{2.5} over the Bohai and Yellow Seas in spring 2018, which is comparable to the 6%*
421 *contribution derived from PM₁₀ samples in this study.”*

422 ➤ Dust source contribution; Page 27, Lines 699-702:

423 *“These results are generally comparable to recent Fe source analysis (based on PMF)*
424 *in Qingdao, a coastal city in the Yellow Sea, where dust (including fresh and aged dust)*
425 *accounted for 71% of total Fe in spring PM_{2.5} samples (Li et al., 2025a) and 46%–80%*
426 *in coarse particles (PM_{>1}) across seasons (Chen et al., 2024).”*

427 ➤ Land anthropogenic sources contribution, Page 29, Lines 724-729:

428 *“For PM_{2.5} samples in coastal NWPI segment (May-June), the contribution to total Fe*
429 *(49%) is comparable to that reported by Qi et al. (2025) during coastal cruise in spring*
430 *(44.5% for total Fe in PM_{2.5}), but lower than their coastal cruise in summer (80.5%).*
431 *For PM_{2.5} samples in open NWPI segment, land anthropogenic sources contributed 29%*
432 *to total Fe and 68% to soluble Fe, which are slightly lower than our previous*
433 *observations on PM_{2.5} in the open Northwest Pacific during spring 2015 (36% and 77%,*
434 *respectively) (Zhang et al., 2024)”*

435 **(3) Comparison of Fe source apportionment results with previous isotope-based** 436 **studies (Pages 28-29, Lines 715-722):**

437 *“These results were comparable with previous PMF-based studies in coastal cities*
438 *(Chen et al., 2024; Li et al., 2025a), but were notably higher than recent Fe isotope–*
439 *based estimates. For example, Ho and Hsieh (2024) reported 35%–99% anthropogenic*
440 *contribution to soluble Fe in the 0.57–1.0 μm size fraction (ammonium acetate buffer*
441 *leach) at Pengjia Island, while Kurisu et al. (2024) found 0–45% in bulk aerosols*

442 *(ultrapure water leach) over the North Pacific. Isotope-based source apportionment is*
443 *highly sensitive to end-member selection. Bunnell et al. (2025) showed that using two*
444 *different anthropogenic Fe isotope end-members could result in nearly a threefold*
445 *difference in the inferred anthropogenic contribution to soluble Fe over the North*
446 *Pacific. This sensitivity and the associated uncertainties may limit the quantitative*
447 *comparability of isotope-based estimates with PMF-derived results.”*
448

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496

497

498 *Minor comments*

499 **Reviewer #2 Minor comment No.1:** total Fe but was major source of soluble Fe"
500 requires the indefinite article "a" before "major" for grammatical correctness. It should
501 read: "...but was a major source of soluble Fe."

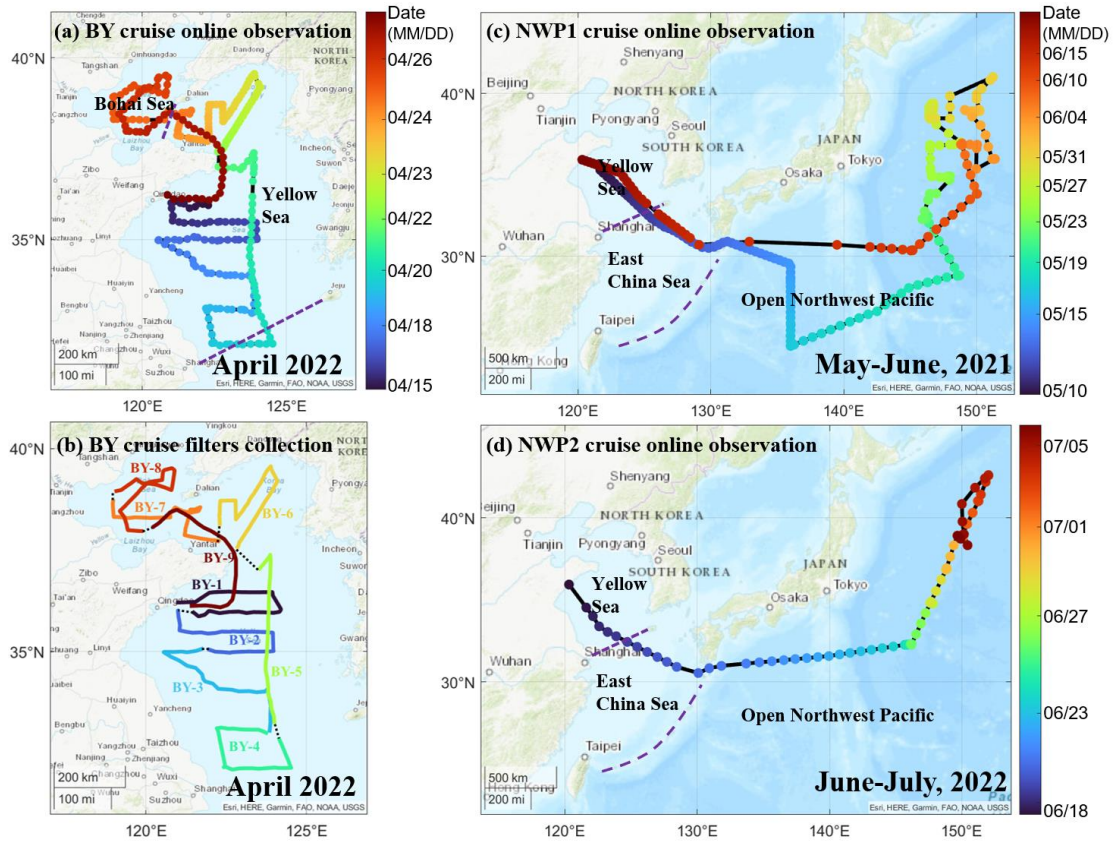
502 **Response to Reviewer #2 Minor comment No.1:** We have added "a" before "major".
503 In addition, we have carefully checked the grammar throughout the entire manuscript
504 to avoid similar errors.

505

506

507 **Reviewer #2 Minor comment No.2:** Figure 1: The cruise track should be clearly
508 marked with dates, particularly highlighting periods when the instrument was
509 operational versus malfunctioning during the NWP2 leg. If the plotted track represents
510 only the periods when the instrument was functioning normally, this must be explicitly
511 stated either in the figure caption or the main text.

512 **Response to Reviewer #2 Minor comment No.2:** This suggestion is very helpful.
513 Figure 1 has been revised to clearly indicate the cruise dates. Moreover, the figure
514 caption now explicitly states that the plotted NWP2 cruise track includes only periods
515 when the instrument was functioning normally.



516

517

Figure 1. Cruise tracks in the Chinese marginal seas and the open Northwest Pacific during 2021–2022. (a) Shipborne online observations *during the BY cruise, where each dot represents an online sample, and the colour indicates the sampling date*; (b) Filter sample collection *during the BY cruise, where each solid line represents an offline filter sample*; (c) and (d) Shipborne online observations *during the NWP1 and NWP2 cruises, respectively, analogous to panel (a)*. BY denotes the cruise conducted in 2022 over the Bohai Sea and Yellow Sea, while NWP1 and NWP2 denote cruises conducted in 2021 and 2022, respectively, in the Northwest Pacific. *For the NWP2 cruise track, only periods when the instrument was functioning normally are shown.*

525

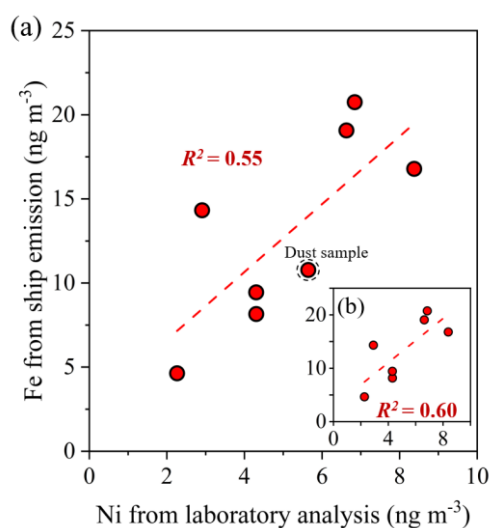
526

527 **Reviewer #2 Minor comment No.3:** In Figure 3c, the use of V for linear regression is
 528 puzzling. Given that dust factor also contributes substantially to V, while Ni is
 529 predominantly explained by ship emissions, it would be more appropriate to use Ni for
 530 the regression analysis. The authors should clarify their choice of V or consider
 531 switching to Ni to better align with the source apportionment results.

532 **Response to Reviewer #2 Minor comment No.3:** We thank the reviewer for this
 533 suggestion. Both V and Ni are widely used tracers of ship emission. In this study, we
 534 prioritized V for the regression analysis because ship-derived Fe shows a stronger
 535 correlation with V (R^2 as 0.68) than with Ni (R^2 as 0.55). Although V was temporarily
 536 influenced by dust contributions during a strong dust event, its overall relationship with
 537 ship-derived Fe remained more robust across the dataset.

538 Following the reviewer's suggestion, we have further added a supplementary figure
539 (Fig. S5) showing the relationship between ship-derived Fe concentrations and Ni. The
540 corresponding discussion has been added in the revised manuscript to further clarify
541 this comparison (Page 11, Lines 288–292):

542 *“We further examined the relationship between ship-derived Fe and another commonly*
543 *used tracer for ship emission, Ni. Excluding the dust-affected sample changed the R^2 of*
544 *their linear regression from 0.55 to 0.60 (Fig. S5), indicating a limited dust influence*
545 *on Ni. Nevertheless, the overall correlation between Ni and ship-derived Fe remained*
546 *weaker than that between V and ship-derived Fe, which supports the higher relative*
547 *contribution of ship emission to V than to Ni in the Northwest Pacific, as revealed by*
548 *modelling (Jiang et al., 2024).”*



549 **Figure S5.** Comparison of ship-derived Fe concentrations resolved by PMF and online Xact 625
550 data versus Ni concentrations measured from offline filter analyses. (a) All samples; (b) excluding
551 the dust-affected sample. The dashed circle indicates the dust sample, and the red dashed lines
552 represent the linear regression lines.
553

554 Reference

555 Jiang, S., Zhang, Y., Yu, G., Han, Z., Zhao, J., Zhang, T., and Zheng, M.: Source-
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557 the East Asian seas, Atmos. Chem. Phys., 24, 8363–8381,
558 <https://doi.org/10.5194/acp-24-8363-2024>, 2024.

559

560

561 **Reviewer #2 Minor comment No.4:** Line 33: "Iron (Fe) in marine aerosols have been
562 extensively studied..." should be "has been" (subject "Iron" is singular).

563 **Response to Reviewer #2 Minor comment No.4:** We have corrected the sentence to
564 "Iron (Fe) in marine aerosols *has* been extensively studied...".

565

566

567 **Reviewer #2 Minor comment No.5:** Line 42: "This made non-dust emissions
568 especially important..."→"This makes..." (present tense is more appropriate for a
569 general statement).

570 **Response to Reviewer #2 Minor comment No.5:** We have revised the sentence by
571 changing "This made" to "This *makes*" to use the present tense.

572

573

574 **Reviewer #2 Minor comment No.6:** Line 75 and Line 526: "Combing" should be
575 "Combining"

576 **Response to Reviewer #2 Minor comment No.6:** We have corrected "Combing" to
577 "*Combining*" at both Line 76 and Line 544 in the revised manuscript.

578

579

580 **Reviewer #2 Minor comment No.7:** Line 164: The phrase "Other than Fe" suggests
581 that Fe is excluded from the analysis, whereas the PMF analysis actually includes Fe.
582 It is recommended to rephrase this to: "In addition to Fe, other elements were primarily
583 used as input variables..." to avoid potential misunderstanding.

584 **Response to Reviewer #2 Minor comment No.7:** Thank you for this helpful
585 suggestion. The sentence has been rephrased to "*In addition to Fe, other elements were*
586 *primarily used as input variables...*".

587

588

589 **Reviewer #2 Minor comment No.8:** Line 179: The term "downwind" appears to be an
590 oversight. To avoid contamination from the vessel's own emissions, the sampler should
591 be located upwind of the stack. Please verify and correct this wording.

592 **Response to Reviewer #2 Minor comment No.8:** Thank you for raising this important
593 point. We would like to clarify that the sampler was indeed located upwind of the stack.
594 However, under rare conditions when the apparent wind blew from the stern toward the
595 bow, the sampler could still become temporarily downwind of the stack. To address
596 this potential issue, we identified and further examined samples collected under such
597 conditions to assess possible contamination by ship exhaust.

598 To improve clarity and avoid misunderstanding, we have revised the text as follows
599 (Page 7, Lines 184-186):

600 “Second, 75 samples were selected only if, during the respective sampling cycle, the
601 inlet was *occasionally located* downwind of the ship’s funnel *due to the apparent wind*
602 *blowing from the stern toward the bow*, and the apparent wind speed *was* maintained at
603 a minimum of 2 m s⁻¹ for at least 5 minutes.”

604
605

606 **Reviewer #2 Minor comment No.9:** Line 344: “northern Yellow Seas” should be
607 “northern Yellow Sea”

608 **Response to Reviewer #2 Minor comment No.9:** “northern Yellow Seas” has been
609 changed to “*northern Yellow Sea*” in the revised manuscript now.

610
611

612 **Reviewer #2 Minor comment No.10:** Line 343: The authors note that "Excluding the
613 EP events" indicates Figure 5 results do not include several extreme pollution episodes.
614 This exclusion should be explicitly stated in the figure caption for clarity. Additionally,
615 while the authors cite prior studies to justify the higher dust contribution at low-latitude
616 sites compared to high-latitude ones, this finding remains somewhat counterintuitive.
617 First, dust transport pathways can vary significantly across different years and events.
618 Second, it is generally accepted that northern regions are more susceptible to dust
619 influences. The authors should provide a more thorough discussion to substantiate the
620 rationality of this observation in their study context.

621 **Response to Reviewer #2 Minor comment No.10:** We apologize for the confusion
622 and would like to clarify that the data shown in Figure 5 already include the EP events.
623 Therefore, the figure caption does not need to be revised. Instead, we have revised the
624 relevant text in the manuscript to clarify this point.

625 In addition, we fully agree with the reviewer that dust transport pathways can vary
626 significantly across different events. We have revised the relevant sentences to clearly
627 indicate that our observations were influenced by specific high-dust events (Page 14,
628 Lines 352–360):

629 “Fig. 5 shows the spatial distribution of Fe concentrations from different sources as
630 determined by PMF analysis. Dust-derived Fe concentrations exhibited pronounced
631 peaks during EP2 and EP3, reaching 283.5 ± 110.2 ng m⁻³ and 801.5 ± 283.6 ng m⁻³,
632 respectively, while remaining much lower during other periods (55.71 ± 55.49 ng m⁻³).
633 Although global model simulations have indicated that the meridional peak of East
634 Asian dust transport to the ocean *occurs* near 40°N (Ito et al., 2019; Zhu et al., 2025),
635 observations in this study showed higher dust-derived Fe *contributions over the lower-*
636 *latitude Yellow Sea (62%) than over the Bohai Sea (34%) (Fig. 5a). This largely*
637 *resulted from two episodic high-dust events (EP2 and EP3), which substantially*
638 *increased the average dust contribution in the Yellow Sea. However, even after*
639 *excluding these two episodes, the average dust contribution in the Yellow Sea remained*

640 *40%, slightly above that of the Bohai Sea, highlighting the relatively greater*
641 *importance of non-dust sources in the Bohai Sea.”*

642

643 References

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652 Pacific: a springtime analysis (2001–2017), *Atmos. Chem. Phys.*, 25, 5175–5197,
653 <https://doi.org/10.5194/acp-25-5175-2025>, 2025.

654

655

656 **Reviewer #2 Minor comment No.11:** Line 607-621: The difference in Fe
657 concentrations between coastal and open-ocean areas is much more pronounced in
658 NWP2 than in NWP1. Could this be related to the difference in aerosol size fractions
659 sampled between the two campaigns? Please discuss this potential factor.

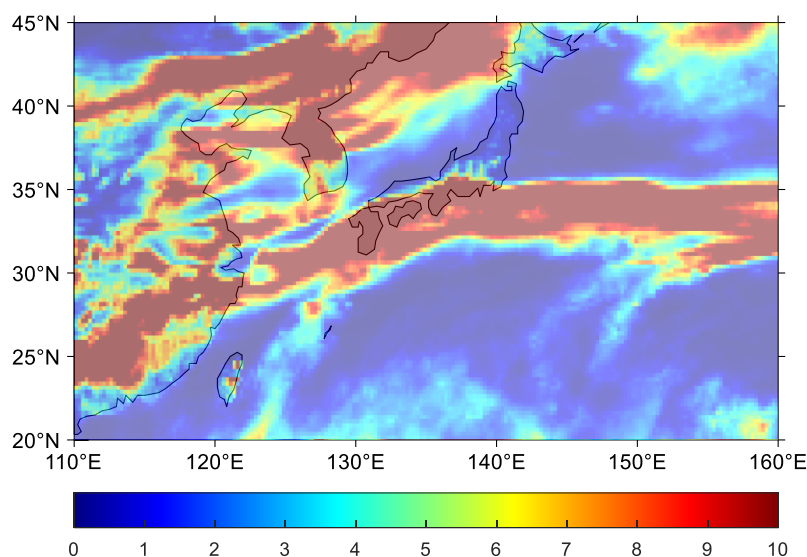
660 **Response to Reviewer #2 Minor comment No.11:** We thank the reviewer for raising
661 this question. Differences in aerosol size fractions may contribute to the observed more
662 pronounced contrast in Fe concentrations between coastal and open-ocean regions.
663 Additionally, large-scale rainfall during NWP2 cruise may also contribute to this
664 observed difference.

665 During the coastal segment of NWP2 cruise, total Fe concentrations in PM₁₀
666 reached a maximum of 883.2 ng m⁻³ (mean: 108.2 ± 248.3 ng m⁻³), exceeding those
667 measured in PM_{2.5} during the coastal segment of NWP1 cruise (maximum: 224.2 ng m⁻³;
668 mean: 76.35 ± 52.40 ng m⁻³). This comparison suggests that sampling larger aerosol
669 size fractions can partly explain the higher Fe concentrations observed in coastal
670 regions during NWP2. However, in the open-ocean regions, an opposite pattern was
671 observed. Iron concentrations in PM₁₀ during NWP2 (23.30 ± 29.42 ng m⁻³) were
672 unexpectedly lower than those measured in PM_{2.5} during NWP1 cruise (47.46 ± 50.57
673 ng m⁻³).

674 Meteorological analysis suggests that the anomalously low Fe level in open-ocean
675 regions during NWP2 was influenced by wet scavenging associated with precipitation.
676 Substantial precipitation occurred over the open Northwest Pacific south of Japan
677 (130°E–144°E) during the NWP2 sampling period (Fig. S11), which would efficiently
678 remove aerosol, resulting in extremely low Fe concentrations in PM₁₀ (7.81 ± 7.62 ng
679 m⁻³) in this region (130°E–144°E), compared with Fe concentrations in PM_{2.5} (54.09 ±
680 26.53 ng m⁻³) over the same region during NWP1 cruise.

681 Overall, these results indicate that while sampling larger aerosol size fractions may

682 have enhanced coastal Fe concentrations during NWP2, the intensified nearshore–
683 offshore gradient was also driven by precipitation-induced scavenging that suppressed
684 Fe concentrations over the open ocean.



685

686 **Figure S11.** Mean precipitation (mm day^{-1}) over the Northwest Pacific from June 20 to 23, 2022.
687 The NWP2 cruise track during this period was located south of the Japan and roughly overlapped
688 with regions of enhanced precipitation.

689

690 The above discussion has been added to the revised manuscript (Page 25, Lines 628–
691 637):

692 “In comparison, during NWP2 cruise (PM_{10} samples), although air mass trajectories
693 were also dominated by marine origins (Fig. S10b), the nearshore-offshore differences
694 in both total Fe and dust-derived Fe were more pronounced than those during NWPI
695 cruise. This pattern was partly driven by enhanced coastal Fe concentrations
696 associated with the larger aerosol size sampled in NWP2. During the YES leg of NWP2,
697 total Fe in PM_{10} reached $108.2 \pm 248.3 \text{ ng m}^{-3}$, exceeding those measured in $PM_{2.5}$
698 during the YES leg of NWPI ($76.35 \pm 52.40 \text{ ng m}^{-3}$). In contrast, over the open ocean,
699 total Fe concentrations during NWP2 ($23.30 \pm 29.42 \text{ ng m}^{-3}$) were lower than those in
700 $PM_{2.5}$ during NWPI ($47.46 \pm 50.57 \text{ ng m}^{-3}$). This anomaly was likely driven by wet
701 scavenging associated with precipitation. Substantial rainfall over the open Northwest
702 Pacific south of Japan (130°E – 144°E) during the NWP2 period (Fig. S11) likely
703 removed atmospheric particles efficiently, resulting in extremely low Fe concentrations
704 ($7.81 \pm 7.62 \text{ ng m}^{-3}$) in this region. Such precipitation-induced wet scavenging over the
705 open ocean further amplified the nearshore–offshore Fe gradient during NWP2.”