Point by Point Response to Review Comments

[Paper #egusphere-2025-4005]

Elevated Anthropogenic Contributions to Trace Elements in Marine Aerosols Compared to Coastal Qingdao in Eastern China

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We sincerely thank the editor and all reviewers for their valuable feedback that we have used to improve the quality of our manuscript. We provide below a point-to-point response to reviewers' comments. The reviewer's comments are in regular **black**; the original (unrevised) text of the manuscript is in ***italicized black***; the response text is in **blue**; and the revisions in the manuscript are in ***red italics***.

Reviewer #5:

General comments:

There are still large uncertainties in source attribution of trace metals in marine aerosols from different anthropogenic sources, despite substantial advancement in the source attribution between natural and anthropogenic sources. This study shows that anthropogenic source is the major contributor to iron (Fe) in $PM_{2.5}$ during summer 2018 based on the measurements from the coastal city of Qingdao (QD) and adjacent offshore regions of Bohai Sea (BS) and Yellow Sea (YS), compared to natural source. This study further suggests that coal combustion contributions to elevated Fe concentrations in the marine aerosols compared to coastal Qingdao. The comprehensive measurements and their application to the PMF model may help us to advance modeling anthropogenic and natural trace metals substantially. However, 8 factors might not directly correspond to the sources of trace metals and their contributions, because the trace metals are minor components of $PM_{2.5}$. Moreover, the attribution of dust to total Fe over BS and YS in spring (16.8% or 25.4%) is significantly smaller than previous studies of source apportionment in the downstream region of East Asian outflow, which might mislead the reader. I have some comments and questions to improve this paper.

Specific comments

Comment 1:

l.24 and l.511: Dust contribution of Fe (16.8% on l.24 and 25.4% on l.511) over the BS and YS in spring is significantly smaller than previous studies in the downstream region of East Asian outflow. Please exclude the sea salt factor to elucidate the dust contribution of Fe. Please also correct the values.

Response:

We thank the reviewer for the helpful comment and fully understand the concern regarding the source attribution results. We have carefully re-examined the PMF results and remain confident in the robustness of our source apportionment.

Unlike many previous studies that only present PMF results, our manuscript provides extensive discussions on the overall consistency and physical plausibility of the $PM_{2.5}$ source apportionment, which strengthens the reliability of the Fe source attribution. One key issue raised by the reviewer is the apparent contribution of the sea salt factor to Fe. We agree that primary sea salt itself should not be a direct source of Fe. However, the purpose of our refined source apportionment is to capture complex atmospheric processes that may influence trace metal distributions. Moreover, according to our subsequent analysis, this is a typical example of the mixture of terrestrial and marine aerosols resolved by PMF, which is exactly one of the reasons we want to highlight this factor.

In fact, mixing processes between mineral dust and sea salt particles during atmospheric transport have been widely reported in previous studies (Andreae et al., 1986; Okada et al., 1990; Wagener et al., 2008; Hsu et al., 2010; Adachi et al., 2020; Knopf et al., 2022; Kwak et al., 2022), including those from our research team (Zhang et al., 2003; Zhang et al., 2006), and such interactions can lead to the association of crustal elements (including Fe) with aged marine particles. Based on the suggestion of Reviewer #6, we have carefully reconsidered the interpretation of this factor and renamed it as an "aged marine aerosol" factor to better reflect the atmospheric processing involved. We have also expanded the discussion and added relevant references (e.g., van Pinxteren et al., 2010; Pey et al., 2013; Liu et al., 2025) to clarify the physical meaning of this factor. Furthermore, in this revised manuscript, we conducted detailed comparisons with previous studies (detailed in response to comment 8).

Importantly, the values of 25.4% in the previous version referred only to the contribution from the pure dust factor (the previously reported 16.8% was an incorrect value and has been corrected). **We agree that reporting this value alone may underestimate the overall influence of dust-related aerosols on Fe over the BS and YS and may therefore mislead readers. When the aged marine aerosol factor, which is closely linked to dust aging and dust–sea-salt mixing, is also considered as part of the dust-related aerosol contribution, the total contribution to Fe over the offshore regions in spring is approximately 60%, which is generally comparable to previous studies conducted in the downstream region of East Asian outflow.**

Therefore, in the revised manuscript, we have retained the original PMF solution but refined the naming and interpretation of several factors, and we have corrected the relevant text in the Abstract (lines 20-40) and Sect. 4.1 (lines 267–275) to clearly distinguish between the pure dust contribution and the dust-related aging process. We believe this revision better reflects the underlying atmospheric processes and avoids misunderstanding of the source apportionment results.

*“**Abstract.** Long-range transport of trace elements (TEs) by aerosols plays a critical role in modulating marine biogeochemistry; however, their source contributions and spatial variability across land-sea gradients remain poorly constrained. Here, we conducted a refined source apportionment of TEs (e.g., Fe, Mn, Cr, V, Ni, Cu, Zn, As, Pb and Cd) in $PM_{2.5}$ collected in the coastal city of Qingdao (eastern China) and adjacent marine regions (the Bohai Sea and Yellow Sea) during spring and summer 2018, to quantitatively resolve terrestrial vs. marine source contributions and identify the key processes controlling their spatial patterns. In spring, all TEs exhibited higher concentrations in Qingdao than in marine atmosphere. In contrast, in summer, Zn, Pb, As, and Cd became more enriched over the marine areas than in Qingdao, with coal combustion accounting for 52.5–78.8% of their concentrations, indicating enhanced anthropogenic impact on the marine atmosphere. For traditional crustal TEs (Fe, Mn and Cr), terrestrial dust dominated in spring Qingdao (e.g., Fe: 81.6%, 2832.0 ng m^{-3}), where the pure dust contributions declined sharply in spring marine areas (Fe: 25.4%, 145.2 ng m^{-3}). However, part of the dust likely underwent aging during transport and was incorporated into the aged marine aerosol factor, which contributed 33.6% of Fe, indicating that dust-related influence remained important offshore and that spring marine aerosols experienced substantial mixing among transported dust, marine processing and anthropogenic emissions. In contrast, coal combustion became the dominant source in summer marine aerosols (Fe: 43.2%, 82.8 ng m^{-3}), exceeding its contribution in Qingdao (Fe: 14.1%, 45.5 ng m^{-3}). Residual oil combustion was identified as the primary source of marine Ni and V (V: 65.7% in spring and 79.8% in summer) and also made substantial contributions to Fe, Mn, and Cr, particularly in summer marine aerosols (e.g., Fe: 26.1%, 50.0 ng m^{-3}). Overall, the refined source apportionment demonstrates that anthropogenic emissions, especially coal combustion and shipping-related residual oil combustion, play a dominant role in shaping the TE composition of marine aerosols over the Bohai and Yellow Seas, while transported dust and its atmospheric aging remain important for crustal elements. These results advance our understanding of land-sea interactions in atmospheric TE cycling and provide new constraints for regional air quality and climate models.”*

Sect. 4.1: *“Factor 5 was characterized by high concentrations of marine source components, including* Na^+, Mg^{2+} *and* Cl^-, *and showed a positive correlation with WS* ($r = 0.35$, $p < 0.01$) *and a strong* *negative correlation with RH ($r = -0.63$, $p < 0.01$) (Sharma et al., 2016; Zhang et al., 2018). These features aligned with the generation of sea salt aerosols production driven by wind-induced disturbance of the ocean surface (Prijith et al., 2014). However, the coexistence of nitrate, sulfate, and harbor-related components (Ca^{2+}, K^{+}) suggested that this factor did not represent fresh sea spray, but rather aged marine aerosol mixed with anthropogenic pollutants during transport (van Pinxteren et al., 2010; Pey et al., 2013; Liu et al., 2025). At the coastal site, its enhanced daytime concentration was consistent with the sea-land breeze patterns (Fig. S6). Time series analysis further showed that Factor 5 had a prevalent influence over the BS and YS (Fig. 4). Notably, in Qingdao during spring, the synchronous peaks of Factor 5 and Factor 3 strongly suggest mixing between marine aerosol and dust during transport."*

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Comment 2:

l.28: This might mislead the reader as if sea salt is the primary sources of trace metals. Please also correct the double sentences on l.431.

Response:

Thanks. Based on the suggestion of Reviewer 6, we have renamed the sea salt factor as an “aged marine aerosol” factor. To avoid the misleading implication, we have revised the text, and the duplicate sentence has been removed. These changes clarify that the factor represents an aged, mixed aerosol type rather than fresh sea salt, while the core message regarding multi-source mixing remains intact.

Lines 29-32: *“However, part of the dust likely underwent aging during transport and was incorporated into the aged marine aerosol factor, which contributed 33.6% of Fe, indicating that dust-related* *influence remained important offshore and that spring marine aerosols experienced substantial mixing among transported dust, marine processing and anthropogenic emissions."*

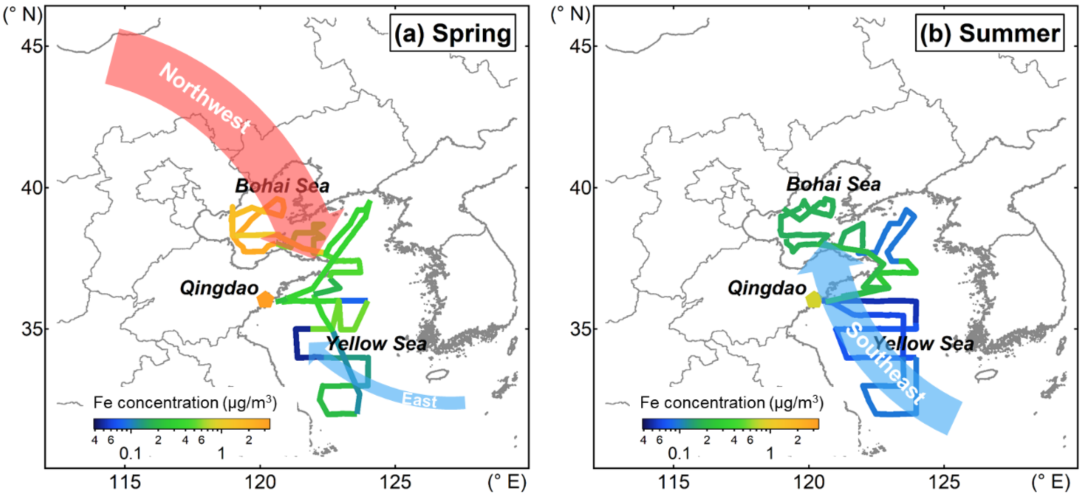
Lines 560-562: *"Springtime marine environments also showed elevated aged marine aerosol contributions to Fe, Mn, Zn, Cd, and Pb (18.5–33.6%), indicating extensive multi-source mixing (dust-marine-anthropogenic) in aerosols. The biogeochemical impact of this complex mixing on the reactivity of elements warrants further investigation."*

<u>Comment 3:</u>

Fig. 1 and Fig. 2: The dominant air mass transport pathways in spring would mislead the reader to understand the contribution of sea salt factor to Fe and Mn. Please reconsider the arrows to explain higher Fe concentrations in spring than summer over the YS.

<u>Response:</u>

We thank the reviewer for this comment. We have revised Fig. 1 to better distinguish the dominant transport pathways. In the spring panel, the northwestern pathway is now highlighted with a thicker and darker arrow to emphasize its primary role in transporting dust and anthropogenic Fe from inland source regions to the BS and YS. The marine pathways are depicted with thinner lines. This modification avoids potential misinterpretation of sea salt contributions.



"Figure 1: Schematic diagram of regional maps with dominant air mass transport pathways (arrows). The solid lines indicate cruise tracks for (a) springtime and (b) summertime cruises over BS and YS during 2018 and the star denotes the location of Qingdao, with the color-filled indicating Fe concentration."

<u>Comment 4:</u>

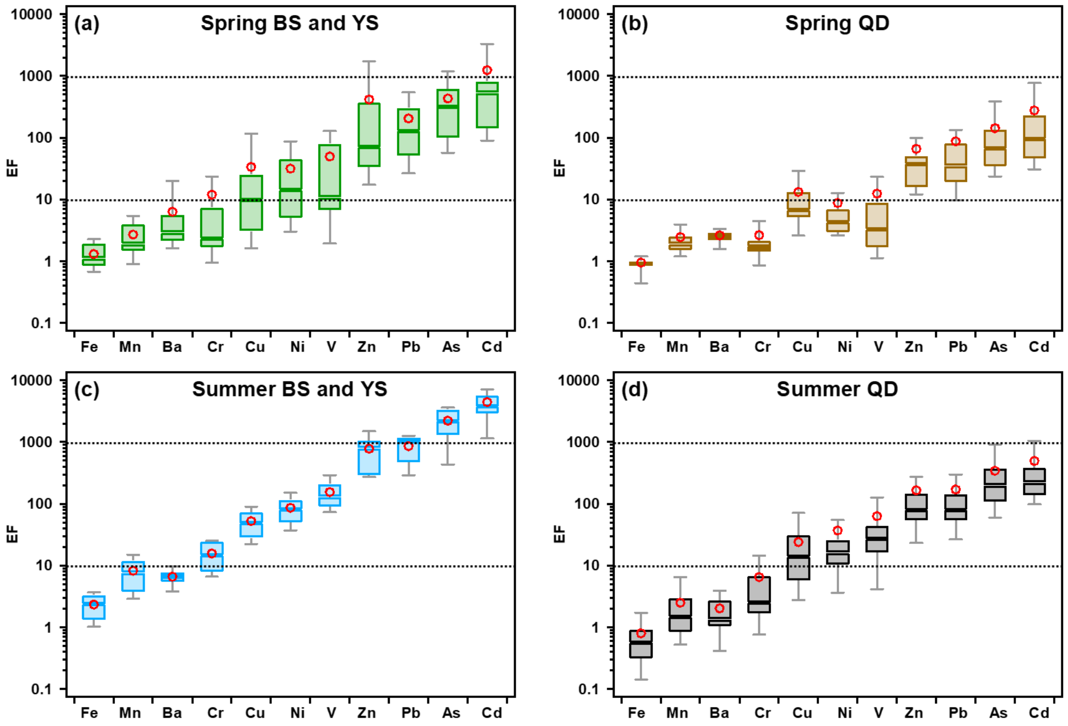
l.148, Fig. 2, Tables S5 and S6, and Fig. S2: Although the Enrichment Factors (EFs) suggest crustal sources of Fe and Mn, their EFs over summer BS and YS are higher than QD, which might support different contributions from different anthropogenic sources. Please show content of metals in $PM_{2.5}$ and discuss the results in the context of the source attribution.

Response:

Thanks for the comment. Following the reviewer's suggestion, we have added two sentences to better relate the EFs with the source apportionment results. First, in Sect. 3 (seasonal variation), we have added text to note that EFs of Fe and Mn over the BS and YS in summer are higher than those observed in Qingdao, and to indicate that this will be further explored in Sect. 4.3.1. Second, in Sect. 4.3.1, we have added a sentence to explicitly link the PMF results with this observation. The revised text now reads:

Sect. 3 (lines 194-197): *"In Qingdao, concentrations of all these elements were notably higher in spring compared to summer (Fig. 2c). Similarly, the EFs of Fe and Mn over the BS and YS in summer were higher than those in Qingdao (Fig. S3), suggesting an enhanced influence of anthropogenic sources in the marine atmosphere during this season. This pattern is further examined in the source apportionment results (Sect. 4.3.1)."*

Sect. 4.3.1 (lines 459-462): *"In summer, coal combustion became the primary contributor to marine Fe and Mn (43.2% and 46.5%, respectively), exceeding Qingdao contributions by 3 times. This finding is consistent with higher EFs of Fe and Mn observed over the BS and YS in summer compared to Qingdao (Fig. S3), further supporting the enhanced influence of anthropogenic sources in the marine atmosphere during this season."*



*“**Figure S2:** Elemental EFs in fine particles over (a) spring BS and YS, (b) spring QD, (c) summer BS and YS, and (d) summer QD. 25th and 75th percentile boxes; 10th and 90th percentile whiskers; the solid line is the median value, and the red circle is the mean value. (It can be seen that the EFs over the BS and YS are generally higher than those in QD.)”*

Comment 5:

l.167: Since you show the median, the high average can be interpreted due to the events. As such, since the outlier informs us the effect of the anthropogenic sources, you should not exclude the event from the analysis. Please show the average values and subsequent PMF analysis including the event and discuss the differences in the results of PMF analysis.

Response:

We sincerely thank the reviewer for this comment and for highlighting the significance of high-concentration events. We fully agree that outliers can provide valuable insights into specific anthropogenic episodes.

Regarding sample SU005, we carefully considered this issue in conjunction with comments from another reviewer during the previous round of revision. Specifically, it was previously noted that this sample exerted a disproportionate influence on the mean statistics given our limited dataset size. Following this input, we conducted rigorous sensitivity tests comparing the PMF results with and without this sample.

Our tests revealed that including such an extreme value compromised the model’s stability, leading to significant discrepancies between the simulated and measured concentrations of trace elements. In small datasets, such distortion can severely bias the source profiles and undermine the overall reliability of the apportionment. Consequently, ensuring the robustness of the PMF solution was our priority; thus, we followed the previous suggestion to exclude this specific sample from both the average calculations and the PMF analysis. In the previous version, we had already evaluated the simulation results of several representative elements. In this revised manuscript, we have explicitly described this quality-control analysis in the main text. lines 159-162:

"Detailed information on the PMF performance can be found in Text S2 and Fig. S2. The comparison between measured and PMF-simulated concentrations for representative elements (Fe, V, and Pb) is shown in Fig. S2, with R^2 *values of 0.97–0.98 and slopes close to 1.0, demonstrating the strong performance and reliability of the PMF model."*

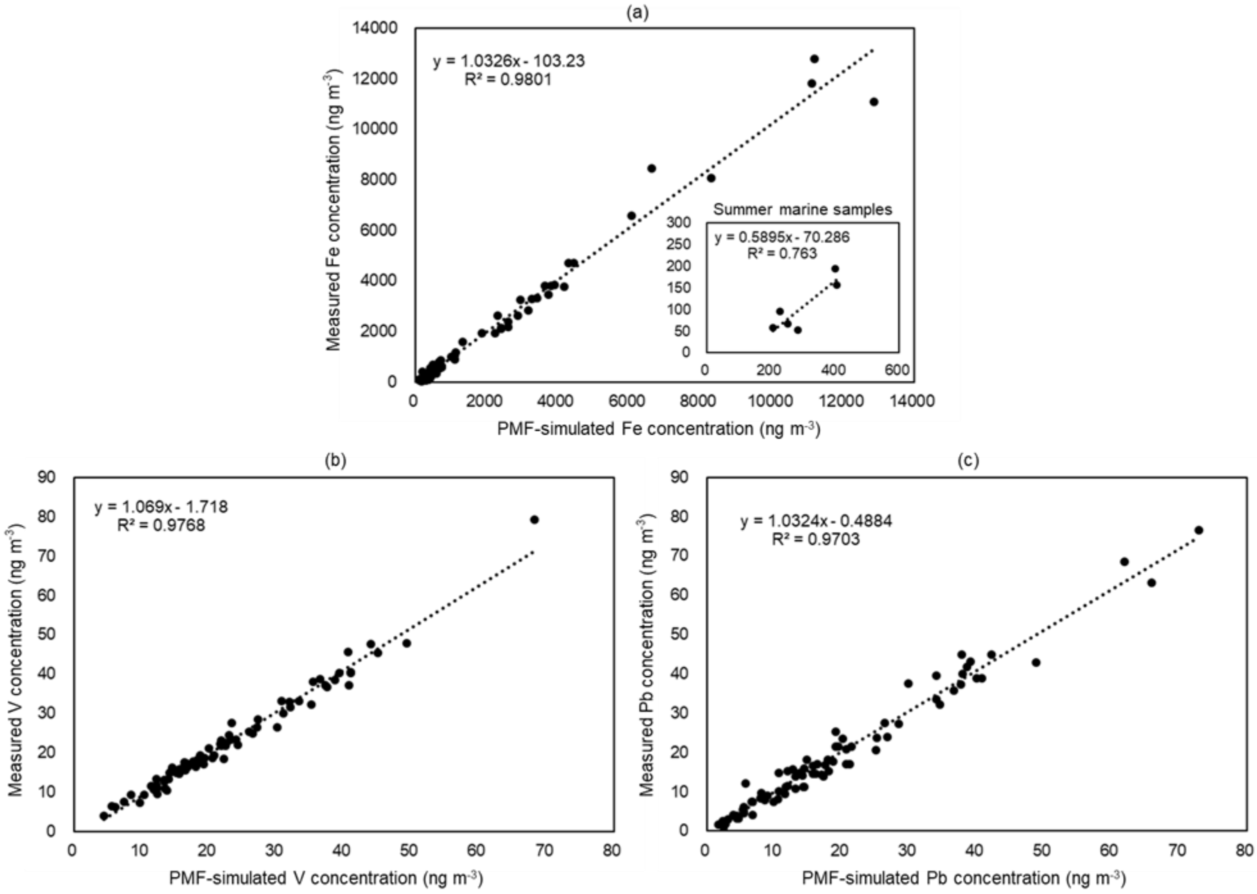


Figure S2. *Comparison of measured and PMF-simulated concentrations (ng* m^{-3}*): (a) Fe, (b) V, and (c) Pb.*

Comment 6

l.188 and Table S5: Please explain the bold and red color in Table S5. How can you explain higher concentrations indicated by bold over ECS than YS?

Response:

We thank the reviewer for the comment. The bold and red font were our markings during revision and will be removed in the final submission as requested by the editorial office. In the current revision, these have been formatted properly. Regarding the higher concentrations of V, Ni, Cu, and Cr over the ECS compared to the YS, this can be explained by regional differences in source emissions and transport processes.

(i) For Cr, its EFs over the YS were mostly in the 1–10 range (Fig. S3), indicating a predominantly crustal source. The lower Cr concentrations over the YS are consistent with the rapid sedimentation of dust particles during long-range transport, as discussed in Sect. 4.2.1. Dust contributed 19.6% (9.7 μg m^{-3}) to $PM_{2.5}$ in Qingdao but only 2.7% (0.5 μg m^{-3}) over the BS and YS, indicating substantial loss during transport. This explains why Cr, being primarily crustal-derived as supported by its EF values, exhibits lower concentrations over the YS compared to the ECS.

(ii) For V and Ni, both primarily from heavy oil combustion, their higher concentrations over the ECS likely reflect stronger emissions from shipping activities in the Yangtze River Delta region, which hosts major ports including Shanghai Port—one of the busiest in the world. In contrast, while Qingdao is also a significant port, the overall shipping density and associated heavy oil combustion emissions in the Yangtze River Delta region are substantially higher, contributing to the observed concentration differences.

(iii) For Cu, its higher concentration over the ECS is consistent with the substantial contribution from vehicle exhaust (20%) reported in Sun et al. (2022), reflecting intensive vehicular emissions in the densely populated Yangtze River Delta.

Reference:

Sun. H., Sun, J., Zhu, C., Yu, L., Lou, Y., Li, R., and Lin, Z.: Chemical characterizations and sources of PM2.5 over the offshore Eastern China sea: Water soluble ions, stable isotopic compositions, and metal elements. Atmos. Pollut. Res., 13, 101410. https://doi.org/10.1016/j.apr.2022.101410, 2022.

Comment 7:

l.285, Fig. 4: Please explain the symbols in the caption, too.

Response:

We thank the reviewer for the comment. We have revised the caption of Fig. 4 accordingly. The updated caption now reads:

"Figure 4: Time series of individual PMF factor concentrations for $PM_{2.5}$ in (a) spring and (b) summer. Bars represent coastal (Qingdao) samples, while open circles with dashed lines denote marine (BS and YS) samples. "SP012" marked in (a) shows the factor concentrations during the sampling period of sample SP012. The detailed procedure for converting the model's factor contribution output to concentrations is provided in Sect. 4.1."

Comment 8:

l.420: Please compare the contribution over YS, ECS, and Northwest Pacific separately. The mixed air masses from the industrial emissions and coal combustion over the YS can introduce highly correlated variables into the PMF, as is described on l.161. Indeed, Fig. S1 show combined vehicular emissions & coal combustion factor when 8-facor is reduced to 7-factor solution. Although multiple factors ranging from 6 to 10 were thoroughly evaluated, how did you consider less factors such as 4 factors in Zhang et al. (2024)?

Why don't you exclude the secondary species for the attribution of trace metals? I also suggest excluding Na^+, Mg^{2+} and Cl^- for the PMF analysis of trace metals because the sea salt is the major component of marine $PM_{2.5}$ but is not the primary sources of Fe and Mn. Please discuss the results whether coal combustion factor can be separated from anthropogenic factors.

Response:

We thank the reviewer for these comments. In the previous version, comparing the average values across the three regions reported by Zhang et al. was indeed not appropriate. Because our dataset does not include samples from the Northwest Pacific, in the revised manuscript we limited the comparison to the coastal regions covered in both studies. Given the differences in the objectives and source-apportionment frameworks of the two studies, we have also provided a more careful and detailed comparison.

(i) As suggested, we now report and compare the source contributions for the YS with other studies.

Lines 470-486: *"Comparisons with previous studies regarding Fe and Mn show both consistencies and discrepancies. Chen et al. (2024) also identified dust as the dominant source of Fe in Qingdao during spring, with a contribution of 88%, which is comparable to our estimate of 81.6%. In contrast, Zhang et al. (2024) reported substantially higher dust contributions over the YS in spring (52–69%) than observed in this study (25.4%) over the BS and YS, together with higher contributions from industry and coal combustion (25.4–33.6% vs. 17.6%), whereas the contribution from ship emissions was broadly comparable (0.9–8.2% vs. 10.2%). Compared with Zhang et al. (2024), our results suggest a more differentiated source structure, particularly over the BS and YS, where continental outflow appears to be shaped by mixing among industrial emissions, coal combustion, vehicular emissions,* *secondary processing, and marine aerosol during transport. Notably, Zhang et al. (2024) did not explicitly resolve the mixing of marine aerosol with transported dust or separate several additional anthropogenic sources. If the aged marine aerosol factor identified here is considered as part of the aged dust-influenced aerosol during transport, the total dust-related contribution would increase to 59.0%, making our results broadly comparable to those of Zhang et al. (2024). Zhang et al. (2024) provided a first-order classification of major source categories, whereas our refined PMF analysis resolves additional source and process complexity and thus offers a more detailed representation of transported aerosols over the marginal seas. The seasonal pattern of Mn source apportionment reported by Yang et al. (2022) for Beijing is also consistent with our results for Qingdao, with dust dominating in spring and industrial emissions becoming more important in summer. Notably, our source apportionment revealed a significant contribution of anthropogenic source to total Fe and Mn in the marine atmosphere, accounting for 41.0% in spring and 90.3% in summer for Fe, and 46.9% and 7.8% for Mn (Fig. 6a, b, and Table S9).”*

(ii) Concerning the factor number selection, we note that source apportionment studies generally follow two complementary frameworks. One is a broad source-classification approach, which aims to distinguish major source categories, such as natural versus anthropogenic sources, or a limited number of dominant source types. This type of framework is commonly adopted in isotope-based studies or PMF analysis using a relatively small set of tracers, and provides an important first-order understanding of aerosol origin (e.g., Mead et al., 2013; Zhang et al., 2024). The other is a more refined source-apportionment approach, which incorporates a broader suite of tracers to better resolve mixed source signatures and atmospheric processing (Wang et al., 2018; Li et al., 2023; Sun et al., 2024). In this framework, the objective is not only to identify broad source categories, but also to reconstruct the complexity of real atmospheric processes in greater detail.

Our study was designed according to the latter framework. Specifically, the PMF analysis was developed based on the **physicochemical interpretability of the bulk $PM_{2.5}$ system**, rather than from the perspective of trace metals alone. For this reason, we intentionally included tracers representing different source types and atmospheric processes. Compared with Zhang et al. (2024), who used 11 trace elements and resolved 4 factors, our study incorporated **23 species**, including trace elements, major water-soluble ions (NO_3^-, NH_4^+, SO_4^{2-}, etc.), and carbonaceous components (OC, EC), and resolved **8 factors**. These two PMF configurations therefore reflect **different apportionment objectives**, rather than simply different choices in factor number. Zhang et al. (2024) provides a useful integrated picture of major source categories, whereas our analysis seeks to move from a broad source classification toward a more detailed and process-oriented interpretation of transported aerosols.

As shown in Fig. S1, when the solution is reduced from 8 factors to 7 factors, **vehicular emissions**

and coal combustion merge into a single combined factor. This result indicates that reducing factor number tends to collapse mechanistically distinct sources into broader mixed categories. Therefore, a 4-factor solution may be appropriate for broad source classification, but is less suitable for the specific objective of this study, namely, to resolve source and process complexity as realistically as possible. In this revision, we add this discussion in Sect. 2.4 in line 139-152:

"The Positive Matrix Factorization (PMF) model is a widely used receptor model to resolve pollution sources and quantify the source contributions to ambient particulate matter concentrations (Paatero and Tapper, 1994). It decomposes the measured data matrix into factor profile, contribution, and residual matrix. In this study, the Environmental Protection Agency (EPA) PMF version 5.0 was utilized for analysis (Norris et al., 2014).

Source apportionment studies generally follow two complementary approaches: a broad classification of major source categories and a more refined apportionment aimed at resolving mixed sources and atmospheric processes in greater detail. Our study follows the latter approach. Therefore, the PMF analysis was constructed based on the physicochemical interpretability of the bulk $PM_{2.5}$ *system, rather than from the perspective of trace metals alone.*

Accordingly, the mass concentrations of 12 elements (V, Cr, Mn, Fe, Ni, Co, Cu, Zn, As, Cd, Ba and Pb), 9 water-soluble ions (Na^+*,* NH_4^+*,* K^+*,* Mg^{2+}*,* Ca^{2+}*,* Cl^-*,* NO_3^-*,* SO_4^{2-} *and* $C_2O_4^{2-}$*), and OC/EC from both Qingdao and cruise campaigns (81 samples) were input to the PMF analysis. Secondary ions and sea-salt related species were retained as process tracers to constrain marine influence, aerosol aging, and internal mixing during transport. Multiple factors ranging from 6 to 10 were thoroughly evaluated to determine the optimal solution. The stability and reliability of the factor solutions were assessed using the displacement (DISP) and bootstrap (BS) uncertainty estimation methods (Norris et al., 2014). "*

(iii) Regarding the reviewer's suggestion to exclude secondary species and sea-salt-related ions (Na^+, Mg^{2+}, and Cl^-) from the PMF analysis of trace metals, we respectfully consider that these species should be retained. Our PMF analysis was not designed to identify only the direct primary sources of Fe and Mn; rather, we first sought to resolve chemically and physically meaningful $PM_{2.5}$ factors, and then quantify the contributions of these factors to trace metals. In marine atmospheres, secondary inorganic species and sea-salt-related ions provide critical constraints on atmospheric processing, marine influence, and internal mixing state. They are particularly important for distinguishing aged marine aerosol, sea-salt-modified particles, and transported continental plumes. Therefore, although

Na^+, Mg^{2+}, and Cl^- are not primary source tracers of Fe or Mn, they are nevertheless important process tracers within the PMF framework. Excluding them would remove valuable information on aerosol aging and mixing, and could artificially force marine or secondary processing signals into dust or anthropogenic factors. We have clarified this rationale in the revised manuscript.

(iv) Finally, we agree that separating coal combustion from other anthropogenic sources is challenging. However, in our interpretation, coal combustion can still be resolved as a distinct combustion-related factor, although some degree of overlap with other anthropogenic influences is expected in a heavily mixed marine atmosphere. Such overlap likely reflects the actual atmospheric mixing state rather than a methodological deficiency. Importantly, retaining this factor remains meaningful because it captures a major component of continental outflow that would otherwise be merged into a broader anthropogenic category in lower-factor solutions.

References:

Li, W., Qi, Y., Qu, W., Qu, W., Shi, J., Zhang, D., Liu, Y., Zhang, Y., Zhang, W., Ren, D., Ma, Y., Wang, X., Yi, L., Sheng, L., and Zhou, Y.: $PM_{2.5}$ source apportionment identified with total and soluble elements in positive matrix factorization, Sci. Total. Environ., 858, 159948, http://dx.doi.org/10.1016/j.scitotenv.2022.159948, 2023.

Mead, C., Herckes, P., Majestic, B. J., and Anbar, A. D.: Source apportionment of aerosol iron in the marine environment using iron isotope analysis, Geophys. Res. Lett., 40, 5722-5727, https://doi.org/10.1002/2013GL057713, 2013.

Sun, M., Qi, Y., Li, W., Zhu, W., Yang, Y., Wu, G., Zhang, Y., Zhao, Y., Shi, J., Sheng, L., Wang, W., Liu, Y., Qu, W., Wang, X., and Zhou, Y. Investigation of a haze-to-dust and dust swing process at a coastal city in northern China part II: A study on the solubility of iron and manganese across aerosol sources and secondary processes. Atmos. Environ., 328, 120532, https://doi.org/10.1016/j.atmosenv.2024.120532, 2024.

Wang, Q., Qiao, L., Zhou, M., Zhu, S., Griffith, S., Li, L., and Yu, J. Z.: Source Apportionment of PM2.5 Using Hourly Measurements of Elemental Tracers and Major Constituents in an Urban Environment: Investigation of Time-Resolution Influence, J. Geophys. Res.-Atmos., 123, 5284-5300, https://doi.org/10.1029/2017JD027877, 2018.

Zhang, T. L., Liu, J. Y., Xiang, Y. X., Liu, X. M., Zhang, J., Zhang, L., Ying, Q., Wang, Y. T., Wang, Y. N., Chen, S. L., Chai, F., and Zheng, M.: Quantifying anthropogenic emission of iron in marine aerosol in the Northwest Pacific with shipborne online measurements. Sci. Total Environ., 912, Article 169158, https://doi.org/10.1016/j.scitotenv.2023.169158, 2024.

Comment 9:

l.424: Despite the significant contribution of anthropogenic combustion to total Fe over the BS and YS, Fig.1 shows larger Fe concentrations over BS than YS. Please discuss previous studies of source apportionment in the downstream region of BS and YS for the implications on the open oceans.

Response:

We thank the reviewer for this comment. Following our revision in response to Comment 8, we have reorganized this part of the discussion. In the revised manuscript, we focus on source comparisons directly relevant to the BS and YS, rather than extending the discussion to downstream open-ocean regions. Regarding the larger Fe concentrations over BS than YS in Fig. 1, we note that the previous version of the manuscript had already included a broader comparison in Sect. 4.2.4, where we discussed source apportionment results across the Western Pacific marginal seas, including the offshore eastern China Sea, Taiwan Island, and the South China Sea, based on studies by Sun et al. (2022) and Yen et al. (2022a, b) (lines 398-427). Therefore, we did not repeat a similar discussion in Sect. 4.3.1.

"4.2.4 Comparison with source apportionment studies in other marginal seas of the Western Pacific

We compared the source apportionment results obtained over the BS and YS in this study with those from studies conducted over the offshore eastern China Sea, around the Taiwan Island, China, and over the South China Sea (SCS) (Sun et al., 2022; Yen et al., 2022a; Yen et al., 2022b). These sea areas all belong to the marginal seas of the Western Pacific, sharing the common influence of the East Asian monsoon and continental outflows. Although direct comparison should be interpreted with caution because of differences in sampling period, receptor location, and PMF inputs among studies, the available results still reveal meaningful regional similarities and contrasts.

Across these marginal seas, $PM_{2.5}$ *sources were generally resolved into secondary inorganic aerosols, dust-related sources (including crustal or fugitive dust as reported in other studies), marine source (including sea salt or oceanic spray), and specific anthropogenic emissions. Among these, secondary aerosols (dominated by nitrate and/or sulfate) and marine-related source were important contributors in most regions. For example, secondary formation (22.9%) and combustion source (30.6%) were reported to make significant contributions over the offshore eastern China Sea in spring (Sun et al., 2022; Fig. S9c), which is qualitatively similar to our findings for spring and summer. Likewise, the notable contribution of marine-related aerosol in this study (22.1%) is comparable to that reported for other sea areas, including 18.6–30.2% around Taiwan Island and 5.6–29.5% over the SCS annually (oceanic spray over the Dongsha Islands; oceanic spray & fugitive dust over the Nansha Islands) (Yen et al., 2022a; Yen et al., 2022b; Fig. S9a, b, and d).*

In contrast, the BS and YS showed a stronger continental influence, as reflected by the clearer resolution of industrial processes and coal combustion related factors. This characteristic was less *pronounced over the more southerly sea areas, suggesting a stronger impact of the industrial structure and coal-dominated consumption of Northern China on the marine regions. Furthermore, vehicular emissions were identified as an important and stable pollution source over the BS and YS (17.6%) in spring and summer, aligning with the substantial contribution (24.5%) reported over the offshore eastern China Sea in spring (Sun et al., 2022). In contrast, studies around the Taiwan Island and over the SCS more commonly resolved a mixed factor combining ship and vehicular emissions, with contributions of 17.4–41.2% annually (Yen et al., 2022a; Yen et al., 2022b). This difference may indicate that traffic and shipping emissions were more readily separated over the BS and YS, although part of the discrepancy may also arise from differences in receptor environments and PMF configurations among studies.*

Overall, the comparison suggests a clear transition in $PM_{2.5}$ *source characteristics from the BS to the YS, with continental anthropogenic influences gradually weakening and marine-related contributions becoming more important southward/offshore, while the adjacent seas of the Western Pacific further reflect increasing impacts from mixed marine and shipping-related emissions.*

Comment 10:

l.431: Why do you include sea salt for the source attribution of Fe and Mn, even though you do not argue that the sea salt is the primary sources of Fe and Mn?

Response:

We thank the reviewer for this comment. This point has been addressed in our previous response. Briefly, sea-salt-related species were retained in the PMF analysis to account for atmospheric mixing and processing, particularly in marine environments. Although sea salt is not considered a primary source of Fe and Mn, its inclusion helps resolve mixed factors and improves the physical interpretability of the results.

Comment 11:

l.468: Please specify secondary aerosol formation of Zn. Why do you include the secondary species for the attribution of trace metals?

Response:

We thank the reviewer for this helpful comment. We have revised the manuscript to clarify both the interpretation of Zn associated with secondary aerosol formation and the rationale for including secondary species in the PMF analysis.

First, regarding Zn, we agree that its presence in a secondary factor should be more clearly explained. In our PMF results, Zn was associated not only with primary anthropogenic sources, but also with a secondary sulfate-rich factor that showed mixing with biomass burning influence. We therefore revised the text to clarify that Zn in this factor does not imply that Zn is produced as a purely secondary species itself; rather, Zn-containing particles emitted from anthropogenic sources can undergo atmospheric aging and partition into secondary aerosol-rich mixtures during transport, especially under sulfate-rich conditions. In addition, biomass burning is also a recognized source of Zn. Accordingly, the revised sentence now reads (lines 506-509):

"Apart from the aged marine aerosol contributions, Zn was associated with multiple anthropogenic sources, including waste incineration, industrial emissions, coal combustion, biomass burning, and secondary sulfate-related aerosol processing. Similar findings have been widely reported in previous PMF studies, where Zn was identified in biomass burning and secondary aerosol factors (Li et al., 2007; Li et al., 2020; Liu et al., 2020; Zhu et al., 2022a; Liu et al., 2025)."

This interpretation is also consistent with previous PMF studies, in which Zn has been reported in biomass burning and secondary aerosol factors (e.g., Li et al., 2007; Li et al., 2020; Liu et al., 2020; Zhu et al., 2022a; Liu et al., 2025).

Second, regarding the inclusion of secondary species in the PMF analysis, our objective was not to identify only the direct primary sources of trace metals, but to resolve chemically meaningful $PM_{2.5}$ source/process factors and then evaluate their contributions to trace metals. Secondary species provide important constraints on atmospheric processing, aging, and mixing state, which are particularly relevant in marine and long-range transport environments. Their inclusion improves the separation of primary emissions from aged or internally mixed aerosol factors, and therefore helps produce source profiles that are more physically and chemically interpretable. For this reason, we retained secondary species in the PMF input.

References:

"Li, R., Wang, Q., He, X., Zhu, S., Zhang, K., Duan, Y., Fu, Q., Qiao, L., Wang, Y., Huang, L., Li, L., and Yu, J. Z.: Source apportionment of PM2.5 in Shanghai based on hourly organic molecular markers and other source tracers, Atmos. Chem. Phys., 20, 12047-12061, https://doi.org/10.5194/acp-20-12047-2020, 2020.

Li, X., Wang, S, Duan, L., Hao, J., Li, C., Chen, Y., and Yang, L.: Particulate and trace gas emissions from open burning of wheat straw and corn stover in China. Environ. Sci. Technol. 41, 6052-6058, https://doi.org/10.1021/es0705137, 2007.

Liu, B., Sun, X., Zhang, J., Bi, X., Li, Y., Li, L., Dong, H., Xiao, Z., Zhang, Y., and Feng, Y.:

Characterization and Spatial Source Apportionments of Ambient PM10 and PM2.5 during the Heating Period in Tian'jin, China, Aerosol Air Qual. Res., 20, 1-13, https://doi.org/10.4209/aaqr.2019.06.0281, 2020.

Liu, X., Zhang, X., Jin, B., Wang, T., Qian, S., Zou, J., Dinh, V. N. T., Jaffrezo, J. -L., Uzu, G., Dominutti, P., Darfeuil, S., Fzvez, O., Conil, S., Marchand, N., Castillo, S., de la Rosa, J., Grange, S., Hueglin, C., Eleftheriadis, K., Diapouli, E., Manousakas, M. -I., Gini, M., Nava, S., Calzolai, G., Alves, C., Monge, M., Reche, C., Harrison, R. M., Hopke, P. K., Alastuey, A., and Querol, X.: Source apportionment of PM10 based on offline chemical speciation data at 24 European sites, npj Clim. Atmos. Sci., 8, 255, https://doi.org/10.1038/s41612-025-01097-7, 2025.

Zhu, W., Qi, Y., Tao, H., Zhang, H., Li, W., Qu, W., Shi, J., Liu, Y., Sheng, L., Wang, W., Wu, G., Zhao, Y., Zhang, Y., Yao, X., Wang, X., Yi, L., Ma, Y., and Zhou, Y.: Investigation of a haze-to-dust and dust swing process at a coastal city in northern China part I: Chemical composition and contributions of anthropogenic and natural sources, Sci. Total. Environ., 851, 158270, https://doi.org/10.1016/j.scitotenv.2022.158270, 2022a."

Reviewer #1:

This manuscript discusses the emission sources and seasonal variability of metal elements in aerosols collected in Qingdao, China, and its adjacent offshore region (the Yellow Sea and the Bohai Sea). The authors have responded well to the numerous comments, and the manuscript has been substantially improved. However, the discussion on Fe dissolution may go beyond what is directly supported by the current dataset, but it is not fundamentally incorrect. I therefore recommend publication in Atmospheric Chemistry and Physics after minor revisions, including a more restrained wording in this section and correction of several minor errors.

Comment 1:

L26: The significant digits for the Fe concentration and solubility are given to four digits (three digits elsewhere).

Response:

We thank the reviewer for pointing out this inconsistency. We have revised the significant digits of the Fe concentration and solubility to align with the format used elsewhere. During this check, we also identified and corrected additional numerical inconsistencies in the Abstract, and polished the language of the Abstract. We have thoroughly reviewed the entire manuscript and unified the number of chemical places throughout.

Lines 27-29: *"For traditional crustal TEs (Fe, Mn and Cr), terrestrial dust dominated in spring Qingdao (e.g., Fe: 81.6%, 2832.0 ng m^{-3}), where the pure dust contributions declined sharply in spring marine areas (Fe: 25.4%, 145.2 ng m^{-3})."*

Lines 32-35: *"In contrast, coal combustion became the dominant source in summer marine aerosols (Fe: 43.2%, 82.8 ng m^{-3}), exceeding its contribution in Qingdao (Fe: 14.1%, 45.5 ng m^{-3}). Residual oil combustion was identified as the primary source of marine Ni and V (V: 65.7% in spring and 79.8% in summer) and also made substantial contributions to Fe, Mn, and Cr, particularly in summer marine aerosols (e.g., Fe: 26.1%, 50.0 ng m^{-3})."*

Comment 2:

L363: There are two periods; please remove one.

Response:

We thank the reviewer for the careful reading. The duplicate period has been removed.

Lines 396-397: *"The lower Ni/V ratio (0.37) in our study suggested residual oil combustion pollutants in the study area may still include higher-sulfur fuel signatures, potentially from regional transport rather than strictly regulated local shipping."*

Comment 3:

L400–402: Please clarify whether the contributions of coal combustion and vehicular emissions to Fe and Mn were higher in marine aerosols than in Qingdao. If so, please also provide the corresponding source contributions to Fe and Mn in Qingdao $PM_{2.5}$ to facilitate comparison.

Response:

We thank the reviewer for the comment. Yes, the relative contributions (percentages) of coal combustion and vehicular emissions to Fe and Mn were higher in marine aerosols than in Qingdao. To facilitate comparison, we have added the corresponding source contributions in Qingdao at lines 454–456. The revised sentences now read:

"In contrast to dust, the relative contributions from vehicular emissions (11.0% and 10.6% for Fe and Mn, respectively) and coal combustion (16.2% and 19.5%), were markedly higher in marine aerosols than in Qingdao (vehicular emissions: 1.8% and 1.8%; coal combustion: 4.9% and 6.2%)."

Comment 4:

L416–419: Is the difference truly statistically significant? The values appear almost identical. This could imply that stationary emissions and residual combustion are influential, or that ship traffic shows little seasonal variability, resulting in a comparable contribution from heavy-fuel-oil combustion throughout the year.

Response:

We thank the reviewer for this comment. To address whether the seasonal differences are statistically significant, we performed a t-test on the absolute concentrations of Fe and Mn from residual oil combustion in Qingdao between spring and summer. The results show that the differences are not statistically significant ($p > 0.05$), consistent with the reviewer's observation. We have revised the text at lines 465–469 accordingly to include this statistical result and its interpretation.

"However, from the perspective of absolute concentration, the contributions of residual oil combustion to Fe and Mn in Qingdao remained lower in summer (77.8 and 2.9 ng m^{-3}, respectively) than in spring (98.7 and 3.7 ng m^{-3}). A t-test on the concentrations showed that these seasonal differences are not statistically significant ($p > 0.05$), suggesting that emissions from residual oil combustion remain relatively stable, likely associated with persistent shipping activities and stationary sources."

Comment 5:

L424-429: This paragraph is appropriate when discussing studies that focus on the Fe solubility in TSP, which includes both coarse and fine aerosol particles, or when comparing Fe solubility between coarse and fine aerosol particles. However, because the present study focuses exclusively on $PM_{2.5}$ (i.e., focus on only fine aerosol particles), it seems inappropriate to apply the same argument directly.

In general, mineral dust is enriched in coarse aerosol particles, whereas combustion-derived anthropogenic Fe is enriched in fine aerosol particles. Therefore, anthropogenic Fe is often considered to have a larger specific surface area than mineral dust and, consequently, to be more readily solubilized through atmospheric chemical processing, as you mentioned in the manuscript. However, the difference is expected to be much smaller than in TSP, because only $PM_{2.5}$ (fine aerosol particles) is analyzed in this study. Indeed, it has been reported that Fe in mineral dust in fine aerosol particles can also be solubilized by acid processing, potentially reaching comparable Fe solubility to those of anthropogenic Fe (Sakata et al., 2023, 2025). As I noted previously, without data on dissolution fractions of Fe and other elements, or dissolved Fe concentrations, emphasizing the importance of anthropogenic Fe for oceanic Fe supply must be regarded as an over-discussion.

Response:

We fully agree with the reviewer's insightful comment and acknowledge that without direct measurements of dissolved Fe or dissolution fractions, speculating on Fe solubility and its drivers would be over-interpreting the data.

Therefore, we have completely removed the section discussing Fe solubility, including the references to physicochemical differences between combustion-derived Fe and mineral dust, as well as the associated mechanisms. The revised paragraph now focuses exclusively on the quantitative results of our source apportionment regarding the contribution of anthropogenic source to total Fe loading in $PM_{2.5}$. The revised paragraph reads (lines 484-486):

"Notably, our source apportionment revealed a significant contribution of anthropogenic source to

total Fe and Mn in the marine atmosphere, accounting for 41.0% in spring and 90.3% in summer for Fe, and 46.9% and 7.8% for Mn (Fig. 6a, b, and Table S9."

Comment 6:

L431–434: The same sentence is repeated twice.

Response:

We thank the reviewer for pointing this out. The duplicate sentence has been removed.

Comment 7:

L431–449: While it is important to discuss potential factors that may affect Fe solubility, the current discussion appears to be overstated. The PMF results only suggest that sea-salt particles and a subset of mineral dust exhibited similar behavior (e.g., transport pathways). However, PMF alone cannot determine whether these components were internally or externally mixed, and it cannot constrain their impacts on Fe solubility in the absence of dissolved metal concentration data. To keep the interpretation appropriately restrained, this part could be made more concise.

Response:

We appreciate the reviewer's critical assessment. We acknowledge that PMF itself is a statistical tool that partitions variance based on temporal covariation, and it cannot directly visually confirm internal mixing, as electron microscopy does. However, we respectfully argue that our PMF analysis, supported by our group's extensive prior work on aerosol mixing states (e.g., Zhang et al., 2003; Zhang et al., 2006), provides robust evidence for the interaction between sea salt and mineral dust. The distinct covariance in the temporal evolution and transport pathways of these factors strongly suggests that the crustal species were not merely co-existing but were incorporated into the sea-salt matrix during transport—a finding consistent with our previous publications on this topic. In response to the comment that the discussion on the impact on Fe solubility was overstated, we have significantly streamlined the text. We have removed the detailed debate regarding pH buffering mechanisms (e.g., TSP vs. submicron acidification) which, while interesting, was too speculative without direct dissolved Fe data. The revised discussion now concisely presents the mixing evidence derived from PMF and provides a more restrained overview of the potential (but complex) implications for bioavailability, as informed by literature. The revised section (lines 441–453) reads:

"Over the marine region, however, the contribution of dust decreased substantially, to 25.4% for Fe and 23.4% for Mn. This significant reduction in the relative contribution of dust over the ocean suggests substantial processing and mixing during transport. Indeed, the PMF model identified "aged marine aerosol" as a distinct source, which contributed 33.6% to Fe and 29.7% to Mn in marine aerosols during spring (Fig. 6a and b). This factor is characterized by strong correlations between sea salt ions and crustal elements (e.g., Fe, Mn), coherent temporal variations, and consistent backward trajectory clusters, indicating substantial mixing of mineral dust with sea spray aerosols (SSAs) during transport (Geng et al., 2014; Hilario et al., 2020). This behavior aligns with previous single-particle observations, showing that mineral dust can be efficiently incorporated into SSA particles during long range transport over the ocean (Andreae et al., 1986; Okada et al., 1990; Zhang et al., 2003; Zhang et al., 2006; Wagener et al., 2008; Hsu et al., 2010a; Adachi et al., 2020; Knopf et al., 2022; Kwak et al., 2022). The implications of such internal mixing for Fe bioavailability remain uncertain. While SSAs could inhibit Fe dissolution via pH buffering (Hsu et al., 2010a), organic ligands in SSAs may also promote the release and stabilization of soluble Fe (Wu et al., 2023). Since dissolved Fe was not measured in this study, we do not attempt to quantify this effect, but note that SSAs mixing is a non-negligible process in the marine Fe budget."

References:

Zhang, D., Iwasaka, Y., Shi, G., Zhang, J., Matsuki, A., and Trochkine, D.: Mixture state and size of Asian dust particles collected at southwestern Japan in spring 2000, J. Geophys. Res.-Atmos., 108(D24), 4760, https://doi.org/10.1029/2003JD003869, 2003.

Zhang, D., Iwasaka, Y., Matsuki, A., Ueno, K., and Matsuzaki, T.: Coarse and accumulation mode particles associated with Asian dust in southwestern Japan, Atmos. Environ., 40, 1205–1215, https://doi.org/10.1016/j.atmosenv.2005.10.037, 2006.

Comment 8:

Figure 4: The bar chart and the line plot appear to correspond to the Qingdao and marine aerosol data, respectively, but this is not explained. Please state this explicitly.

Response:

We thank the reviewer for the comment. We have revised the caption of Fig. 4 accordingly. The updated caption now reads:

"Figure 4: Time series of individual PMF factor concentrations for PM$_{2.5}$ in (a) spring and (b) summer. Bars represent coastal (Qingdao) samples, while open circles with dashed lines denote marine (BS and YS) samples. "SP012" marked in (a) shows the factor concentrations during the sampling period of sample SP012. The detailed procedure for converting the model's factor contribution output to concentrations is provided in Sect. 4.1."

Reviewer #6:

Overview: The revised version of this paper is much improved and more clearly explains the methodology used to complete this study. I do still have concerns that the authors should carefully consider before re-submitting, particularly regarding the interpretation of the PMF factors.

Major Comments:

Comment 1:

1. I still have questions and concerns about the methods that need more information. My first one is, were certified reference materials digested in order to certify which metals you can confidently measure from your digestion method? Also, what type of mass analyzer does the ICP use?

Response:

We thank the reviewer for these important comments.

Certified reference materials were used to evaluate the accuracy of the digestion and analytical procedures. The reference materials were digested and analyzed using the same procedure as the aerosol samples. The recoveries of target elements were within acceptable ranges (typically 98.5–117.5%; e.g., mean recoveries of 98.5% for Fe and 105.6% for Mn), indicating that the digestion method provided reliable measurements for the analyzed metals. This analytical has been extensively applied and validated in our laboratory over nearly two decades (Shi et al., 2012; Shi et al., 2013). We add the references in the revised manuscript line 118.

Elemental concentrations were determined using an inductively coupled plasma-mass spectrometry (ICP-MS, model: iCAP Qc, manufacturer, Thermo Fisher Scientific, Germany), which is equipped with a quadrupole mass analyzer. This information has been clarified in the revised manuscript (lines 115-118).

"The concentrations of elements in the extracts were measured using an inductively coupled plasma-mass spectrometry (ICP-MS, iCAP Qc, Thermo Fisher Scientific, Germany) equipped with a quadrupole mass analyzer. Rh, Sc, and Th were used as internal standards to correct for matrix effects and instrumental drift. A standard solution was analyzed as a quality control sample for every 10 samples (Shi et al., 2012)."

References:

Shi, J.-H, Gao, H.-W., Zhang, J., Tan, S.-C., Ren, J.-L, Liu, C.-G., Liu, Y., and Yao, X.: Examination of causative link between a spring bloom and dry/wet deposition of Asian dust in the Yellow Sea, China, J. Geophys. Res., 117, D17304, https://doi.org/10.1029/2012JD017983, 2012.

Shi, J.-H., Zhang, J., Gao, H.-W., Tan, S.-C., Yao, X.-H., and Ren, J.-L.: Concentration, solubility and deposition flux of atmospheric particulate nutrients over the Yellow Sea, Deep-Sea Res. Pt. II, 97, 43-50, https://doi.org/10.1016/j.dsr2.2013.05.004, 2013.

Comment 2:

2. The PMF time series and discussion should be in the main paper and not in the SI as it is the most important part of the paper.

Response:

We thank the reviewer for this comment. Following the suggestion, we have moved the description of how the PMF factor concentration time series were reconstructed from the Supplementary Information to the main text. This addition has been placed in Sect. 4.1 after the introduction of Fig. 4 (lines 226-230), as follows:

"Eight factors were identified by PMF. The resolved factor profiles and time series are presented in Fig. 3 and Fig. 4 respectively. The time series of individual PMF factor concentrations (e.g., Fig. 4) were derived from the factor contributions output by the PMF model. A factor-specific scaling coefficient was determined by relating the PMF-derived factor contributions to the corresponding absolute concentration. This coefficient was then applied to the full factor contribution time series to reconstruct the absolute concentration time series."

"Figure 4: Time series of individual PMF factor concentrations for $PM_{2.5}$ in (a) spring and (b) summer. Bars represent coastal (Qingdao) samples, while open circles with dashed lines denote marine (BS and YS) samples. "SP012" marked in (a) shows the factor concentrations during the sampling period of sample SP012. The detailed procedure for converting the model's factor contribution output to concentrations is provided in Sect. 4.1."

Comment 3:

3. I have questions about some of the PMF factors. Is Factor 1 ammonium nitrate and ammonium chloride? If so, I would be more explicit here.

Response:

We thank the reviewer for this comment. We have revised the description of Factor 1 accordingly. The updated text now reads (lines 232-234):

"Factor 1 was identified as the secondary nitrate factor, characterized by high loadings of NO_3^-, NH_4^+ *and* Cl^- *(Fig. 3) (Wu et al., 2017; B. Xu et al., 2023). Based on its chemical profile, this factor represents a mixture of ammonium nitrate and ammonium chloride."*

Comment 4:

4. Similar to the point above, Factor 5 is labeled as "sea salt" but this is incorrect. I would reclassify as "aged marine aerosol" since it is clearly not fresh sea spray.

Response:

We thank the reviewer for this valuable suggestion and agree that "aged marine aerosol" is a more appropriate classification than "sea salt" for Factor 5. Accordingly, we have revised the factor label throughout the manuscript, including the main text, figures, and SM, and updated the related descriptions to better reflect its chemical characteristics and atmospheric processing. Due to the extensive nature of the revisions, they are not fully presented here.

Lines 267-275: *"Factor 5 was characterized by high concentrations of marine source components, including* Na^+, Mg^{2+} *and* Cl^-*, and showed a positive correlation with WS* ($r = 0.35$, $p < 0.01$) *and a strong negative correlation with RH* ($r = -0.63$, $p < 0.01$) *(Sharma et al., 2016; Zhang et al., 2018). These features aligned with the generation of sea salt aerosols production driven by wind-induced disturbance of the ocean surface (Prijith et al., 2014). However, the coexistence of nitrate, sulfate, and harbor-related components* (Ca^{2+}, K^+) *suggested that this factor did not represent fresh sea spray, but rather aged marine aerosol mixed with anthropogenic pollutants during transport (van Pinxteren et al., 2010; Pey et al., 2013; Liu et al., 2025). At the coastal site, its enhanced daytime concentration was consistent with the sea-land breeze patterns (Fig. S6). Time series analysis further showed that Factor 5 had a prevalent influence over the BS and YS (Fig. 4). Notably, in Qingdao during spring, the synchronous peaks of Factor 5 and Factor 3 strongly suggest mixing between marine aerosol and dust during transport."*

References:

"van Pinxteren, D., Brüggemann, E., Gnauk, T., Müller, K., Thiel, C., and Herrmann, H.: A GIS based approach to back trajectory analysis for the source apportionment of aerosol constituents and its first application, J. Atmos. Chem., 67, 1-28, https://doi.org/10.1007/s10874-011-9199-9, 2010.

Pey, J., Alastuey, A., and Querol, X.: PM_{10} and $PM_{2.5}$ sources at an insular location in the western Mediterranean by using source apportionment techniques, Sci. Total. Environ., 456-457, 267-277, http://dx.doi.org/10.1016/j.scitotenv.2013.03.084, 2013.

Liu, X., Zhang, X., Jin, B., Wang, T., Qian, S., Zou, J., Dinh, V. N. T., Jaffrezo, J. -L., Uzu, G., Dominutti, P., Darfeuil, S., Fzvez, O., Conil, S., Marchand, N., Castillo, S., de la Rosa, J., Grange, S., Hueglin, C., Eleftheriadis, K., Diapouli, E., Manousakas, M. -I., Gini, M., Nava, S., Calzolai, G., Alves, C., Monge, M., Reche, C., Harrison, R. M., Hopke, P. K., Alastuey, A., and Querol, X.: Source apportionment of PM_{10} based on offline chemical speciation data at 24 European sites, npj Clim. Atmos. Sci., 8, 255, https://doi.org/10.1038/s41612-025-01097-7, 2025."

Lines 330-336: *"Aged marine aerosol, sulfate & BB, coal combustion, vehicular emissions and residual oil combustion showed higher contributions in the marine area, despite lower or comparable absolute concentrations to the coastal site (Fig. 5). Aged marine aerosol contributions were 30.9% in the marine area, versus 13.0% at the coastal site, consistent with the fact of its marine origin. Furthermore, its temporal peaks at Qingdao closely resembled those of the dust factor (Fig. 4a). It is hypothesized that during dust events, aged marine aerosol may mix with the transported dust, making the two factors difficult to distinguish and leading to the higher concentration of aged marine aerosol at Qingdao (6.4 μg m^{-3}) compared to those over the marine area (5.7 μg m^{-3})."*

Lines 365-368: *"Similarly, aged marine aerosol concentrations were higher at the coastal site (0.9 μg m^{-3}) compared to marine areas (0.6 μg m^{-3}), which may be partly attributed to enhanced sea spray generation near the shoreline due to stronger wave breaking (Zhou et al., 2025); summer southeasterly winds may also facilitate marine aerosol transfer inland, thereby influencing the air quality of coastal urban areas (Figs.S4c, d, and S8d)."*

Lines 506-508: *"Apart from the aged marine aerosol contributions, Zn was associated with multiple anthropogenic sources, including waste incineration, industrial emissions, coal combustion, biomass burning, and secondary sulfate-related aerosol processing."*

Lines 535-537: *"A refined PMF analysis identified eight aerosol sources, including secondary nitrate, secondary sulfate & biomass burning, dust, vehicular emissions, aged marine aerosol, residual oil combustion (ship emissions), coal combustion, and waste incineration & industrial pollutants."*

Lines 547-548: *"Marine-related aerosols, particularly aged marine aerosols and ship emissions, dominated marine areas, with residual oil combustion contributing up to 9.1% in spring and 14.7% in summer."*

Supplementary Material (SM): ***"Table S7.*** *Pearson correlations (r) of PMF factor contributions with meteorological parameters (relative humidity (RH) and wind speed (WS)), and gas pollutants concentrations (SO_2, NO_2, O_3 and CO).*

Factor	*RH*	*WS*	SO_2	NO_2	O_3	*CO*
Secondary nitrate	*–0.18*	*–0.17*	*0.30**	*0.45***	*0.26**	*0.39***
Secondary sulfate & biomass burning	*0.36***	*–0.25**	*-0.46***	*-0.52***	*0.39***	*0.54***
Dust	*–0.72***	*0.18*	*0.19*	*0.35***	*–0.13*	*–0.13*
Dust (spring, coastal)	-	*0.43**	-	-	-	-
Vehicular emissions	*0.01*	*–0.36***	*0.11*	*0.10*	*–0.37***	*0.30**
Vehicular emissions (summer)	-	-	-	*0.78***	-	-
Aged marine aerosol	*–0.63***	*0.35***	*0.31**	*0.46***	*–0.05*	*–0.03*
Residual oil combustion	*0.06*	*–0.24**	*0.38***	*0.19*	*–0.06*	*–0.05*
Waste incineration & industrial emissions	*–0.33***	*0.10*	*0.31**	*0.50***	*–0.06*	*0.24*
Coal combustion	*–0.23**	*0.00*	*0.35***	*0.54***	*0.18*	*0.27**

Note. The highest correlation coefficients for each factor are denoted in bold. No gas pollutants data was available for the cruise campaign. The meteorological parameters of the former eight samples in summer Qingdao are missing values.

**. Correlation is significant at the 0.05 level (2-tailed).*

***. Correlation is significant at the 0.01 level (2-tailed)."*

Tables S9 and S10 are not shown here due to their length, but have been revised as requested, along with all related figures.

Comment 5:

5. For Factor 2, why is Ca so high in a biomass burning factor?

Response:

We thank the reviewer for this question. We have examined Factor 2 and offer the following clarifications:

First, the loading of Ca^{2+} in this factor is relatively limited. According to our PMF results, Factor 2 accounts for only 3.9% of the total Ca^{2+}, indicating that Ca^{2+} is not a dominant component of this biomass-burning-related factor.

Second, the presence of minor Ca^{2+} in a biomass burning factor has also been reported in previous studies. For example, Zhang et al. (2024) and Yen et al. (2022) both identified biomass burning factors containing measurable Ca^{2+}, with contributions to Ca^{2+} that were even higher than those found in our study. This indicates that the association of minor Ca^{2+} with biomass burning is not unique to our dataset. A plausible explanation is that the biomass burning emissions may be accompanied by the entrainment of soil or ash-related particles during combustion or by mixing with resuspended crustal materials during transport.

Therefore, a small contribution of Ca^{2+} in this factor is physically reasonable.

References:

Zhang, T. L., Liu, J. Y., Xiang, Y. X., Liu, X. M., Zhang, J., Zhang, L., Ying, Q., Wang, Y. T., Wang, Y. N., Chen, S. L., Chai, F., and Zheng, M.: Quantifying anthropogenic emission of iron in marine aerosol in the Northwest Pacific with shipborne online measurements. Sci. Total Environ., 912, Article 169158. https://doi.org/10.1016/j.scitotenv.2023.169158, 2024.

Yen, P. -H., Yuan, C. -S., Wu, C. -H., Yeh, M. -J., Tseng, Y. -L., Soong, K. -Y.: Transport route-based cluster analysis of chemical fingerprints and source origins of marine fine particles (PM2.5) in South China Sea. Sci. Total Environ., 806, 150591. https://doi.org/10.1016/j.scitotenv.2021.150591, 2022b.

Comment 6:

6. It's not clear to me why some concentrations will be higher over the marine environment than at the coast if the coast has the main sources—especially for lines 279-280. This does not make sense to me.

Response:

We thank the reviewer for this comment. Specifically, the BS and YS are not remote open-ocean environments, but are surrounded by densely populated and highly industrialized coastal regions. For the coal combustion factor, the higher concentrations observed over marine areas can be explained by the spatial distribution of coastal coal-fired power plants and the direction of regional atmospheric transport. A large number of coal-fired power plants are located along the coastline and act as persistent sources of coal-combustion-derived aerosols. Under prevailing meteorological conditions, emissions from these coastal sources can be transported seaward, leading to elevated concentrations over adjacent marine regions. In contrast, these sources are not always located upwind of the Qingdao sampling site, and their influence at the coastal site may therefore be weaker. We have clarified this point in the revised manuscript (lines 300-308):

"The spatial distributions of coal-fired power plants, $PM_{2.5}$ emissions from coal-fired power plants, and atmospheric As concentration in China collectively indicated that the coal combustion factor was mainly associated with transport from coastal source regions (Fig. S8) (Tian et al., 2014; Wang et al., 2016; Zhang et al., 2020). In particular, the dense concentration of coal-fired power plants along the coastline serves as a persistent source of coal-burning aerosols over the sea. Under prevailing winds, emissions from these coastal plants can be transported offshore, resulting in elevated concentrations over marine areas. However, these coal-related sources were not consistently located upwind of the Qingdao sampling site and therefore have minimal influence on concentrations there. Furthermore, although this factor contributed only a limited fraction of $PM_{2.5}$ mass, it made a substantial contribution to heavy metal concentrations, which will be discussed in detail in Sect. 4.3."

Comment 7:

7. It might help to divide the results by season and to have a first section on the prevailing meteorology to help explain the concentrations more clearly.

Response:

We thank the reviewer for this constructive suggestion. We have revised Sect. 3 by adding three subsection titles to better organize the spatiotemporal analysis. These subheadings clarify the structure of our discussion, which already covered seasonal patterns and land-sea comparisons but lacked explicit signposting.

"3.1 Overall concentrations and spatial variations"

"3.2 Seasonal variations"

"3.3 Comparison with other studies"

Regarding the meteorological context, we present the dominant air mass transport pathways during the spring and summer campaigns in Fig. 1. Furthermore, detailed air mass trajectory classifications and their relationships with trace element concentrations are provided in Fig. S5 and Table S8 in the SM. We placed these materials in the SM because they serve as supporting reference rather than core findings, given length constraints and the paper's primary focus on source apportionment.

Additionally, we have discussed the seasonal differences in source contributions between land and sea through the subsections in Sect. 4.2. We believe this balance maintains readability while ensuring the meteorological context informs the interpretation of concentration variations and source apportionment results.

"4.2.1 Enhanced anthropogenic inputs to marine areas driven by spring westerly"

"4.2.2 Dominance of biomass burning and vehicular emissions in summer marine areas"

Comment 8:

8. Section 4.2.3 needs standard deviations and significance tests to more clearly articulate whether changes in chemistry are being observed over time.

Response:

We thank the reviewer for this comment. The discussion in Sect. 4.2.3 is based on published data from Wu et al. (2017), for which only mean values are available; we were unable to obtain the original dataset. This limitation was acknowledged in our previous revision, and the current text frames the discussion around observed trends rather than definitive changes.

Minor Comments:

Comment 9:

1. Line 37, remove "a profusion of" and replace "can engage in intricate physicochemical processes, resulting in the formation of" to "contribute to"

Response:

We thank the reviewer for this suggestion. Following the comment, we have revised line 46 accordingly.

"Anthropogenic activities release pollutants that contribute to atmospheric fine particulate matter ($D_p \leq 2.5$ μm, $PM_{2.5}$)."

Comment 10:

2. Line 45, please also cite (Mackey et al., 2012)

Response:

We thank the reviewer for this suggestion. Following the comment, we have added the reference Mackey et al. (2012) at lines 50-51:

"Some trace elements deposited in the open oceans, such as Mn, Fe, Ni, Cu, Zn, and Cd, have biological roles, generally as cofactors or part of cofactors of enzymes or as structural elements in proteins (Morel and Price, 2003; Mackey et al., 2012)."

Reference:

"Mackey, K. R. M., Buck, K. N., Casey, J. R., Cid, A., Lomas, M. W., Sohrin, Y., and Paytan, A.: Phytoplankton responses to atmospheric metal deposition in the coastal and open-ocean Sargasso Sea. Front. Microbiol., 3, https://doi.org/10.3389/fmicb.2012.00359, 2012."

Comment 11:

3. Line 50, in addition to sources, atmospheric processes also impact solubility.

Response:

We thank the reviewer for this comment. Following the suggestion, we have revised lines 55-57 to explicitly state that atmospheric processes also impact solubility and have added the corresponding references:

"In most cases, only the soluble fractions are more likely to be bioavailable (Shi et al., 2012), and the solubility is closely associated with their sources and atmospheric physicochemical processes (Baker et al., 2006; Sholkovitz et al., 2012; Sun et al., 2024; Zhu et al., 2022b)."

Comment 12:

4. Line 112: why were samples extracted at such a cold temperature (0 deg C)?

Response:

We thank the reviewer for this question. The extraction was carried out at approximately 0 °C in order to minimize chemical alteration, volatilization losses, and other unwanted reactions during the leaching process, thereby ensuring a more reliable determination of water-soluble ion concentrations in atmospheric particles. In practice, this temperature was maintained using an ice-water bath, which is a commonly adopted laboratory procedure, although such operational details are not always explicitly described in published methods. This approach is consistent with methods adopted in previous studies (Shi et al., 2013; Meng et al., 2017). For example, Shi et al. (2013) indicated that "A quarter of the filters was extracted by purified water (> 18.2 MΩ cm) for 40 min of ultrasonic agitation at 0 °C", and Li et al. (2024) noted that "water-soluble matters were extracted by ultrasonic vibration at approximately 0 °C for 40 min".

References:

Li, W., Qi, Y., Liu, Y., Wu, G., Zhang, Y., Shi, J., Qu, W., Sheng, L., Wang, W., Zhang, D., and Zhou, Y.: Daytime and nighttime aerosol soluble iron formation in clean and slightly polluted moist air in a coastal city in eastern China. Atmos. Chem. Phys., 24, 6495-6508, https://doi.org/10.5194/acp-24-6495-2024, 2024.

Meng, Y., Li, P., Cao, W., Shi, J., Gao, H., and Yao, X.: Size distribution of particulate trace elements in mass concentration and their size-dependent solubility in the atmosphere in Qingdao, China (in Chinese), China Environ. Sci., 37, 851-858, 2017.

Shi, J.-H., Zhang, J., Gao, H.-W., Tan, S.-C., Yao, X.-H., and Ren, J.-L.: Concentration, solubility and deposition flux of atmospheric particulate nutrients over the Yellow Sea, Deep-Sea Res. Pt. II, 97, 43-50, https://doi.org/10.1016/j.dsr2.2013.05.004, 2013.

Comment 13:

5. Lines 190-192: not following this sentence. Please rephrase.

Response:

We thank the reviewer for the comment. We have rephrased the sentence at lines 211–212 for clarity. The revised sentence now reads:

"Compared with summer island measurements reported by Yuan et al. (2023), the concentrations of most elements in this study were within the same order of magnitude."

Comment 14:

6. Line 299, there are plenty of acronyms in this paper. No need to add another. Please spell out what WI &IE mean. Same on line 373 with OECS.

Response:

We thank the reviewer for the comment. Following the suggestion, we have spelled out "WI&IE" as "waste incineration & industrial emissions" and "OECS" as "offshore East China Sea" throughout the main text. In figures and tables, the abbreviations are retained due to space limitations. Their full meanings have been explicitly provided in the corresponding captions and notes to ensure clarity. Due to the extensive nature of the revisions, they are not fully presented here.

Line 289: *"Factor 7 was defined as the waste incineration & industrial emissions factor."*

Lines 317-318: *"Secondary nitrate, dust, and waste incineration & industrial emissions exhibited significantly lower concentrations and relative contributions in marine environments compared to the coastal site."*

Lines 399-401: *"We compared the source apportionment results obtained over the BS and YS in this study with those from studies conducted over the offshore eastern China Sea, around the Taiwan Island, China, and over the South China Sea (SCS) (Sun et al., 2022; Yen et al., 2022a; Yen et al., 2022b)."*

Comment 15:

7. Lines 213-214, ships can also burn marine diesel fuel that would have the same signature. This should be pointed out.

Response:

We thank the reviewer for this comment. We note that the lines indicated (213-214) discuss Factor 1, which is unrelated to ship emissions. After careful review, we suspect that the reviewer intended to refer to lines 313-314. Since the suggestion regarding ship emissions is more directly relevant to the residual oil combustion factor, which includes ship emissions, we have revised the discussion on residual oil combustion accordingly as follows:

Lines 343-350: *"As shown in Figs.4a, S8a and c, time series peaks of vehicular emissions factor over the ocean coincided with the samples collected from air masses originating in northern and central China, i.e., samples SP010, SP014, and SP017.*

Residual oil combustion exhibited a lower average concentration in marine areas (1.7 µg m^{-3}*) than in Qingdao (2.9 µg m*$^{-3}$*). This is consistent with the influence of ship and port emissions on coastal urban areas. The time series peaks of residual combustion factor over the ocean were often associated with air masses transported from sea to land (e.g., samples SP006 and SP008). Unlike the waste incineration & industrial emissions factor, these four anthropogenic factors demonstrated that varying transport paths, particle size, and source proximity could affect land-sea distribution patterns of their contributions."*

Comment 16:

8. Line 328, and higher RH would also enhance sulfate production.

Response:

We thank the reviewer for this suggestion. We have revised line 328 to include high relative humidity as an additional factor enhancing sulfate production through aqueous-phase oxidation.

Lines 358-361: *"This reduction was attributed to the thermal decomposition of NH_4NO_3 under high summer temperatures and enhanced secondary sulfate facilitated by photochemical reactions with the high temperature and sufficient light, as well as aqueous-phase oxidation promoted by high RH (Wu et al., 2017; Kim et al., 2022)."*

Comment 17:

9. Could the high sea spray over the coast vs the marine environment be due to enhanced wave breaking at the shore?

Response:

We thank the reviewer for this helpful suggestion. We agree that enhanced wave breaking near the shoreline could partly explain the higher marine aerosol contribution observed at the coastal site relative to the offshore environment. In the revised manuscript, we have added this point to the discussion and now clarify that the elevated aged marine aerosol concentration at the coastal site may result from both enhanced sea spray production in the nearshore zone and the inland transport of marine aerosol by southeasterly winds (Zhou et al., 2025). The revised sentence is as follows (lines 365-368): *"Similarly, aged marine aerosol concentrations were higher at the coastal site (0.9 μg m^{-3}) compared to marine areas (0.6 μg m^{-3}), which may be partly attributed to enhanced sea spray generation near the shoreline due to stronger wave breaking (Zhou et al., 2025); summer southeasterly winds may also facilitate marine aerosol transfer inland, thereby influencing the air quality of coastal urban areas (Figs.S4c, d, and S8d)."*

Reference:

"Zhou, S., Salter, M., Bertram, T., Azevedo, E. B., Reis, F., and Wang J.: Shoreline wave breaking strongly enhances the coastal sea spray aerosol population: Climate and air quality implications, Sci. Adv. 11, eadw0343, https://doi.org/10.1126/sciadv.adw0343, 2025."

Comment 18:

10. Lines 434-440, the authors need to remove these sentences as internal mixing cannot be assumed without single particle analysis.

Response:

We thank the reviewer for this comment. To address the concern, we have adjusted the wording of the relevant sentences to more accurately reflect that the discussion of internal mixing is based on previous studies rather than our own single-particle analysis. The revised text now reads:

Lines 441-450: *"Over the marine region, however, the contribution of dust decreased substantially, to 25.4% for Fe and 23.4% for Mn. This significant reduction in the relative contribution of dust over the ocean suggests substantial processing and mixing during transport. Indeed, the PMF model identified "aged marine aerosol" as a distinct source, which contributed 33.6% to Fe and 29.7% to Mn in marine aerosols during spring (Fig. 6a and b). This factor is characterized by strong correlations between sea salt ions and crustal elements (e.g., Fe, Mn), coherent temporal variations, and consistent backward trajectory clusters, indicating substantial mixing of mineral dust with sea spray aerosols (SSAs) during transport (Geng et al., 2014; Hilario et al., 2020). This behavior aligns with previous single-particle observations, showing that mineral dust can be efficiently incorporated into SSA particles during long range transport over the ocean (Andreae et al., 1986; Okada et al., 1990; Zhang et al., 2003; Zhang et al., 2006; Wagener et al., 2008; Hsu et al., 2010a; Adachi et al., 2020; Knopf et al., 2022; Kwak et al., 2022)."*

Comment 19:

11. Lines 431-449 should be removed as solubility was not measured.

Response:

We thank the reviewer for this comment. We agree that the discussion on Fe and Mn solubility should be more concise, as solubility was not directly measured in this study. Following the suggestion, we have condensed this section to focus on citing previous studies that have explored these mechanisms, while avoiding overinterpretation of our PMF results. The revised text now reads as follows.

Lines 441-453: *"Over the marine region, however, the contribution of dust decreased substantially, to 25.4% for Fe and 23.4% for Mn. This significant reduction in the relative contribution of dust over the ocean suggests substantial processing and mixing during transport. Indeed, the PMF model identified* *"aged marine aerosol" as a distinct source, which contributed 33.6% to Fe and 29.7% to Mn in marine aerosols during spring (Fig. 6a and b). This factor is characterized by strong correlations between sea salt ions and crustal elements (e.g., Fe, Mn), coherent temporal variations, and consistent backward trajectory clusters, indicating substantial mixing of mineral dust with sea spray aerosols (SSAs) during transport (Geng et al., 2014; Hilario et al., 2020). This behavior aligns with previous single-particle observations, showing that mineral dust can be efficiently incorporated into SSA particles during long range transport over the ocean (Andreae et al., 1986; Okada et al., 1990; Zhang et al., 2003; Zhang et al., 2006; Wagener et al., 2008; Hsu et al., 2010a; Adachi et al., 2020; Knopf et al., 2022; Kwak et al., 2022). The implications of such internal mixing for Fe bioavailability remain uncertain. While SSAs could inhibit Fe dissolution via pH buffering (Hsu et al., 2010a), organic ligands in SSAs may also promote the release and stabilization of soluble Fe (Wu et al., 2023). Since dissolved Fe was not measured in this study, we do not attempt to quantify this effect, but note that SSAs mixing is a non-negligible process in the marine Fe budget."*

REFERENCES CITED

Mackey, K. R. M., Buck, K. N., Casey, J. R., Cid, A., Lomas, M. W., Sohrin, Y., & Paytan, A. (2012). Phytoplankton responses to atmospheric metal deposition in the coastal and open-ocean Sargasso Sea. Frontiers in Microbiology, 3, doi.org/10.3389/fmicb.2012.00359.