Reply to Anonymous Referee #2

We thank Anonymous Referee #2 for the valuable comments on our manuscript. Here we provide point-to-point responses to the Referees' comments. For clarity, the Referees' comments are marked in black, authors' responses are marked in **blue**, and changes in the manuscript are marked in **red**.

This study by Hu et al. presents a comprehensive study on the chemical composition, optical properties, and sources of aerosols over the Bohai Sea and Yellow Sea during summer 2023, based on shipboard observations of TSP and PM2.5. The authors combine detailed chemical analyses, optical measurements, stable isotopes, PMF source apportionment, and air mass trajectory analysis to demonstrate that, despite a higher proportion of marine air masses in summer, terrestrial (particularly coastal) emissions dominate the aerosol characteristics in these marginal seas. The topic is important for understanding land–sea interactions in heavily anthropogenically influenced marginal seas. The dataset is valuable and the conclusions are well-supported by the integrated analyses. With some corrections to technical errors and clarifications, this manuscript is suitable for publication in Atmospheric Chemistry and Physics. I recommend minor revision.

Comments:

1. Page 3: Change "from 15 to July 23, 2023," to "from July 15 to 23, 2023". Similarly, change "from 11 to August 13, 2023" to "from August 11 to 13, 2023".

Author reply:

This has been corrected. Page 3, Line 75–76:

The cruise campaign was conducted aboard the R/V Lanhai 101, with samples taken in the BS and northern YS from July 15 to 23, 2023, and in the southern YS from August 11 to 13, 2023.

2. Page 3: The flow rate should be 1.05 m^3 min^{-1}.

Author reply:

This has been corrected in the revised manuscript, Page 3, Line 76–78:

Two high–volume air samplers (Laoying 2031) were located on the first deck of the ship to collect TSP and $PM_{2.5}$ samples at a flow rate of 1.05 m^3 min^{-1} on quartz filters (preheated at 500°C for 4 hours).

3. Page 3: "a standard solution of 5mg L^{-1} was used…". Specify what ions are in this standard.

Author reply:

The standard solution of 5 mg L^{-1} in the manuscript is a mixed standard solution of eight ions including Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, and NO_3^-, with the concentration of each ion being 5 mg L^{-1}. This has been updated in the revised manuscript.

Page 3, Line 87–91:

Water–soluble ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, and NO_3^-) were extracted from filters and analyzed by ion chromatography. In order to verify the stability of the instrument, a standard solution of 5 mg L^{-1} of the above-mentioned ions was used to calibrate the instrument after every 5 samples to ensure that the relative standard deviation of repeated measurements of the same sample is less than 6%.

4. Page 7: The choice of 1000 m as the boundary layer height is reasonable, but it can vary due local meteorology. Consider adding sensitivity analysis (e.g., testing 500 and 1500 m) to show if Rtbl/Rmbl values are robust.

Author reply:

Based on the Referee's comment, we calculated the values of Rtbl and Rmbl at boundary layer heights of 500 and 1500 m, respectively. The results are shown in following Table R1.

The retention ratio of terrestrial air masses (Rtam) and marine air masses (Rmam) are calculated based on the landing point position during the air mass transport process, and are independent of the boundary layer height. We found that changes in boundary layer height mainly have impacts on the retention ratio of terrestrial boundary layer air mass (Rtbl) in the northern sea region. When the height of the boundary layer decreases from 1500 m to 500 m, the Rtbl value decreases from 0.63 ± 0.24 to 0.34 ± 0.27, while the retention ratio of marine boundary layer air mass (Rmbl) remains unchanged. This is mainly attributed to the significant height variation of air masses from northeast China when transported over land, while the variation over the ocean is relatively small. However, no matter how the height of the boundary layer changes, what remains unchanged is that the value of Rmbl is always higher than that of Rtbl. From Figure R1, the height of the boundary layer in the sampling region is mainly below 1000 m or 500 m. Moreover, at boundary layer heights of 1000 m and 500 m, Rtbl changed by about 0.1, without altering its lower characteristics in the northern sea region. Therefore, a boundary layer height of 1000 m can be used as a reference upper limit, and the results are reasonable and robust.

Table R1. Changes in the proportion of four types of air masses in the northern and southern sea regions at different boundary layer heights.

Boundary layer height (m)	Sea region	Rtam	Rmam	Rtbl	Rmbl
1500	Northern	0.55 ± 0.28	0.45 ± 0.28	0.63 ± 0.24	0.91 ± 0.17
	Southern	0.31 ± 0.27	0.69 ± 0.27	0.60 ± 0.32	0.99 ± 0.02
1000	Northern	0.55 ± 0.28	0.45 ± 0.28	0.44 ± 0.26	0.91 ± 0.17
	Southern	0.31 ± 0.27	0.69 ± 0.27	0.60 ± 0.32	0.99 ± 0.02
500	Northern	0.55 ± 0.28	0.45 ± 0.28	0.34 ± 0.27	0.91 ± 0.17
	Southern	0.31 ± 0.27	0.69 ± 0.27	0.57 ± 0.31	0.99 ± 0.02

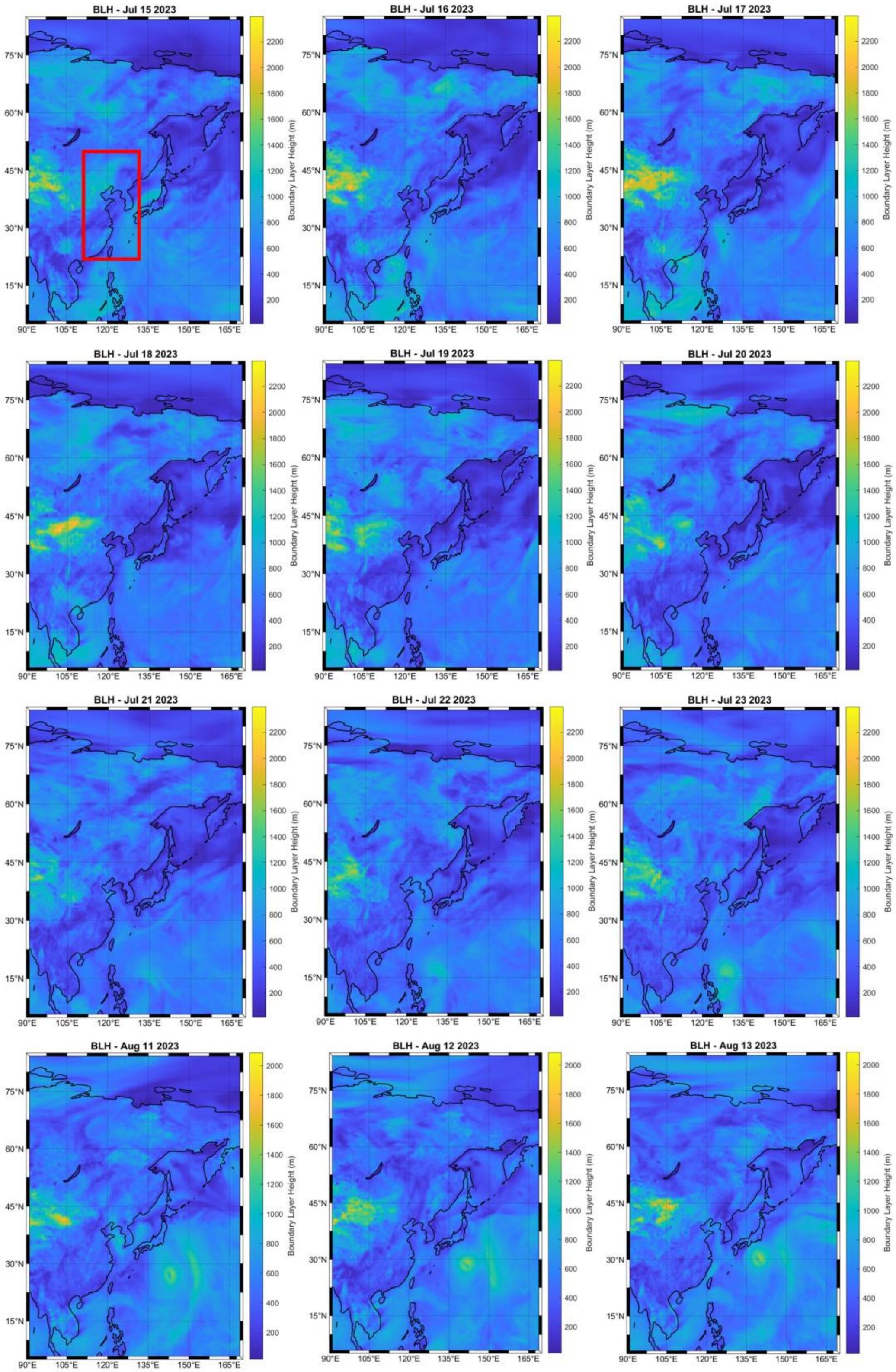

Figure R1. Boundary layer height variation of Bohai Sea, Yellow Sea and its surrounding regions (red box) during the sampling period. The boundary layer height data was downloaded from the ERA5 dataset of the European Centre for Medium Range Weather Forecasts (ECMWF): https://cds.climate.copernicus.eu/datasets.

We have added the above Table R1 and Figure R1 to the Supplement, and the following text of Section 3.1 in the revised manuscript has been updated (Page 7, Line 183–186): The satellite observation results also showed that the boundary layer height of the study region during the sampling period was mainly below 1000 m or 500 m, and the difference in retention ratio of air masses calculated at these boundary layer heights was not significant (Table S1 and Fig. S2). Therefore, we chose 1000 m (equivalent to ~900 hPa) as the upper limit of the boundary layer (Deng et al., 2022).

5. Page 8: "…the concentration of particulate matter is significantly correlated with the calculated Rtbl", provide the correlation coefficient here.

Author reply:

We have added correlation coefficients and significance levels in the revised manuscript, Page 8, Line 203–205:

Firstly, the concentration of particulate matter is significantly correlated with the calculated Rtbl ($R = 0.829$ and $P < 0.01$ for TSP, and $R = 0.811$ and $P < 0.05$ for $PM_{2.5}$), indicating that aerosols in marginal seas are greatly influenced by terrestrial sources (Fig. S3A and B).

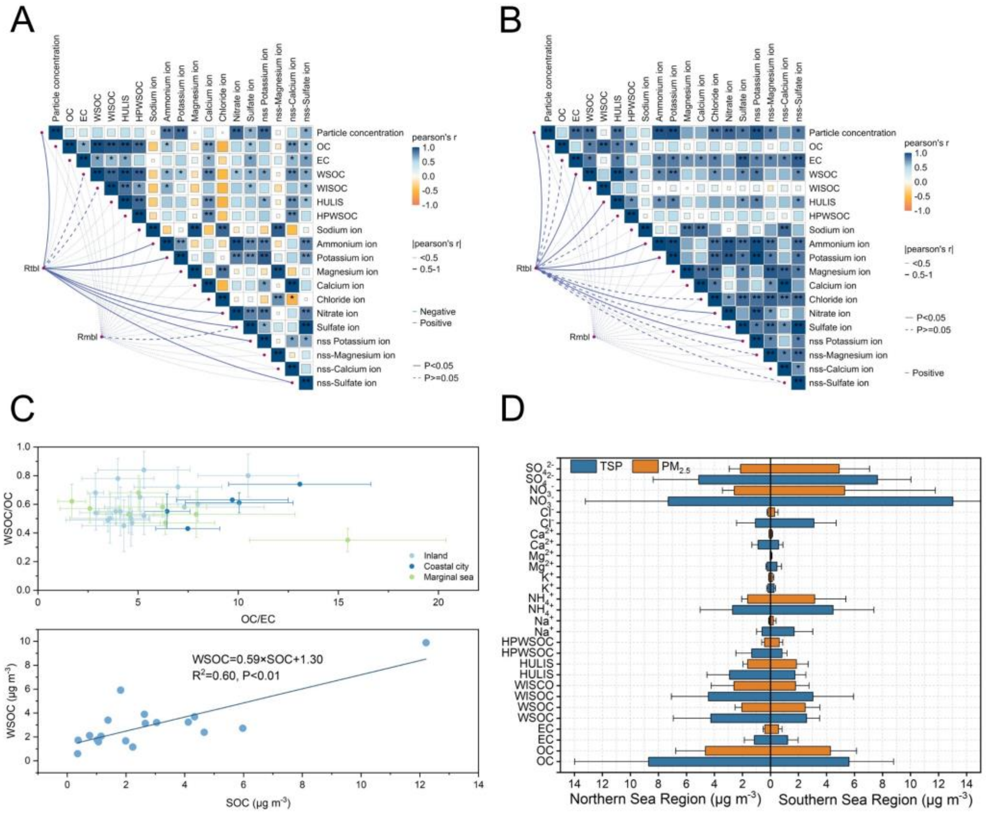

Figure S3. Correlation map of major components in (A) TSP and (B) $PM_{2.5}$. (C) OC/EC and WSOC/OC ratios reported in field studies and the correlation between WSOC and SOC. (D) Concentration comparison of the chemical composition in different sea regions.

References

Deng, J., Ma, H., Wang, X., Zhong, S., Zhang, Z., Zhu, J., Fan, Y., Hu, W., Wu, L., Li, X., Ren, L., Pavuluri, C. M., Pan, X., Sun, Y., Wang, Z., Kawamura, K., and Fu, P.: Measurement report: Optical properties and sources of water-soluble brown carbon in Tianjin, North China – insights from organic molecular compositions, Atmos. Chem. Phys., 22, 6449–6470, https://doi.org/10.5194/acp-22-6449-2022, 2022.