Responses to comments from the reviewer 1

Revised Review Comments:

The paper by Chen et al. provides a clear analysis of the molecular characterization of organic aerosols in urban and seaside environments within a coastal city in southeast China. The dataset is credible and thoroughly analyzed, enhancing our understanding of molecular-level organic aerosol characteristics in coastal areas. The manuscript is generally well written, though it contains several English errors that require attention. I recommend acceptance after minor revisions, as detailed below:

Response: We appreciate your overall positive assessment of the manuscript. We have thought about the issues you raised and carefully revised the entire manuscript accordingly. Detailed responses are listed below, and the main correlations are marked in blue in the "Revised manuscript with changes marked" file.

Lines 116-117: Rewrite the sentence to:

"Potential influence from shipping activities, due to the proximity of Xiamen Port, may impact the seaside site."

Response: We rewrote the site introduction, and this sentence has been modified. (in lines 117–122)

Line 149: Clarify what the correlation coefficients represent.

Response: We rewrote the sentence in lines 152–156. "Meanwhile, parallel experiments were conducted to evaluate the reproducibility of sampling and analytical procedures. These tests showed excellent agreement between the OA signal intensities of duplicate samples, with linear regression slopes of 0.84–1.13 and correlation coefficients (r^2) upwards to 0.997 (Fig. S2)."

Lines 157-158: The BC-related data is not used in the article and could be deleted.

Response: We have corrected it.

Lines 190-191: Rewrite the sentence as:

"The sum of the four compound classes was denoted as CHOX, where X indicates the potential presence of N, S, or both."

Response: We rewrote the sentence in lines 195–196.

Line 228: Ensure consistent terminology for "O_3 concentrations and UVB intensity."

Response: We have corrected it. And we ensured the consistency of terminology throughout the full text.

Lines 343-345: "The signal proportion of compounds with DBE/C > 0.7 was comparable between the two sites (CHO: ~25%; CHON: ~16%)…"

Question: Were low-carbon-number compounds (e.g., $C_2H_2O_4$) excluded from this analysis?

Response: The low-carbon-number compounds were not excluded from the dataset. We calculated the signal proportion of compounds with DBE/C > 0.7 after excluding $C_2H_2O_4$, which was 10% for the urban site and 8% for the seaside site. This indicates that regardless of whether small-molecule compounds are excluded, the proportion of compounds with DBE/C > 0.7 remains higher at the urban site than at the seaside site. The results demonstrate that urban OA compounds systematically exhibit higher DBE/C ratios compared to those at the seaside site.

Lines 417-420: "…urban OA is more influenced by anthropogenic emissions, dominated by aromatic species, whereas marine-influenced OA exhibits higher proportions…"

Note: Verbs should be in singular form for consistency.

Response: We have corrected it. We rewrote the sentence combined with other comments. "This finding demonstrates a consistent spatial pattern that urban OA is more strongly influenced by anthropogenic emissions and is dominated by aromatic species, whereas marine-influenced OA exhibits relatively higher proportions of aged, aliphatic compounds.” (in lines 423–427)

Lines 434-435: “…owing to the identification of abundant $C_nH_{2n-4}O_xS$ and $C_nH_{2n+2}O_xS$ homologues…”

Suggestion: Provide additional context on gas-phase contributions.

Response: Thanks for your suggestion. The abundant reduced sulfur ($C_nH_{2n-4}O_xS$ and $C_nH_{2n+2}O_xS$, $x<4$) might be generated from the oxidation of dimethyl sulfide (DMS) emitted by marine phytoplankton. In addition, fatty acids and other lipid molecules carried in primary organic sea spray aerosols can react with other organic compounds to form the above-mentioned Aliphatic-like OSs. We have added explanations regarding the sources and formation pathways of these S-containing compounds in the corresponding text.

“The elevated abundances of Aliphatic-like OSs and Reduced-sulfur compounds in seaside OA were likely attributed to the oxidation of biogenic reduced sulfur gases, particularly dimethyl sulfide (DMS) emitted from the ocean (Shen et al., 2022; Siegel et al., 2021).” (in lines 463–466)

Additional references:

Shen, J., Scholz, W., He, X.-C., Zhou, P., Marie, G., Wang, M., Marten, R., Surdu, M., Rörup, B., Baalbaki, R., Amorim, A., Ataei, F., Bell, D. M., Bertozzi, B., Brasseur, Z., Caudillo, L., Chen, D., Chu, B., Dada, L., Duplissy, J., Finkenzeller, H., Granzin, M., Guida, R., Heinritzi, M., Hofbauer, V., Iyer, S., Kemppainen, D., Kong, W., Krechmer, J. E., Kürten, A., Lamkaddam, H., Lee, C. P., Lopez, B., Mahfouz, N. G. A., Manninen, H. E., Massabò, D., Mauldin, R. L., Mentler, B., Müller, T., Pfeifer, J., Philippov, M., Piedehierro, A. A., Roldin, P., Schobesberger, S., Simon, M., Stolzenburg, D., Tham, Y. J., Tomé, A., Umo, N. S., Wang, D., Wang, Y., Weber, S. K., Welti, A., Wollesen de Jonge, R., Wu, Y., Zauner-Wieczorek, M., Zust, F., Baltensperger, U., Curtius, J., Flagan, R. C., Hansel, A., Möhler, O., Petäjä, T., Volkamer, R., Kulmala, M., Lehtipalo, K., Rissanen, M., Kirkby, J., El-Haddad, I., Bianchi, F., Sipilä, M., Donahue, N. M., and Worsnop, D. R.: High Gas-Phase Methanesulfonic Acid Production in the OH-Initiated Oxidation of Dimethyl Sulfide at Low Temperatures, Environ. Sci. Technol., 56, 13931-13944. doi:10.1021/acs.est.2c05154, 2022.

Lines 488-490: “…reactions of NO with organic peroxy radicals (generated from monoterpenes oxidation by OH radicals or O_3) during the daytime, or via nighttime

NO_3 radical-initiated reactions…"

Question: Were aqueous/heterogeneous phase reactions considered?

Response: Indeed, aqueous and heterogeneous phase reactions are critical for the formation of organic nitrates in the aerosol phase. Currently proposed major formation pathways for organic nitrates include the addition of nitrate radicals ($NO_3\cdot$) to C=C bonds and the reactions of organic peroxyl radicals with nitric oxide (NO). Particulate organic nitrates are primarily generated by gas-particle partitioning of gaseous organic nitrates and the heterogeneous reaction of $NO_3\cdot$ with organic aerosols (Yang et al., 2025). Our initial analysis focused predominantly on gas-phase oxidation. We have revised the manuscript to explicitly address these multiphase formation processes.

"These organonitrates are mainly formed through the oxidation of VOCs such as alkanes, alkenes and monoterpenes by OH, O_3, and NO_3 in the presence of NO_x (Lee et al., 2016; Ng et al., 2017; Yan et al., 2019; Yang et al., 2025), and subsequently partition into the particle phase. Additionally, particulate organonitrates can also be generated via heterogeneous reactions of organic compounds with NO_3 (Nah et al., 2016)." (in lines 518–523)

Additional references:

Lee, B. H., Mohr, C., Lopez-Hilfiker, F. D., Lutz, A., Hallquist, M., Lee, L., Romer, P., Cohen, R. C., Iyer, S., Kurtén, T., Hu, W., Day, D. A., Campuzano-Jost, P., Jimenez, J. L., Xu, L., Ng, N. L., Guo, H., Weber, R. J., Wild, R. J., Brown, S. S., Koss, A., Gouw, J. de, Olson, K., Goldstein, A. H., Seco, R., Kim, S., McAvey, K., Shepson, P. B., Starn, T., Baumann, K., Edgerton, E. S., Liu, J., Shilling, J. E., Miller, D. O., Brune, W., Schobesberger, S., D'Ambro, E. L., and Thornton, J. A.: Highly functionalized organic nitrates in the southeast United States: Contribution to secondary organic aerosol and reactive nitrogen budgets. P. Natl. Acad. Sci. USA, 113, 1516-1521. doi:10.1073/PNAS.1508108113, 2016.

Nah, T., Sanchez, J., Boyd, C. M., and Ng, N. L.: Photochemical Aging of α-pinene and β-pinene Secondary Organic Aerosol formed from Nitrate Radical Oxidation. Environ. Sci. Technol., 50, 222-231. doi:10.1021/acs.est.5b04594, 2016.

Ng, N. L., Brown, S. S., Archibald, A. T., Atlas, E., Cohen, R. C., Crowley, J. N., Day, D. A., Donahue, N. M., Fry, J. L., Fuchs, H., Griffin, R. J., Guzman, M. I., Herrmann, H., Hodzic, A., Iinuma, Y., Jimenez, J. L., Kiendler-Scharr, A., Lee, B. H., Luecken, D. J., Mao, J., McLaren, R., Mutzel, A., Osthoff, H. D., Ouyang, B., Picquet-Varrault, B., Platt, U., Pye, H.

O. T., Rudich, Y., Schwantes, R. H., Shiraiwa, M., Stutz, J., Thornton, J. A., Tilgner, A., Williams, B. J., and Zaveri, R. A.: Nitrate radicals and biogenic volatile organic compounds: oxidation, mechanisms, and organic aerosol. Atmos. Chem. Phys., 17, 2103-2162. doi:10.5194/acp-17-2103-2017, 2017.

Yang, Y., Huang, L., Zhao, M., Wu ,Y., Xu, Y., Li, Q., Wang, W., and Xue, L.: Multiphase reactions of organic peroxides and nitrite as a source of atmospheric organic nitrates. Nat. Commun.. 16, 5437. doi:10.1038/s41467-025-60696-3, 2025.

Lines 493-495: “…N_2O_5 signals in Case 1…”

Question: How does O_3 concentration compare in Case 1? Is it lower?

Response: The O_3 concentration in case 1 showed a higher value under the influence of regional transport, which led to the formation of more NO_3 radicals by NO_2+O_3. As we mentioned in the text, “During Case 1, elevated O_3 concentrations resulting from regional transport (Fig. S10a) likely facilitated nocturnal NO_3 radical formation via reactions with locally emitted NO_2.”