

Response to Comments from Reviewer #1

We thank you for the constructive comments and suggestions, which are very positive to improve scientific contents of the manuscript. We have revised the manuscript appropriately and addressed your comments point-by-point for consideration as below. The remarks from yours are shown in black, our responses (in blue) and the corresponding edits in the manuscript (in red) are shown below. All the page and line numbers mentioned following are refer to the revised manuscript without change tracked.

Reviewer #1: This manuscript presents an analysis of air pollutant daytime measurements of NO₂, HCHO and CHOCHO obtained using the MAX-DOAS technique in a rural coastal site in Hainan Island, China from June to August 2024. The analysis focuses on the dependence of air pollutant levels to the dominant airflow patterns at the measurement site, and distinguishes between days with and without sea-breeze, as well as days with tropical cyclones. The main results are: (i) days without sea breeze show higher pollutant concentrations compared to sea breeze days; (ii) typhoon days show vertical transport of pollutants to higher altitude; (iii) high values of glyoxal-to-NO₂ and HCHO-to-NO₂ ratios are indicative of a VOC-limited regime, and the glyoxal-to-NO₂ ratio is reliable for studying the ozone formation sensitivity. The article is generally well-written and the performed analysis seems to be robust, although I could not check the details because the MAX-DOAS dataset is not openly accessible. The findings of the paper concern pollutant patterns in coastal environments, but I am not convinced that they can be generalized to other environments. The article could be considered for publication once the following points are adequately addressed in a revised version.

R: We thank you for the constructive and encouraging assessment of our manuscript and for recognizing the value of our analysis. In accordance with the journal's data policy, we will make the MAX-DOAS dataset openly available upon publication to ensure transparency and facilitate independent verification of our results. Regarding the generalizability of our findings, we acknowledge that certain quantitative metrics are specific to Hainan's local conditions; however, the underlying physical and chemical processes identified in this study—such as the SB-driven cooling and suppression of vertical pollutant transport, pollutant redistribution under tropical cyclones, and the diagnostic value of photochemical indicators across different ACPs—are governed by universal mechanisms that also operate in many coastal and island environments. We have clarified these points and explicitly discussed both the limitations and the applicability of our conclusions in the revised manuscript.

Specific comments:

1. 1.158-167: Please mention that the ERA5-L fields are provided at 0.1 degree. Despite the finer resolution of ERA5-L compared to the parent ERA5 dataset, it is still relatively

coarse. An evaluation of the ERA5 or ERA5-L reanalysis against local meteorological measurements at the site should be included. It would be useful to evaluate ERA5-L under cyclonic conditions.

R: Thank you for your comment. We conducted a comparative analysis of temperature and relative humidity (RH) between ERA5 and the nearest meteorological station to our study site—Danzhou station (19.31°N, 109.35°E, 170 m a.s.l.) (Fig. R1). However, it should be noted that although Danzhou station is the closest meteorological site available, it still far away from the measurement site (~33 km). Such distance may also contribute to the discrepancies in temperature and humidity. Overall, the comparison shows relatively high agreement of temperature and RH between these two data sources.

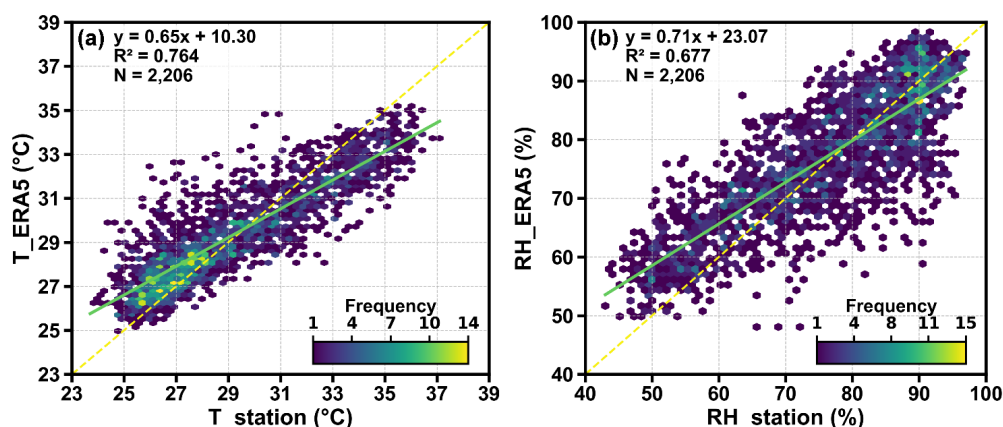


Figure R1. Correlation between Temperature (T_{ERA5} and $T_{station}$) and Humidity (RH_{ERA5} and $RH_{station}$) from ERA5_land and Danzhou Meteorological Station (19.31°N, 109.35°E) during summer 2024. The color of each hexagonal bin represents the frequency of data points within that grid cell. The yellow dashed line indicates the 1:1 line, and the green line shows the regression line between the two datasets.

As shown in Fig. R2, the Danzhou station and ERA5_Land exhibit broadly consistent variations in WS and wind direction WD throughout the study period. The ERA5_Land WS is derived from the 10 m u- and v-wind components and represents grid-averaged conditions, whereas the Danzhou station is influenced by strong local surface roughness caused by nearby buildings and vegetation, resulting in lower observed WS. Despite these differences in magnitude, both datasets display similar prevailing wind directions on non-typhoon days, a shift toward southwesterly winds during typhoon periods, and comparable diurnal patterns in WS, as shown in Fig. R2(b), (c) and (d).

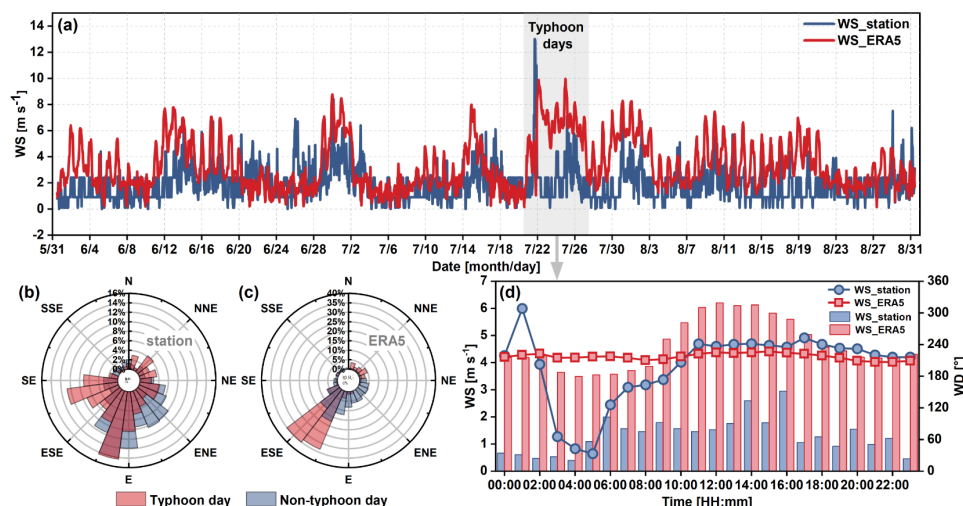


Figure R2. Comparison of WS and WD between the Danzhou station and ERA5 Land. (a) Hourly variations in WS for the Danzhou station and ERA5 Land over the entire observation period; (b) WD patterns at the Danzhou station and (c) ERA5 Land during typhoon and non-typhoon days; (d) Diurnal variations of mean WS and WD for the Danzhou station and ERA5 Land on typhoon days. The gray shaded area indicates the range of the typhoon's duration. Wind direction is plotted in polar coordinates with percentage frequency indicated by concentric circles.

Although ground-based observations can better capture near-surface meteorological conditions, their strong short-term fluctuations—particularly in wind fields—introduce substantial noise and complicate the identification of sea–land breeze circulations. More importantly, the Danzhou station is located far from the measurement site, making its data unrepresentative. In contrast, ERA5 Land provides smoother and more continuous spatiotemporal fields that coherently depict the evolution of large- and mesoscale wind structures. As a result, ERA5 Land has been widely applied in sea–land breeze research worldwide and is generally regarded as reliable and internally consistent for such analyses (Azorin-Molina et al., 2011; Hallgren et al., 2023; Xia et al., 2022; Zhao et al., 2022).

In summary, despite certain uncertainties, ERA5 Land offers physically consistent estimates of all required variables and supplies complete, continuous, and quality-controlled time series—advantages that ground-based observations cannot fully match. The comparison with station measurements further confirms good agreement between these two datasets. These features make ERA5 Land particularly suitable for investigating sea–land breeze circulations in this study.

2. 1.208-210: The existence of additional sources is suggested here to explain the delayed HCHO enhancement (at 10am). Can you give more details? Note that the delayed HCHO peak formation could be due to VOCs that due to their longer lifetimes produce HCHO with delay. Please clarify.

R: Thank you for your comment. It should be clarified that our original statement, "linked to morning traffic emissions," was intended to emphasize this specific photochemical pathway: anthropogenic VOCs emitted by early morning traffic are oxidized by OH radicals under increasing solar radiation, leading to a delayed HCHO

peak around 10:00 local time. We acknowledge that our initial wording was somewhat ambiguous and did not fully convey the meaning. We have therefore revised the manuscript and please refer to Line 243-244.

In the manuscript:

"In contrast, the delayed HCHO peak (10:00 LT, VMRs = 5.14 ppbv) can be attributed to the photochemical oxidation of early morning traffic emitted VOCs under increasing solar radiation."

3. 1.215-218: The residence time of CHOCHO and HCHO are actually quite similar, globally 2.9 h for CHOCHO and 5 h for HCHO. The profile shapes of Fig.3 for both species are also very similar, which contradicts the view expressed in the paper that CHOCHO remains confined within 500 m of the surface because of its shorter lifetime. In addition, it should be noted that methane oxidation contributes to HCHO at the higher levels (especially above 2km), where the CHOCHO levels are close to zero. Can you further elaborate why the lower photolysis rates would contribute to the late afternoon rebound for both species? I wonder why the reduced mixing, as illustrated in Fig.3, is not enough to explain the rebound. After all, reduced solar radiation simultaneously increases the lifetime and depletes the photochemical production of secondary species such as HCHO and CHOCHO. I recommend to include a figure similar to Fig.3 but showing the diurnal variation of the columns. Vertical mixing variations is expected to affect less the columns than the surface concentrations.

R: Thank you for your comment. We reconsider the comparison of HCHO and CHOCHO vertical profiles. In short, HCHO benefits from sustained and spatially widespread secondary production, such as methane oxidation and early-emitted VOC oxidation (Fortems-Cheiney et al., 2012; Liao et al., 2025; Wolfe et al., 2019). By contrast, CHOCHO is formed primarily through oxidation of later-emitted specific VOCs (e.g., aromatics and isoprene) (Chan Miller et al., 2017; Fu et al., 2008; Myriokefalitakis et al., 2008). Additionally, the much larger Henry's law constant of CHOCHO leads to more efficient uptake by humid aerosols, which further limits its upward transport (Du et al., 2025; Kroll et al., 2005; Sander, 2015; Waxman et al., 2015). Together, these factors offer a more plausible explanation for the different daytime vertical distributions of the two species. From the diurnal variation, it's easy to find that HCHO shows considerable abundance up to 800~1000 m while CHOCHO is more confined to lower altitudes, as shown in the left panels of Fig. 4 (b) and (c). We have revised the manuscript accordingly to clarify these points and remove the earlier ambiguity. Please refer to Line 249-252.

"In contrast, CHOCHO remains confined to lower altitudes (<500 m) (Fig. 4d, left), mainly because its weaker secondary formation at higher altitudes compared to HCHO and stronger uptake by humid aerosols that depletes its abundance (Fortems-Cheiney et al., 2012; Liao et al., 2025; Sander, 2015; Waxman et al., 2015; Wolfe et al., 2019)."

We acknowledge that explaining the late-afternoon rebound of HCHO and CHOCHO solely by reduced photolysis is insufficient, as weaker solar radiation also suppresses

their photochemical production, complicating the determination of source and sink contributions. According to your suggestion, we have added Fig. R3 (Fig. S9) to show the diurnal variations of surface AEC, NO₂, HCHO, and CHOCHO VMRs and column-integrated AOD, VCDs of NO₂, HCHO, and CHOCHO. The results indicate that both surface and column abundances of these species increase in the late afternoon, inferring that reduced mixing can not explain the rebound of these species enough. Meanwhile, the weakened photo-induced sink and enhanced anthropogenic emissions like traffic and residential cooking may increase the surface concentration and also contribute to the total column abundance. Nonetheless, reduced atmospheric mixing remains an important factor, allowing pollutants to accumulate near the surface. We have accordingly expanded the discussion in the main text and please refer to Line 252-254.

"The late afternoon rebound in HCHO and CHOCHO concentrations may be due to the combining effects of increased anthropogenic emissions of traffic and cooking activities, as well as reduced atmospheric mixing with temperatures drop and solar radiation decreases (Fig. S9)."

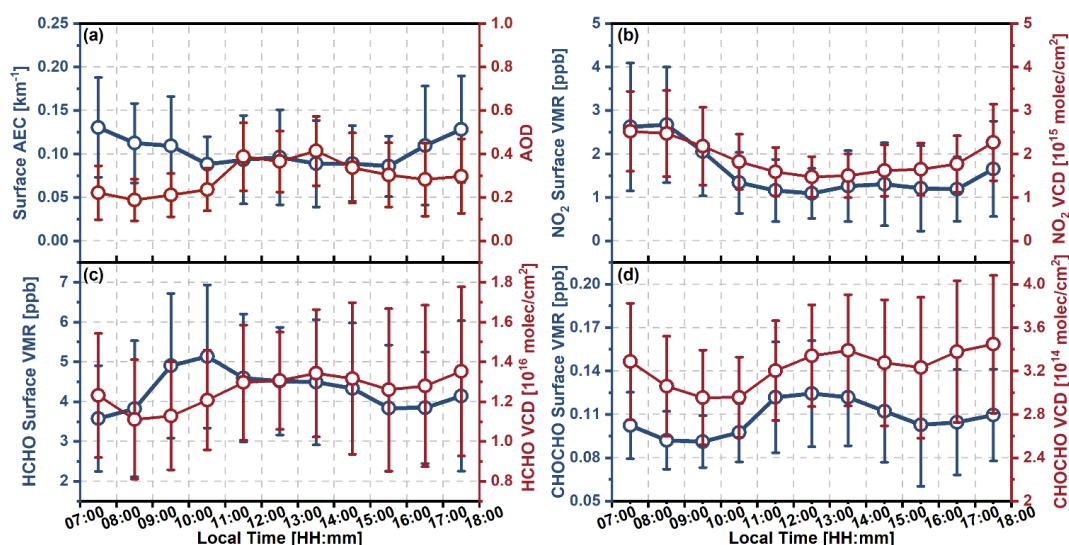


Figure R3. Diurnal cycles of (a) surface AEC and AOD, along with surface VMRs and VCDs of (b) NO₂, (c) HCHO and (d) CHOCHO.

4. The surface VMRs for HCHO and CHOCHO are different in Section 4 and in Section 3.1. Which one is correct?

R: Thank you for your comment. We would like to clarify that the surface VMRs of HCHO and CHOCHO reported in Section 3.1 refer to observational values from previous studies conducted in Beijing and Guangzhou. The values presented in Section 4 correspond to the actual observations obtained in this work. We have revised the manuscript accordingly to make this clearer. Please refer to Line 208-210.

"Notably, HCHO and CHOCHO concentrations (4.33 ± 1.07 ppbv and 0.10 ± 0.02 ppbv, respectively) are comparable to values reported for summertime metropolitan regions, such as Beijing (HCHO: 4.41 ppbv) and Guangzhou (CHOCHO: 0.13 ppbv) (Hong et

al., 2024)."

5. The manuscript attributes the high glyoxal-to-NO₂ and HCHO-to-NO₂ ratios to BVOC sources and suggests that they are dominant compared to anthropogenic sources. Could you include bottom-up emission maps of BVOC, anthropogenic NO_x and VOCs over the island?

R: Thank you for your comment. We have included Fig. S16 (Fig. R4) in the Supplement materials to show the bottom-up emissions of isoprene, anthropogenic NO_x, and VOCs over the island during summer 2024. The data are obtained from the CAMS with a spatial resolution of 0.25°×0.25° for isoprene and 0.1°×0.1° for anthropogenic NO_x and VOCs (Copernicus Atmosphere Monitoring Service) global emission inventories (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-inventories?tab=overview>). It should be noted that CAMS currently provides isoprene as the only BVOC species for 2024, while other BVOC components are not yet available. Anthropogenic NO_x and VOCs represent the total emissions from 12 major source sectors.

As shown in Figure R4, the isoprene emission intensity at the observation site is lower than that in densely vegetated areas such as Wuzhi Mountain. However, both anthropogenic NO_x and VOCs emissions in this region are much lower than those in urban areas (e.g., Haikou and Sanya) and are also lower than the local isoprene flux. This emission pattern explains the relatively high glyoxal-to-NO₂ and HCHO-to-NO₂ ratios, especially considering that other BVOC sources (e.g., monoterpenes and sesquiterpenes) are not included. In addition, the spatial distribution clearly indicates that the upwind regions influenced by sea-breeze and typhoon flows are characterized by higher isoprene emissions and relatively low anthropogenic VOCs sources, supporting our conclusion that BVOC dominate in this area. The corresponding emission maps and related discussion have been added to the revised manuscript. Please refer to Line 193-195 and Line 415-419.

"Monthly emission fields of isoprene (0.25°×0.25°), anthropogenic NO_x, and VOCs (0.1°×0.1°) were also obtained from the CAMS (Copernicus Atmosphere Monitoring Service) global emission inventories (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-inventories?tab=overview>)."

"As shown in Fig. 9d, all ACPs demonstrated mean R_{GF} values < 0.04 across altitudes, indicating BVOC dominance in the FK, which can be supported by the bottom-up emission maps from CAMS reanalysis data (Fig. S16). It shows higher emission than anthropogenic NO_x and VOCs emissions—especially considering that other BVOC species were not included here, e.g., monoterpenes and sesquiterpenes."

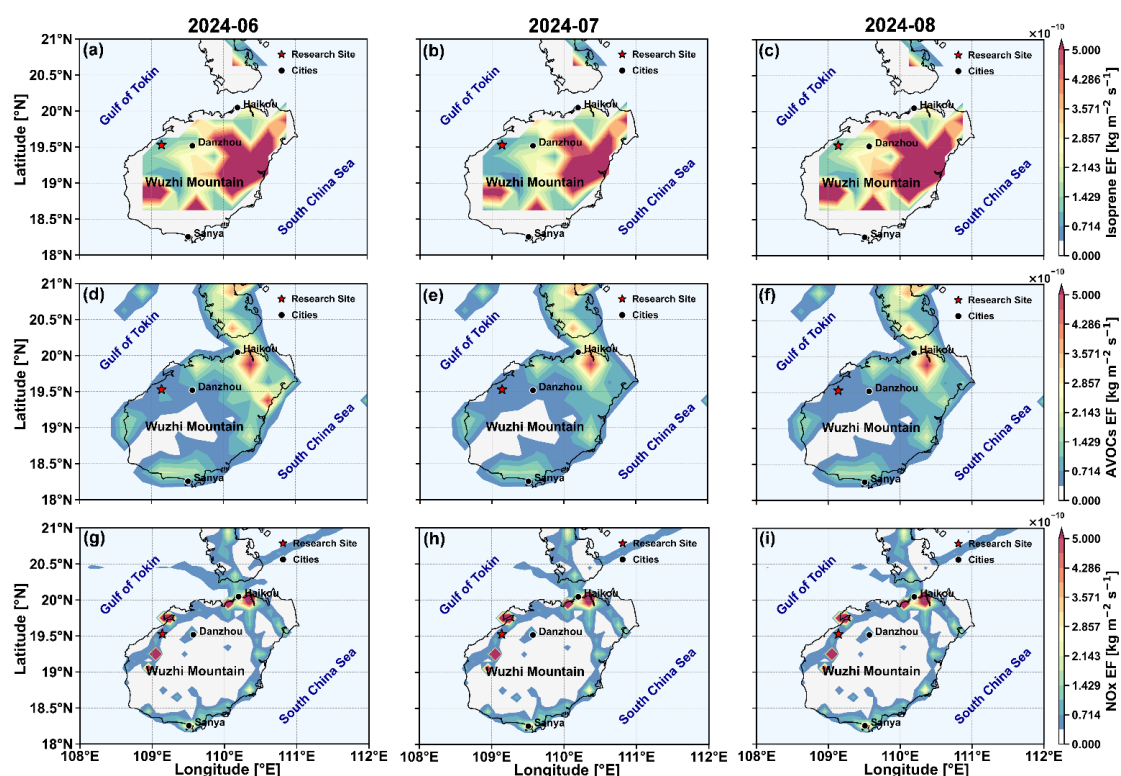


Figure R4. Monthly mean of (a–c) isoprene, (d–f) anthropogenic VOCs (AVOC), and (g–i) anthropogenic NO_x emission fluxes (EF) over Hainan Island for June–August 2024.

6. The manuscript should include a comparison with air pollutant levels from other coastal locations or islands based on literature studies. Besides the typhoon days, what is special about Hainan Island? Can we generalize the findings to other locations?

R: Thank you for your comment. We have appropriately supplemented the manuscript with a comparative analysis of air pollutant concentrations between this study and other coastal regions or islands. Please refer to Line 201-214.

"The observed summer aerosol loading (mean AEC and AOD measuring 0.11 ± 0.03 km⁻¹ and 0.29 ± 0.1 , respectively) exhibited markedly lower values compared to urban agglomerations like Beijing and Shanghai (where AOD is around 0.4) (Fan et al., 2025; Peng et al., 2025), and slightly higher than in coastal Thailand, where monsoonal rainfall efficiently scavenges aerosols (Peengam et al., 2025). The average NO₂ surface VMRs (1.61 ± 0.53 ppbv) was 3 to 5 times lower than the reported values in major Chinese megacity centers, such as the Beijing–Tianjin–Hebei region (7.62 ± 1.39 ppbv) and the Yangtze River Delta (7.45 ± 0.87 ppbv) (Lou et al., 2025; Ministry of Ecology and Environment of the People's Republic of China, 2024a, b, c), reflecting minimal local traffic and industrial emissions. Notably, HCHO and CHOCHO concentrations (4.33 ± 1.07 ppbv and 0.10 ± 0.02 ppbv, respectively) are comparable to values reported for summertime metropolitan regions, such as Beijing (HCHO: 4.41 ppbv) and Guangzhou (CHOCHO: 0.13 ppbv) (Hong et al., 2024). This likely stems from biogenic emissions from surrounding tropical ecosystems compensating for diminished anthropogenic sources. However, the observed HCHO and CHOCHO substantially

higher than values reported in less anthropogenically influenced islands and coastal regions, including the coastal area of Mt. Lao, Qingdao, the Galápagos Islands, and Cape Verde (Lawson et al., 2015; Mahajan et al., 2014; Mahajan et al., 2010; Zhao et al., 2024)."

Similar to the Community Comment #14, we have added a dedicated paragraph in the conclusion section outlining the extent to which our findings can be generalized and the key limitations of our study. Please refer to Line 535-543.

"This study reveals several summertime physical and chemical processes that are characteristic of Hainan Island, so the quantitative values reported here—such as pollutant concentrations, ORA parameter ranges and FNR/GNR thresholds—are shaped by the island's unique land–sea configuration, monsoon regime, and emission environment and should be treated as site-specific. However, the SB-driven cooling and suppression of vertical pollutant transport, the typhoon-related scavenging, redistribution, and uplift, and the diagnostic value of different photochemical indicators under different ACPs are processes that commonly occur across many coastal and island settings. Thus, while specific numerical thresholds may vary, the underlying conceptual processes and the ORA framework offer a transferable basis for interpreting similar coastal phenomena, particularly in regions with comparable climatic and geographic conditions."

Technical comments:

7. 1.40: read 'due to the fact that it originates...'

R: Thank you for your comment. We have revised the sentence and please refer to Line 49:

"NO₂ concentrations typically decrease exponentially with altitude due to the fact that it originates mainly from surface-level anthropogenic emissions, ..."

8. 1.47: remove 'regimes' (repetition)

R: Thank you for your comment. We have removed the repeated word. Please refer to Line 56-57.

9. 1.52: 'Coastal atmospheric environments are significantly influenced by sea-land breeze circulation and typhoons': Typhoons only affect a specific region of the globe. Rephrase.

R: Thank you for your comment. We have revised the sentence to clarify that. Please refer to Line 62-63.

"Coastal atmospheric environments are significantly influenced by sea-land breeze (SLB) circulation and also typhoons in some regions (e.g., the western North Pacific)."

10. 1.63: 'in island', something is missing here

R: Thank you for your comment. We have revised the sentence to correct the grammatical structure. Please refer to Line 72-75.

"Nevertheless, research gaps persist regarding vertical distributions of NO₂, HCHO, and CHOCHO under SB and typhoon conditions, particularly in island where complex topography and unique atmospheric environments may cause these pollutants to behave differently than over continental or coastal regions."

11. 1.64: 'located far from mainland China', be more specific

R: Thank you for your comment. We have updated the description and please refer to Line 76.

"Hainan Island is located away from China's mainland and lies approximately 20 km from the nearest point in Guangdong Province."

12. 1.64-72: Add information about the island (area, population, large cities, powerplants, fraction of vegetation)

R: Thank you for your comment. As suggested, we have integrated key information about the island. Please refer to Line 77-81.

"The island spans approximately 32,900 km² and supports a population of 10.48 million people concentrated in coastal cities (e.g., Haikou, Sanya, and Danzhou), while also maintaining dense vegetation with over 60% forest coverage featuring unique tropical ecosystems (Hainan Provincial Bureau of Statistics, 2024). Adjacent to the South China Sea, its topography and geographic position render the island frequently influenced by SB and typhoons (Fu et al., 2023; Liang and Wang, 2017; Zhang et al., 2014)."

13. 1.66: 'Given the superior air quality', sounds weird. Replace 'superior' by 'good'

R: Thank you for your comment. We have replaced "superior" with "good" in the revised manuscript and please refer to Line 81.

14. 1.72: 'are still unclear'. Replace by 'are not investigated yet'

R: Thank you for your comment. We have revised the text accordingly, and please refer to Line 86-87.

15. 1.73: read 'measurements'

Response: Thank you for your comment. We have corrected the text to "measurements." Please refer to Line 88.

16. l.83: Beibu Gulf is not shown on Fig.1

Response: Thank you for pointing this out. Since "Beibu Gulf" is the Chinese name for the region, while "Gulf of Tonkin" (in Fig. 1) is the internationally recognized name for this region. To ensure consistency and avoid confusion, we have revised the manuscript to use the internationally recognized name, "Gulf of Tonkin", throughout both the text and figures. Please refer to Line 97-98.

"The observation site (altitude approx. 100 m above sea level) lies within a topographically complex region bordered to the northwest by the Gulf of Tonkin and to the southeast by the Wuzhi Mountain (Fig. 1a)."

17. l.83: 'Wuzhi mountain (18.9°N, 109.7°E)'. What are these coordinates?

R: Thank you for your comment. We have removed the coordinates, as they specifically refer to the Wuzhi Mountain main peak and are not necessary for the context. Please refer to Line 98.

18. l.91: add an 'r' to 'dime'

R: Thank you for pointing this out. We have corrected the typo, so it now reads "dimer". Please refer to Line 107.

19. l.97: remove 's' from 'Supplements', here and elsewhere in the paper

R: Thank you for your comment. We have removed the extra "s" and revised all instances throughout the manuscript and please refer to Line 113, 171 and 354.

20. Fig.1a: The color of highways is orange, not yellow

R: Thank you for your comment. We have revised the figure caption to correctly describe the highway color as orange instead of yellow. Please refer to Line 137.

21. l.146-156: Fig.S4 is central to the analysis and the filters mentioned are not explained in the main manuscript. Either include a new section on ORA and move Fig.S4 to the main manuscript, or keep it as is and include a brief description on the filters in the main manuscript. Could you also provide the percentage of the days identified as sea breeze days or refer to the relevant section?

R: Thank you for your comment. Following your suggestion, we have moved Fig. S4 to the manuscript as Fig. 2 now shown in Section 2.2. To keep the fluency and readability of the manuscript, we have kept the detailed parameter definitions and justifications in the Supplementary Text S1, while the main manuscript now includes a

concise description of the purpose and function of each filtering step for clarity. In addition, the specific dates identified as sea-breeze days are provided in Table S3 of the Supplement, and the percentage of such days is discussed in Section 3.2. Please refer to Line 170-171.

"The specific functions of the modules and filters are detailed in Text S2 (Supplement) and identification results are summarized in Table S3 and Section 3.2."

22. 1.170: 'series daily', add 'of' between the two

R: Thank you for your comment. We have corrected the phrase to be "series of daily." Please refer to Line 198.

23. 1.179: 'This likely stems from biogenic emissions'. Could you provide biogenic emission maps?

R: Thank you for your comment. We have added emission maps of isoprene, anthropogenic NO_x, and VOCs in the Supplementary Material (Fig. S16) and incorporated corresponding discussions in the revised manuscript. Please also refer to response to Comment #5 for more details above.

24. 1.181: 'refined', can you be more specific?

R: Thank you for your comment. We have revised the manuscript to specify the temporal resolution more clearly and please refer to Line 215.

"At an hourly temporal resolution (Fig. S6), HCHO and CHOCHO exhibited.... "

25. 1.206: Replace 'The early peaks' by 'The high HCHO and CHOCHO morning levels'

R: Thank you for your comment. We have revised the text accordingly, and please refer to Line 241-242.

26. 1.207: 'in the previous night', replace by 'during daytime'

R: Thank you for your comment. We have revised the text, replacing "in the previous night" with "during daytime" as suggested. Please refer to Line 242.

27. 1.209: remove 'beyond this mechanism'

Response: Thank you for your comment. We have removed it as suggested. Please refer to Line 244.

28. 1.212: replace 'A' by 'a'

Response: Thank you for your comment. We have corrected the capitalization, and please refer to Line 247.

29. Section 3.3. Move Table S5 to the main manuscript to quick reference

Response: Thank you for your comment. In accordance with your suggestion, we have moved Table S5 to Section 3.3 as Table 1 in the main manuscript.

30. 1. 326: Yang et al. 2024. Replace by a textbook reference here

Response: Thank you for your comment. We have updated the citation and please refer to Line 367.

31. 1.326: add space 'land'

Response: Thank you for your comment. We have added the missing space. Please refer to Line 367.

32. 1.379: 'to the local area'. Do you mean the measurement site?

Response: Thank you for your comment. Yes, we mean the measurement site. We have revised the text to "to the measurement site" for clarity. Please refer to Line 423.

33. 1.456: '...several megacities', Please provide reference here

Response: Thank you for your comment. We have added an appropriate reference to support the statement regarding several megacities and please refer to Line 502.

Reference:

- Azorin-Molina, C., Tijm, S., and Chen, D.: Development of selection algorithms and databases for sea breeze studies, *Theor. Appl. Climatol.*, 106, 531-546, <https://doi.org/10.1007/s00704-011-0454-4>, 2011.
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