Response Letter

Dear Editors:

We sincerely thank you for facilitating the review process and for providing us with valuable feedback. The comments we received have been instrumental in enhancing the quality and clarity of our manuscript.

We have carefully considered all the comments and have made comprehensive revisions to address each point raised. Our detailed responses to the reviewers' comments are provided below.

Yours sincerely,
Zining Yang and co-authors

Reviewer #2

General comments:

- ***Yang et al. investigate the role of Chaohu Lake in modulating surface $PM_{2.5}$ concentrations over the lake and the surrounding regions. They do so using high-resolution WRF-Chem simulations configured both with and without the presence of Chaohu Lake. The simulations indicate that the lake acts as a particle storage zone during the daytime due to suppressed boundary layer height and reduced dry deposition rates, leading to $PM_{2.5}$ accumulation and increased particle lifetime. During nighttime, $PM_{2.5}$ concentrations over land decrease substantially in the presence of the lake, driven by enhanced land-breeze circulation. The authors further examine aerosol sources (primary and secondary) and their contributions to $PM_{2.5}$. I found the methodology to be sound and the interpretation of the results convincing. Uncertainties related to the emission inventory are also documented in detail. Overall, the results highlight the importance of properly representing surface properties for improved air quality forecasting. I recommend publication in Atmospheric Chemistry and Physics after the authors address the following (mostly minor) comments.***

Response: We are grateful to the reviewer for the thorough and careful reading of the manuscript, and for the accurate and comprehensive summary of our study. We are pleased that the reviewer found the methodology sound, the interpretation of results convincing, and the treatment of emission inventory uncertainties sufficiently documented. We also appreciate the reviewer's recognition that our findings highlight the importance of properly representing surface properties for improved air quality forecasting. We have carefully addressed all of the reviewer's subsequent specific comments, as detailed in the responses below. We believe the revisions have further strengthened the manuscript and hope it now meets the standards for publication in Atmospheric Chemistry and Physics.

Specific comments:

- ***1. Line 207: Instead of "Another type of option include…", I suggest using "Other model configurations include…".***

Response: We thank the reviewer for this helpful suggestion to improve the clarity and precision of our language. We have revised the sentence according to the reviewer's recommendation. In the revised manuscript, the original text "Another type of option include…" has been changed to "Other model configurations include…". This revision makes the expression more concise and appropriate for describing model setup choices.

- ***2. Line 214: A brief explanation of why the first five days of the simulations was considered spin-up would be helpful. Additionally, why were the days between 5 and 20 March 2019 selected for this study? Does this period have any particular seasonal characteristics that make it relevant for lake-breeze development.***

Response: We sincerely appreciate the reviewer for this insightful question. We have expanded our explanations in the revised manuscript to clarify the rationale for the 5-day spin-up period and the selection of 5-20 March 2019 as our study period.

1、Regarding the spin-up period

The first five days were designated as spin-up to allow both meteorological and chemical fields to transition from their initial and boundary conditions to states governed by the model's own dynamical, physical, and chemical processes. This is particularly critical for WRF-Chem simulations because $PM_{2.5}$ evolution is influenced not only by emissions and transport but also by the complex interplay of boundary layer development, turbulent mixing, gas-phase chemistry, aerosol thermodynamics, and deposition processes. During this initialization period, the atmospheric chemistry module requires time to establish realistic distributions of gaseous precursors such as NO_x, VOCs, SO_2, and NH_3, as well as aerosol species including sulfate, nitrate, ammonium, and organic aerosols through emissions, chemical transformations, and transport. All these processes need sufficient time to reach relatively consistent and stable states. Previous WRF-Chem studies have demonstrated that 3-5 days is typically sufficient for chemical species concentrations to stabilize and minimize the influence of initial conditions, particularly for secondary aerosols that depend on complex nonlinear photochemical reactions. The spin-up period ensures that the diagnosed lake effects are not dominated by model adjustment artifacts from the simulation initialization phase.

2、Regarding the selection of 5-20 March 2019

This specific period was chosen based on several complementary scientific considerations that make it particularly suitable for investigating lake impacts on $PM_{2.5}$ under polluted conditions in eastern China. First, March represents a pollution season when $PM_{2.5}$ concentrations in the Yangtze River Delta region are typically much higher than in summer, making it easier to identify the impacts of lake-induced meteorological perturbations on air pollution. This timing combines two advantageous characteristics: elevated $PM_{2.5}$ levels characteristic of the broader pollution season extending from October to April, and sufficiently strong lake-land thermal contrasts to drive significant lake-breeze circulations and associated boundary layer adjustments. Specifically, early spring in eastern China exhibits conditions closely relevant to this study. The atmosphere remains within the pollution season, maintaining relatively high $PM_{2.5}$ levels, while the gradually intensifying solar radiation after winter promotes substantial daytime boundary layer development and local thermally-driven circulations. Meanwhile, the lake still exhibits pronounced thermal inertia differences relative to the surrounding land surface. These conditions favor the development of lake-induced mesoscale circulations and are well-suited for analyzing their impacts on $PM_{2.5}$.

Second, the selected 15-day period exhibits meteorological conditions favorable for isolating intrinsic lake effects. The period was characterized by predominantly clear-sky conditions with low total column cloud water and ice content, less than 0.1 kg m^{-2} in most areas, and negligible precipitation with hourly accumulation below 0.5 mm. These stable weather conditions minimize confounding influences from cloud microphysics and wet scavenging, allowing the lake-induced perturbations on boundary layer structure, turbulent mixing, and pollutant distributions to be more clearly detected and attributed. The moderate background winds, typically 2-5 m s^{-1} during this period, allow local lake-breeze circulations to develop without being

completely overwhelmed by synoptic-scale flows, while still maintaining sufficient regional transport to affect $PM_{2.5}$ distributions.

Third, from a practical perspective, given the extremely high computational cost of 1-km resolution convection-permitting WRF-Chem simulations with full aerosol-chemistry coupling, the 10-day analysis period following 5-day spin-up represents a balance between capturing diurnal variations of lake effects and remaining computationally feasible. This duration is sufficient to examine the recurring patterns of lake-breeze development, boundary layer evolution, and their impacts on $PM_{2.5}$ across multiple diurnal cycles under relatively consistent synoptic conditions.

To provide context for our study period selection, we examined ERA5 reanalysis 850 hPa wind fields for three representative months: January representing winter conditions, March representing early spring, and October representing autumn. As shown in Figure S1 (in the revised manuscript or our response to the other reviewer's comments), these three seasons exhibit distinctly different synoptic circulation patterns over eastern China. January is more strongly controlled by winter monsoon circulation, March exhibits a transitional circulation background, while October shows autumn circulation characteristics distinct from the former two. These seasonal differences in background circulation patterns suggest that the intensity and spatial extent of lake-breeze development may vary across seasons. We acknowledge that March corresponds to an early spring transitional circulation background, and the lake-breeze characteristics observed in this study may differ from those under winter, summer, or autumn conditions. Future work will explore how lake effects on air quality vary across different seasons and synoptic regimes. The controlled experimental design with Lake and Nolake simulations employing identical initial and boundary conditions ensures that the synoptic-scale circulation constitutes a common background forcing in both simulations. Therefore, the simulation differences primarily reflect perturbations induced by lake presence under the studied meteorological conditions. This allows us to rigorously quantify the physical and chemical processes through which the lake influences air quality during pollution seasons. The mechanisms revealed in this study are most applicable to weather conditions similar to this spring period.

We have clarified this contextualization in the revised manuscript.In the revised Methods section (Section 2.2), we have added a detailed explanation of this question:

"This period corresponds to the pollution season when $PM_{2.5}$ concentrations are typically much higher than in summer, and lake-land thermal contrasts remain sufficiently strong to drive significant lake-breeze circulations. Importantly, the episode was characterized by predominantly clear-sky conditions, with total column cloud water and cloud ice content remaining at low levels (less than 0.1 kg/m^2 in most areas) and negligible precipitation (hourly accumulation greater than 0.5 mm). These conditions are favorable for isolating the intrinsic lake effects while minimizing confounding influences from cloud microphysics and wet scavenging on $PM_{2.5}$ distributions (detailed spatial distributions are not shown here). Additionally, March represents a transitional season between winter and summer circulation patterns, which facilitates the investigation of interactions among lake-induced meteorological perturbations, boundary layer evolution, and $PM_{2.5}$ pollution. Given the

extremely high computational cost of 1-km resolution WRF-Chem simulations, the 10-day period can capture diurnal variations of lake effects while remaining computationally feasible. It should be noted that ERA5 reanalysis dataset (https://rda.ucar.edu/datasets/ds630.0/, last access: 15 April 2019) indicates significant differences in large-scale circulation across different months during the pollution season (October, January, March) (see Figure S1 in Supporting Information), and the selected period represents springtime transitional conditions with moderate background winds. Therefore, our results should be interpreted as lake effects under specific springtime meteorological conditions, and the lake impact mechanisms may differ in other seasons, rather than being statistically representative of all pollution seasons. The sensitivity experiments employ identical initial and boundary conditions, ensuring that simulation differences primarily reflect perturbations induced by lake presence."

In the revised manuscript (Section 4), we have added a detailed explanation about this discussion:

"The lake effects revealed in this study should be interpreted in the context of the meteorological background and the limitations of the simulation period. This study aims to quantitatively isolate the net lake impacts on $PM_{2.5}$ and identify the underlying physical-chemical mechanisms through high-resolution sensitivity experiments, rather than conducting long-term climatological statistical analysis, given the extremely high computational cost of 1-km resolution WRF-Chem simulations. The selected period in March 2019 corresponds to the pollution season when $PM_{2.5}$ concentrations are typically much higher than in summer, and lake-land thermal contrasts remain sufficiently strong to drive significant lake-breeze circulations. However, this study is not statistically representative of all pollution seasons. The 850 hPa wind fields from ERA5 reanalysis for January, March, and October (Figure S1) indicate significant differences in large-scale circulation patterns over eastern China across winter, spring, and autumn. January is more strongly controlled by winter monsoon circulation, March exhibits a transitional circulation pattern, while October shows distinct autumn circulation characteristics different from the former two. This implies that the intensity, spatial extent, and even the dominant pathways of lake impacts on $PM_{2.5}$ may vary with seasonal circulation backgrounds. Additionally, the study period was characterized by predominantly clear skies and moderate background winds, with weak cloud content and precipitation, which was a deliberate aspect of the study selection strategy to facilitate the isolation of intrinsic lake effects. Although frontal passages can influence $PM_{2.5}$ through wind field reorganization, boundary layer structural adjustments, thermal changes, and wet scavenging processes, this study was not dominated by persistent, large-scale, strong frontal precipitation events, and thus frontal scavenging was not a primary controlling factor in this analysis. Since the Lake and Nolake experiments employ identical initial and boundary conditions, the synoptic-scale circulation constitutes a common background forcing in both simulations, and thus the simulation differences primarily reflect perturbations induced by lake presence. In summary, this study are more applicable to stable weather conditions similar to this springtime transitional period. Future research should systematically evaluate lake impacts on pollutants across multiple seasons and different weather patterns (including frontal events) to establish a more comprehensive understanding of lake-air quality interactions and quantify their seasonal and circulation dependencies."

- ***3. Line 220: The abbreviation "YRD" is not defined in the manuscript.***

Response: We sincerely appreciate the reviewer for pointing out this omission. Following the reviewer's suggestion, we have now explicitly defined the abbreviation "YRD" in the revised manuscript. In Section 2.2, we have added the following definition: "The intermediate domain covers the Yangtze River Delta (YRD) region in East China, consisting of 250 × 250 grid cells (111.8°-121.8°E, 27°-37°N) at 5 km resolution." This definition now appears before all subsequent uses of "YRD" in the manuscript, ensuring clarity for readers.

- ***4. Line 260: How were the quasi-global WRF-Chem simulations on a 1° × 1° grid obtained for use as initial and boundary conditions for trace gases and different aerosol species?***

Response: We thank the reviewer for the attention to the initial and boundary condition setup. The quasi-global WRF-Chem simulations on a 1° × 1° grid were obtained by conducting a quasi-global scale WRF-Chem simulation. The simulation domain consists of 360 × 145 grid cells covering the latitude-longitude range of 67.5°S-77.5°N, 180°W-180°E, with a horizontal resolution of 1° × 1°. After preparing the meteorological fields and emission data, we conducted the quasi-global simulation for the same time period as our regional high-resolution simulation to provide initial fields and lateral boundary conditions for trace gases and different aerosol species for the regional simulation. This nesting approach ensures that our regional high-resolution simulation has reliable background concentration fields.

More details about the quasi-global WRF-Chem simulation can be found in Zhao et al. (2013). We have added the following statement in the revised manuscript (Section 2.2): "More details about the quasi-global WRF-Chem simulation can be found in Zhao et al. (2013b)."

Reference:

Zhao, C., Chen, S., Leung, L. R., Qian, Y., Kok, J. F., Zaveri, R. A., and Huang, J.: Uncertainty in modeling dust mass balance and radiative forcing from size parameterization, Atmos. Chem. Phys., 13, 10733–10753, https://doi.org/10.5194/acp-13-10733- 2013, 2013b.

- ***5. Line 349 and Figure 3: Do the local times correspond to 10 March 2019? A reference date is missing in the manuscript. Alternatively, are these panels averaged over the simulations between 10 and 20 March 2019?***

Response: We thank the reviewer for pointing out this ambiguity. To clarify, all times shown in Figure 3 and throughout the manuscript represent averages over the simulation period from 10 to 20 March 2019, not a single date. We have added explicit clarification in the revised manuscript.

In the main manuscript, we have revised the sentence to:

"The spatial distribution of $PM_{2.5}$ surface concentrations in the Lake experiment and the differences between the Lake and

Nolake experiments at 08:00, 14:00, 20:00, and 02:00 LT, averaged over 10-20 March 2019, is illustrated in Figure 3."

We have also added a clarifying statement in the Section 3.1:

"Unless otherwise specified, all analyses presented in this study represent temporal averages over the 10-day period from 10 to 20 March 2019."

We have also revised all figure captions throughout the manuscript to explicitly indicate that the results represent temporal averages over 10-20 March 2019. This ensures readers clearly understand that the figures show averaged conditions over the entire analysis period rather than the result from a single day.

These revisions ensure consistency throughout the manuscript and eliminate any potential confusion about temporal averaging.

- ***6. Figure S4: By "satellite-derived hourly 5 km resolution ground-level $PM_{2.5}$ for Eastern China, 2018," do you mean that the satellite observations correspond to the year 2018? If so, this does not appear to match the simulation period used in this study.***

Response: We appreciate the reviewer for raising this important question regarding the temporal correspondence of the satellite data. The reviewer is correct: the ChinaHigh$PM_{2.5}$ dataset used in this figure corresponds to the year 2018, which does not exactly match our simulation period of March 2019. We sincerely apologize for not providing adequate clarification on this point in the original manuscript, which may have led to misunderstanding about the intended purpose of this figure.

We want to emphasize that we do not use this satellite dataset for quantitative validation of our simulation results. The temporal mismatch between the satellite observations (2018) and our simulation period (March 2019), combined with the inherent retrieval uncertainties in satellite-derived $PM_{2.5}$ estimates (particularly pronounced over water surfaces where surface reflectance properties differ significantly from land), makes rigorous quantitative comparison both inappropriate and unfeasible. Different meteorological conditions, emission patterns, and seasonal variations between these two periods would confound any attempt at direct quantitative comparison. Instead, we present this satellite data purely for qualitative purposes, demonstrating that the phenomenon of elevated daytime $PM_{2.5}$ concentrations over lake surfaces relative to surrounding urban areas is physically plausible and represents a recurring pattern supported by independent observational evidence, rather than being a spurious artifact of our specific model configuration or simulation setup. Figure S6 shows that daytime lake-surface $PM_{2.5}$ concentrations are comparable to or exceed those in adjacent urban areas, with this feature being most pronounced during 11:00-16:00 local time (Figure S6d-i). This spatial pattern is qualitatively consistent with the accumulation mechanism identified in our simulations of the Lake experiment, lending credibility to our mechanistic interpretation even though the specific magnitude and timing may differ between years.

To address the reviewer's concern and eliminate any potential confusion about the purpose and limitations of this comparison, we have substantially revised and expanded the relevant text in the manuscript. We now explicitly acknowledge

the temporal mismatch, clearly state the purely qualitative nature of this comparison, explain in detail why quantitative validation using this dataset is not feasible, and clarify what insights this comparison does and does not provide. The relevant paragraph has been added to the revised manuscript (Section 3.1):

"To further support the reliability of the simulated $PM_{2.5}$ accumulation over the lake surface, we examine the spatial distribution of satellite-derived hourly high-resolution near-surface $PM_{2.5}$ data over eastern China from the ChinaHigh$PM_{2.5}$ dataset (Wei et al., 2021), which provides hourly near-surface $PM_{2.5}$ concentrations during 08:00–17:00 LT. The corresponding results are shown in Figure S6. It should be emphasized that this comparison is not intended as a quantitative validation of our simulation results. The ChinaHigh$PM_{2.5}$ data used here are from 2018 rather than the simulated period of March 2019, precluding an exact temporal match with our model output. In addition, satellite-derived $PM_{2.5}$ estimates are subject to inherent retrieval uncertainties, particularly over water surfaces, which renders meaningful quantitative comparison with model output unfeasible. Figure S6 is therefore presented purely for qualitative purposes, to demonstrate that elevated $PM_{2.5}$ concentrations over the lake surface relative to surrounding areas represent a physically plausible phenomenon supported by independent observational evidence, rather than an artifact of the model configuration. Figure S6 shows that daytime lake-surface $PM_{2.5}$ is comparable to or exceeds that over adjacent urban areas, with this feature being most pronounced during 11:00–16:00 LT (Figures S6d–i), which is qualitatively consistent with the accumulation mechanism identified in our simulations. We acknowledge that direct quantitative validation of lake-surface $PM_{2.5}$ using in-situ observations would be highly desirable. Unfortunately, in-situ air quality observations over lake surfaces and shoreline areas remain extremely scarce in China, with very limited publicly available data. Targeted observational deployments are discussed further in Sect. 4."

Reference:

Wei, J., Li, Z., Pinker, R. T., Wang, J., Sun, L., Xue, W., Li, R., and Cribb, M.: Himawari-8-derived diurnal variations in ground-level $PM_{2.5}$ pollution across China using the fast space-time Light Gradient Boosting Machine (LightGBM), Atmospheric Chemistry and Physics, 21, 7863-7880, https://doi.org/10.5194/acp-21-7863-2021, 2021.

- ***7. Figure 3 caption: By surface, do you mean the vertical level nearest to the surface?***

Response: We thank the reviewer for seeking this clarification. Yes, by "surface" we refer to the lowest vertical level in the model simulation, which represents the atmospheric conditions nearest to the ground surface. In the WRF-Chem model configuration used in this study, the surface level corresponds to the first model layer, with the model output representing the mean conditions within this layer. To avoid any ambiguity, we have updated this explanation in the revised manuscript to explicitly clarify this definition. The revised caption now reads: "The spatial distribution of $PM_{2.5}$ near-surface concentrations (the lowest model level) in the Lake experiment and the differences between the Lake and Nolake experiments at 08:00, 14:00, 20:00, and 02:00 LT, averaged over 10-20 March 2019, is illustrated in Figure 3."

- ***8. Several details regarding the observational data are missing. For example, Figure S4 in the Supplementary Material should be included in Figure 3, with same color map and limits, to qualitatively demonstrate that the simulated concentrations reasonably represent the observed spatial and temporal variations. In addition, it should be stated whether clouds and associated precipitation occurred during the simulation period, as these factors can affect $PM_{2.5}$ concentrations.***

Response: We sincerely thank the reviewer for this thoughtful suggestion regarding the presentation of observational data. We have carefully considered this recommendation and believe that, while we greatly appreciate the reviewer's intent to strengthen the observational support for our findings, incorporating Figure S6 (previously Figure S4) into Figure 3 with the same color scale and limits would not be appropriate for our study. We explain our reasoning in detail below.

Figure 3 and Figure S6 serve fundamentally different purposes and were not intended for direct comparison. As we clarified in our response to Comment 6, the purpose of Figure S6 is not to provide quantitative validation of our simulated $PM_{2.5}$ concentrations, but rather to demonstrate that the phenomenon of elevated $PM_{2.5}$ concentrations over lake surfaces during daytime is physically plausible. It represents a real atmospheric feature supported by independent observational evidence, rather than an artifact of our model configuration. We recognize that this distinction was not sufficiently clear in the original manuscript, potentially leading to the assumption that a direct quantitative comparison was intended. However, this was never our intent. In response, we have revised Section 3.1 in the manuscript to explicitly clarify the purpose of Figure S6. Several important considerations make this satellite data unsuitable for direct quantitative comparison with our model output. First, the satellite-derived data shown in Figure S6 correspond to 2018, while our simulations represent March 2019. This temporal mismatch means the two datasets reflect different meteorological conditions, emission patterns, and seasonal variations. Second, satellite-retrieved $PM_{2.5}$ concentrations have inherent uncertainties, particularly pronounced over water surfaces where surface reflectance properties differ significantly from land. Given these considerations, we can only use such data qualitatively to assess whether the general spatial pattern we simulate (relatively elevated $PM_{2.5}$ over the lake surface compared to surrounding areas) is physically plausible, rather than attempting to quantitatively validate specific concentration values or temporal evolution. Placing Figure S6 alongside Figure 3 with identical color scales, as the reviewer suggests, would fundamentally misrepresent the purpose of our analysis. It would create the impression that we are conducting a direct quantitative comparison between model output and satellite retrievals, which is neither our intention nor scientifically justified given the temporal mismatch and retrieval uncertainties discussed above and detailed in our response to Comment 6.

Therefore, we believe it is more appropriate to keep Figure S6 with its own color scale optimized to show the spatial patterns in the satellite data, clearly separated from the model-experiment comparisons shown in Figure 3. We have substantially revised the relevant text in Section 3.1 of the manuscript to explicitly clarify that Figure S6 is presented purely for qualitative assessment of physical plausibility rather than quantitative validation, and to explain the specific limitations

that preclude quantitative comparison. The detailed explanation of the temporal mismatch, retrieval uncertainties, and the purely qualitative nature of this comparison can be found in our response to Comment 6 and the corresponding revisions in Section 3.1 of the revised manuscript.

Regarding the broader issue of observational validation. We understand the need for comprehensive assessment of available observational data. As we also addressed in our response to Reviewer 1, we have substantially revised the manuscript to provide systematic model-observation validation. We organize our response into two aspects:

(1) Quantitative validation against urban observations:

We have added a dedicated model evaluation section (Section 2.4 and Section 3.1) that systematically validates our Lake experiment against in-situ observations. Specifically, we selected 10 national air quality monitoring stations operated by the Ministry of Environmental Protection of China within the Hefei area to validate $PM_{2.5}$ surface concentrations, and four automatic weather stations (AWSs) to validate meteorological fields. The locations of these stations are marked in Figure 1b (red dots for pollutant stations, purple dots for AWSs).

As shown in the newly added Figure S4, the model successfully reproduces the temporal evolution of 2-meter temperature and 10-meter wind speed throughout the study period. The simulated 2-meter temperature agrees well with observations, indicating that the model accurately characterizes the surface energy budget and thermodynamic conditions. Figure S5 demonstrates the comparison between simulated and observed $PM_{2.5}$ diurnal variations, showing that the model captures the key features of the observed diurnal cycle well, including nocturnal accumulation under stable boundary layer conditions and daytime concentration decrease driven by boundary layer development and enhanced turbulent mixing. While some overestimation of nighttime concentrations exists (primarily attributed to insufficient representation of turbulent mixing intensity under stable nocturnal boundary layer conditions), the simulated diurnal variations are generally consistent with observations, and the overall performance is satisfactory for investigating lake-urban air quality interactions.

It is worth noting that the Nolake experiment, in which Chaohu Lake is replaced by cropland, is a controlled sensitivity experiment designed to isolate lake effects and does not represent an observable atmospheric state, so independent observational validation is neither feasible nor necessary. Given the overall satisfactory performance of the Lake experiment demonstrated above, the simulation framework is considered reliable, and the lake-induced signals identified through the differential analysis between the two experiments are sufficiently credible to support the discussion in the following sections.

We have added Section 2.4 to introduce new observational meteorological and environmental data:

"2.4 Observational data

2.4.1 Meteorological data

The meteorological data were obtained from automatic weather stations (AWSs), which were established based on the operational standards issued by the China Meteorological Administration (CMA, 2018). The hourly data underwent quality control (QC) by local meteorological bureaus of Anhui, following World Meteorological Organization guidelines (Estevez et

al., 2011). The QC included checks of consistency, such as internal, temporal-spatial, and climatic range validations. These QC data were used to determine daily mean, minimum, and maximum meteorological variables. The AWSs recorded various parameters, including air temperature (T, °C), wind speed (U, m/s), air pressure (P, Pa), and wind direction. In this study, we focus on the 3-hourly 2 m temperature and 10 m wind speed obtained from four AWS stations located in the study region. The four AWS sites are marked by purple solid dots in Figure 1b.

2.4.2 Environmental data

Ground observations of hourly $PM_{2.5}$ surface concentrations during March 2019 were obtained from the website of the Ministry of Environmental Protection of China (MEP of China). As our study concentrates on the Hefei region, we selected 10 monitoring stations within this area for detailed analysis. These stations are marked by red solid dots in Figure 1b.

While hourly observations for both meteorology and pollutants are available, model outputs are provided at 3-hour intervals to balance computational efficiency and storage requirements. Hourly output data would provide higher time resolution but significantly increase storage demands. Given that we ran simulations at 1km resolution, hourly outputs would have generated prohibitively large data volumes. On the other hand, this 3-hour output interval remains sufficient for our primary research objective of investigating the diurnal reversal effect of lake impacts on $PM_{2.5}$ concentrations and elucidating the coupling mechanisms between physical processes (turbulent mixing, dry deposition, local circulation) and chemical processes. This approach effectively captures the distinct daytime pollution enhancement and nighttime purification patterns without losing essential detail for understanding lake-urban air quality interactions. To ensure consistent temporal resolution between model and observations, hourly observations were sampled to match our 3-hour model output intervals."

In the revised manuscript (Section 3.1), we have added a detailed comparison between simulated and observed meteorological variables and pollutant concentrations across all observational stations:

"Before presenting the simulation results of $PM_{2.5}$ surface concentrations over lake and urban areas during daytime and nighttime, a systematic evaluation of the Lake experiment is first conducted to verify the capability of the simulation framework in reproducing real atmospheric conditions with the lake present. The evaluation covers the meteorological fields and $PM_{2.5}$ surface concentrations during the study period from March 10 to 20, 2019, which are compared against in-situ observational data averaged over 10 MEP sites in Hefei.

The model's performance in reproducing meteorological conditions is assessed by comparing the simulated 10-meter wind speed and 2-meter temperature with observational data from four AWSs in the Hefei region, as shown in Figure S4. Overall, the model performs well in simulating both variables and successfully reproduces the temporal evolution throughout the study period. The simulated 2-meter temperature agrees well with observations, indicating that the model accurately characterizes the surface energy budget and thermodynamic conditions that form the physical basis for analyzing the lake-land thermal contrast in this study, as shown in Figure S4a. The model overestimates peak wind speed during the strong wind event around March 20, likely attributable to complex mesoscale interactions. Nevertheless, this bias does not compromise the overall

assessment of circulation characteristics throughout the study period, as demonstrated in Figure S4b. The model's ability to reproduce $PM_{2.5}$ surface concentrations is then assessed against observational data from 10 MEP sites in the Hefei region. Figure S5 shows the comparison between the simulated and observed diurnal variations of $PM_{2.5}$ averaged over the study period. The model captures the key features of the observed diurnal cycle well, including the nocturnal accumulation of $PM_{2.5}$ under stable boundary layer conditions and the daytime concentration decrease driven by boundary layer development and enhanced turbulent mixing. The simulated diurnal variation are generally consistent with observations, while the overestimation of nighttime concentrations is primarily attributed to insufficient representation of turbulent mixing intensity under stable nocturnal boundary layer conditions in the model (Yang et al., 2025).

It is worth noting that the Nolake experiment, in which Chaohu Lake is replaced by cropland, is a controlled sensitivity experiment designed to isolate lake-induced effects and does not represent an observable atmospheric state, so independent observational validation is neither feasible nor necessary. Given the overall satisfactory performance of the Lake experiment demonstrated above, the simulation framework is considered reliable, and the lake-induced signals identified through the differential analysis between the two experiments are sufficiently credible to support the discussion in the following sections."

(2) Qualitative assessment using satellite observations and discussion of lake-surface observational gaps:

As discussed above and in our response to Comment 6, we use satellite-derived data (Figure S6) purely for qualitative assessment of the physical plausibility of elevated $PM_{2.5}$ over lake surfaces. However, we must acknowledge an important observational gap: direct in-situ air quality measurements over lake surfaces and lakeside regions remain extremely scarce in China, with publicly available data being quite limited. This observational scarcity is a recognized challenge in studying lake-atmosphere interactions and is not unique to our study. While our urban validation demonstrates the model's capability to reproduce atmospheric conditions where observations exist, the most significant lake effects we identify occur precisely over the lake surface and nearshore areas where observational infrastructure is currently absent. Although international research occasionally deploys targeted measurements over lake surfaces, such observations are extremely rare in China.

We have added detailed discussions of this observational limitation and the need for future targeted measurements in Section 4 of the revised manuscript:

"Additionally, the key limitation of current lake-urban air quality research is the scarcity of direct observations over lake surfaces and lakeside regions. Although this study has validated the simulations against urban observation networks, the most significant lake effects we identified occur precisely over lake surfaces and nearshore areas where observational infrastructure is absent. While satellite-retrieved $PM_{2.5}$ products provide qualitative support for lake surface accumulation phenomena, their spatiotemporal resolution and retrieval uncertainties over water surfaces are insufficient to meet the needs for detailed mechanistic validation, underscoring the necessity of systematic field observations. This observational challenge is particularly acute in China. Many major cities have developed along inland lakes, yet systematic lake-atmosphere monitoring remains

extremely limited compared to North America and Europe. Future research should prioritize the establishment of comprehensive observation networks specifically designed for lake-urban pollution gradients. Such networks should include monitoring stations deployed at multiple locations along lakeshores and cross-sectional observations along lake-urban corridors (such as the A-B-C transect examined in this study) to measure $PM_{2.5}$ concentrations, chemical composition, and meteorological parameters. Lake-based platform observations (buoys or low-altitude drones) can capture spatial heterogeneity and transient features such as lake-breeze fronts, while vertical profiling measurements (tethered balloons, drones, or ground-based remote sensing) can observe boundary layer structure and lake-land breeze circulation. Additionally, measurements of dry deposition velocities and surface fluxes over both lake and land surfaces, combined with dense low-cost sensor networks monitoring fine-scale spatial patterns, will provide multidimensional data support for understanding lake effects. These observations will not only directly validate the lake-induced $PM_{2.5}$ gradients and vertical mixing signals identified in this study but also reveal small-scale turbulent mixing and chemical transformation mechanisms. Filling the observational gap in lake environments represents a critical frontier for advancing air quality research in rapidly urbanizing inland lake regions globally."

The reviewer also raised an important point regarding the potential influence of clouds and precipitation on $PM_{2.5}$ concentrations during the simulation period. We thank the reviewer for this consideration and provide the following clarification:

We have conducted a comprehensive analysis of cloud and precipitation conditions during our study period (March 10-20, 2019). Figure 1 shows the spatial distribution of temporally-averaged total column cloud water and cloud ice content, and Figure 2 presents the corresponding precipitation distribution across different times of the day. The analysis clearly demonstrates that the study period was characterized by predominantly clear-sky conditions with negligible cloudiness across the entire domain. The total column cloud water and cloud ice content remained below 0.1 kg/m² throughout most of the region, with only sporadic and spatially isolated minor cloud patches appearing at certain times. More importantly, precipitation was virtually absent during the analysis period, with domain-wide accumulated precipitation remaining below 0.5 mm/h across nearly the entire region. Only extremely localized and minimal precipitation events (< 2 mm/h) occurred in very limited areas at specific times. These trace amounts of precipitation are insufficient to produce any meaningful wet scavenging effect on $PM_{2.5}$ concentrations. Therefore, we can confidently conclude that neither cloud formation nor precipitation exerted significant influence on the $PM_{2.5}$ distribution patterns and lake effects identified in our study. The clear-sky, dry conditions that prevailed during this episode provide an ideal setting for isolating and examining the thermodynamic and dynamic mechanisms of lake-atmosphere-pollution interactions without the confounding effects of cloud microphysics or wet removal processes.

We have added a clarifying statement in the revised manuscript (Section 2.2) noting: "Importantly, the episode was characterized by predominantly clear-sky conditions, with total column cloud water and cloud ice content remaining at low levels (< 0.1 kg/m^2 in most areas) and negligible precipitation (hourly accumulation < 0.5 mm). These conditions are favorable for isolating the intrinsic lake effects while minimizing confounding influences from cloud microphysics and wet scavenging

on $PM_{2.5}$ distributions (detailed spatial distributions are not shown here). ”

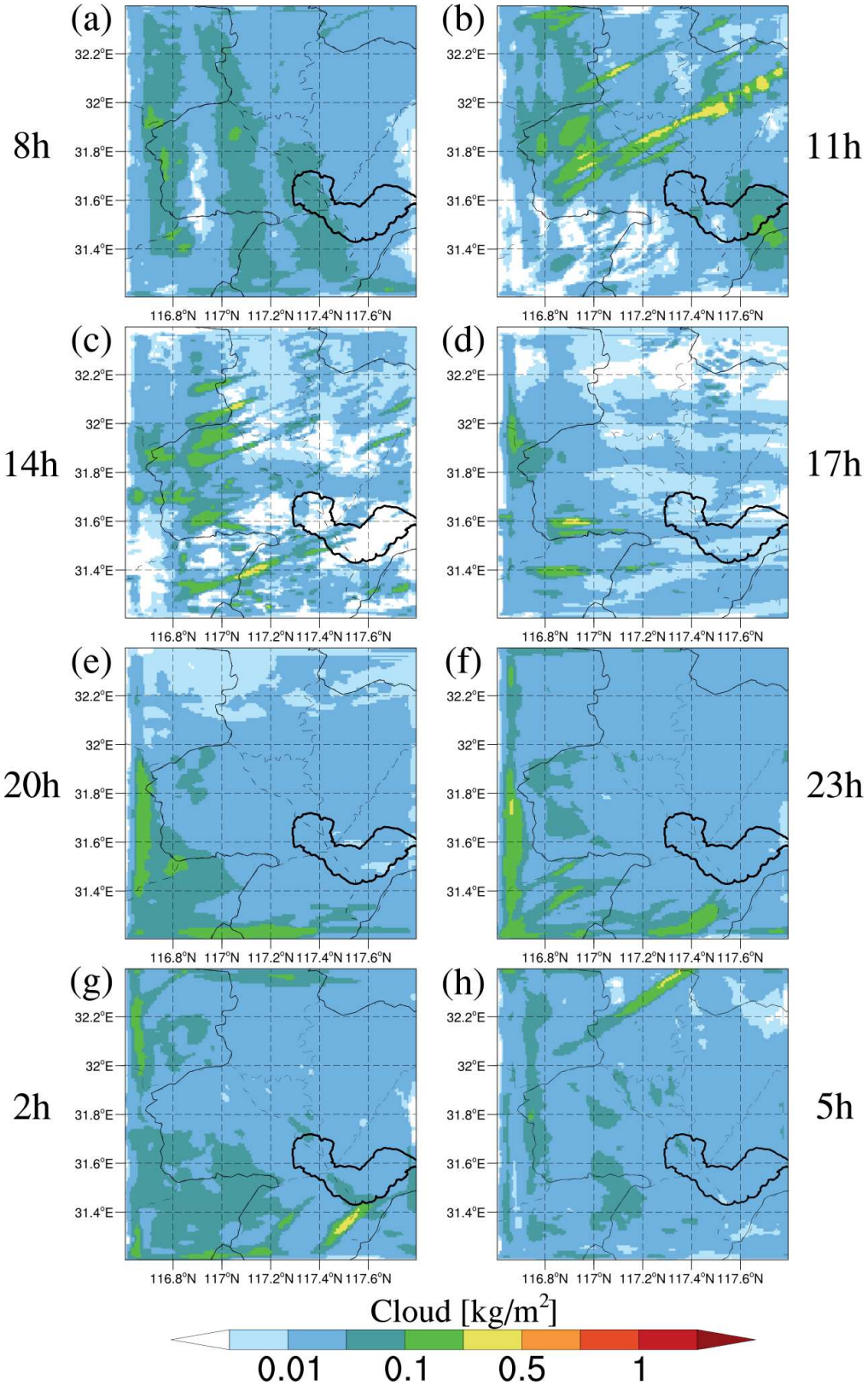

Figure 1. Spatial distribution of total column cloud water and cloud ice content at 08:00, 11:00, 14:00, 17:00, 20:00, 23:00, 02:00, and 05:00 local time (LT), averaged over the study period.

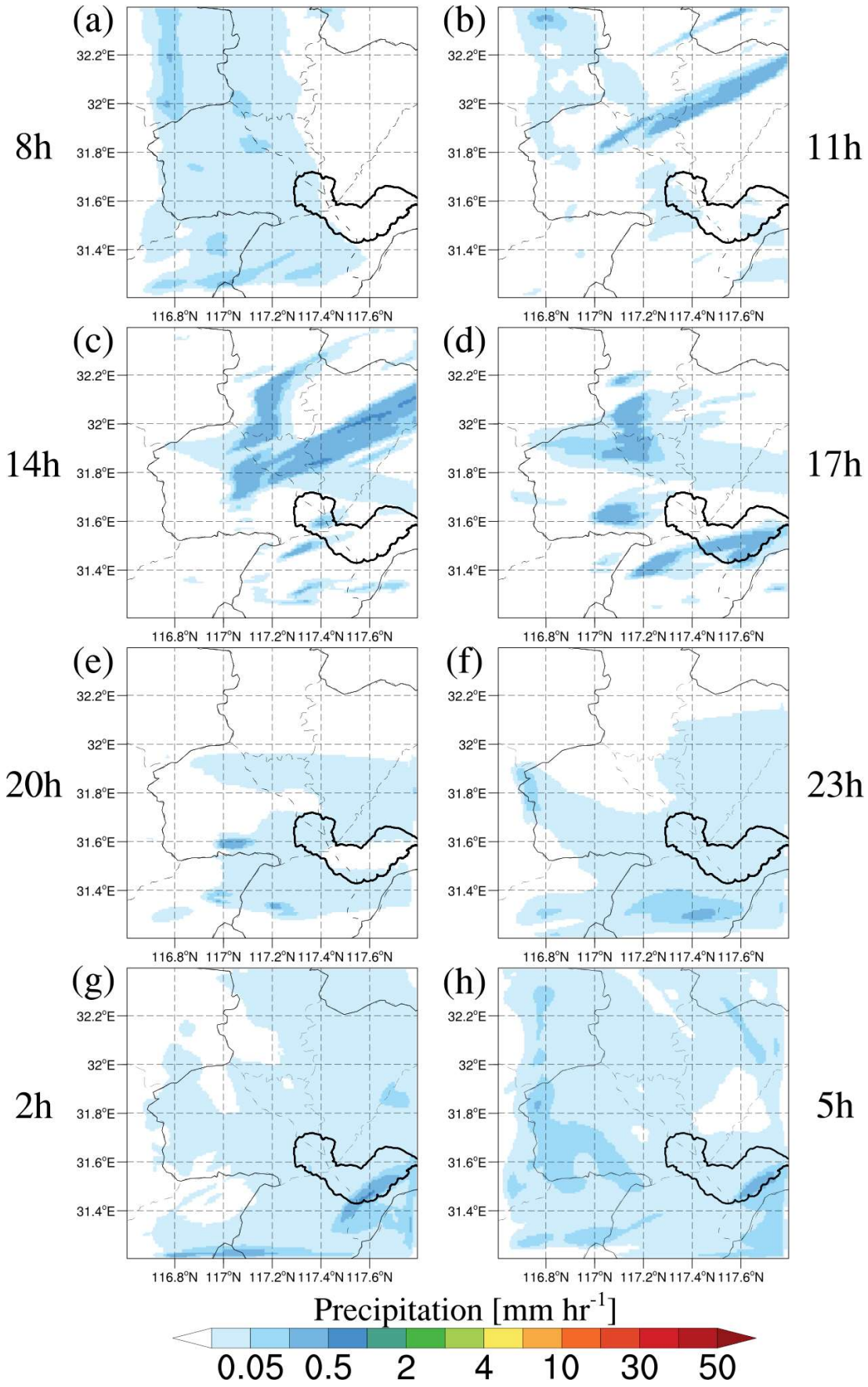

Figure 2. Spatial distribution of precipitation at 08:00, 11:00, 14:00, 17:00, 20:00, 23:00, 02:00, and 05:00 local time (LT) averaged over the study period.

Reference:

CMA, 2018: Technical Specifications for Maintenance of Regional Automatic Weather Stations. QX/T 465–2018. (in Chinese). Available at: http://cmastd.cmatc.cn/standardView.jspx?id=3076. Accessed on 5 May 2022., 2018.

Estevez, J., Gavilan, P., and Giraldez, J. V.: Guidelines on validation procedures for meteorological data from automatic weather stations, Journal of Hydrology, 402, 144-154, https://doi.org/10.1016/j.jhydrol.2011.02.031, 2011.

Yang, Z. N., Du, Q. Y., Yang, Q. K., Zhao, C., Li, G. D. Z., Xia, Z. H., Xu, M. Y., Yuan, R. M., Li, Y. B., Xia, K. H., Gu, J., and Feng, J. W.: Modeling urban pollutant transport at multiple resolutions: impacts of turbulent mixing, Atmospheric Chemistry and Physics, 25, 8831-8857, https://doi.org/10.5194/acp-25-8831-2025, 2025.

- ***9. Line 374: Replace "reasonableness" with "reliability".***

Response: We thank the reviewer for this suggestion to improve the precision of our language. We agree that "reliability" is a more appropriate and scientifically rigorous term in this context. Following the reviewer's recommendation, we have replaced "reasonableness" with "reliability" in the revised manuscript. This revision better conveys the credibility and trustworthiness of our model simulation results.

- ***10. Figure 4 caption: Replace "…as a function of distance from point B" with "as a function of distance from point B toward A, as marked in Figs. 2 and 3.".***

Response: We thank the reviewer for this helpful suggestion to improve the clarity and completeness of the figure caption. Following the reviewer's recommendation, we have revised the caption of Figure 4 in the revised manuscript. The phrase "as a function of distance from point B" has been replaced with "as a function of distance from point B toward A, as marked in Figure S2 and S3". This revision more explicitly indicates the direction of the transect and provides clear reference to where points A and B are defined in earlier figures, making it easier for readers to interpret the cross-sectional results.

- ***11. Section 3.1.2 title: Replace "Component-dependent…" with "Aerosol-species dependent…"***

Response: We thank the reviewer for this suggestion to improve the terminology precision in our section title. We agree that "Aerosol-species dependent" is more specific and scientifically accurate than "Component-dependent" in describing the variations among different aerosol chemical species. Following the reviewer's recommendation, we have revised the title of Section 3.1.2 in the revised manuscript from "Component-dependent…" to "Aerosol-species dependent…" This revision enhances clarity and better reflects the chemical speciation analysis presented in this section.

- ***12. Section 3.2 title: Replace "The influence mechanism of…" with "Factors controlling the variations of…"***

Response: We thank the reviewer for this valuable suggestion to improve the section title. We agree that "Factors controlling

the variations of…" more accurately describes the content of this section and is more direct and concise than "The influence mechanism of…" Following the reviewer's recommendation, we have revised the title of Section 3.2 in the revised manuscript from "The influence mechanism of…" to "Factors controlling the variations of…" This revision better captures the analytical focus of this section on identifying and quantifying the controlling factors.

- ***13. Lines 457–458: Although I understand that secondary $PM_{2.5}$ dominates the concentration differences, the phrase "secondary $PM_{2.5}$ plays a particularly prominent role" is somewhat unclear and could be rephrased for clarity.***

Response: We thank the reviewer for this constructive suggestion. We agree that the original phrase "secondary $PM_{2.5}$ plays a particularly prominent role" was somewhat vague. To improve clarity, we have revised this sentence to more explicitly describe the quantitative difference between secondary and primary $PM_{2.5}$. The revised sentence in the revised manuscript now reads: "During daytime, the difference in secondary $PM_{2.5}$ concentrations between the Lake and Nolake experiments is substantially larger than that of primary $PM_{2.5}$." This revision make the statement more precise and easier to understand.

- ***14. Lines 459–462: It is not clear how this conclusion can be inferred from Figures 5 and 6. The color scale for the differences is limited to 2, and panel (d) in both figures appears nearly identical. I recommend merging Figures 5 and 6 into a single figure and adjusting the color bar limits to better highlight the differences.***

Response: We sincerely thank the reviewer for this valuable comment. We acknowledge the reviewer's concern regarding the color scale limitations and have carefully addressed this issue.

Regarding the color scale range:

The reviewer correctly noted that our original color scale was limited to 0-2 μg/m^3, which may not immediately reveal the 5-10 μg/m^3 and 0-5 μg/m^3 ranges mentioned in the text. To demonstrate that our stated values are accurate, we have prepared additional spatial distribution maps with expanded color bar ranges showing primary and secondary $PM_{2.5}$ at different times (Figures 5A, 5B, S9A and S9B in this response). These figures clearly show the concentration difference ranges (5-10 μg/m^3 for secondary $PM_{2.5}$ and 0-5 μg/m^3 for primary $PM_{2.5}$) discussed in the manuscript, particularly over the lake surface and lakefront areas.

However, we have chosen to retain the original color scale (0-2 μg/m^3) in the main manuscript and supplementary materials for the following reason: while the larger differences (5-10 μg/m^3 and 0-5 μg/m^3) are indeed prominent over the lake and immediate lakefront areas, the concentration differences in areas 5-20 km or farther from the lake range from 2-4 μg/m^3. To ensure that spatial variations across the entire study domain are clearly visible and comparable, the original color scale provides better overall visualization. The expanded color scale figures provided here in our response serve to validate the specific values discussed in the text.

Regarding merging Figures 5 and 6:

We appreciate the reviewer's suggestion to merge these two figures. We agree that these figures are closely related and should be presented together. However, we believe that merging them into a single figure would result in panels that are too small to discern important spatial details. Therefore, we have restructured them as Figure 5A and Figure 5B (Figure S9A and Figure S9B) in the revised manuscript, keeping them as separate but consecutive figures. This arrangement maintains their visual clarity while emphasizing their interconnection. The corresponding changes have been made throughout the revised manuscript.

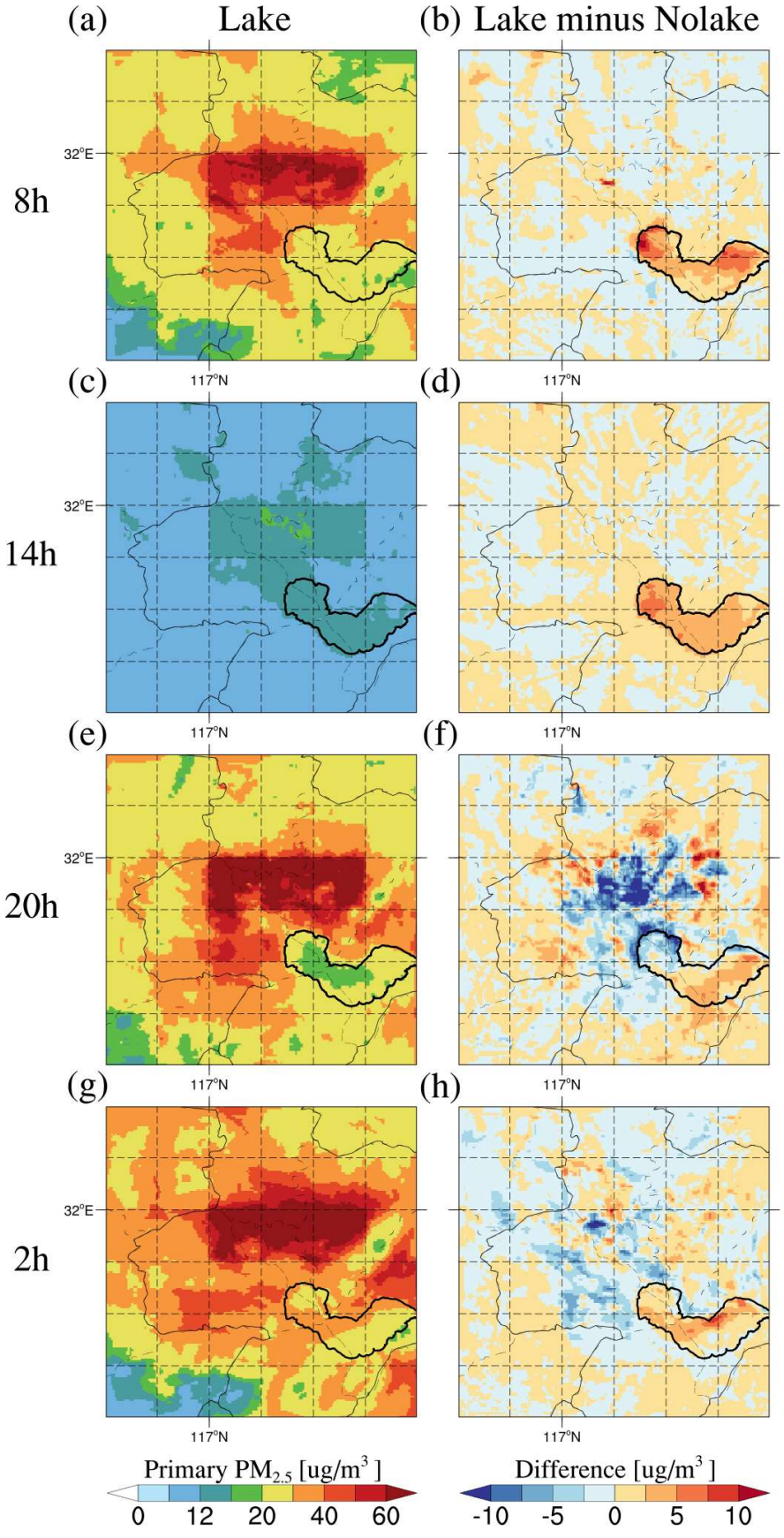

Figure 5A. The spatial distribution of primary $PM_{2.5}$ surface concentrations (sum of black carbon (BC), organic carbon (OC), and other inorganics (OIN)) in the (a、c、e、g) Lake experiment and (b、d、f、h) the differences between Lake and Nolake experiments (Lake minus Nolake) at 08:00, 14:00, 20:00, and 02:00 LT across the study area, averaged over 10-20 March 2019.

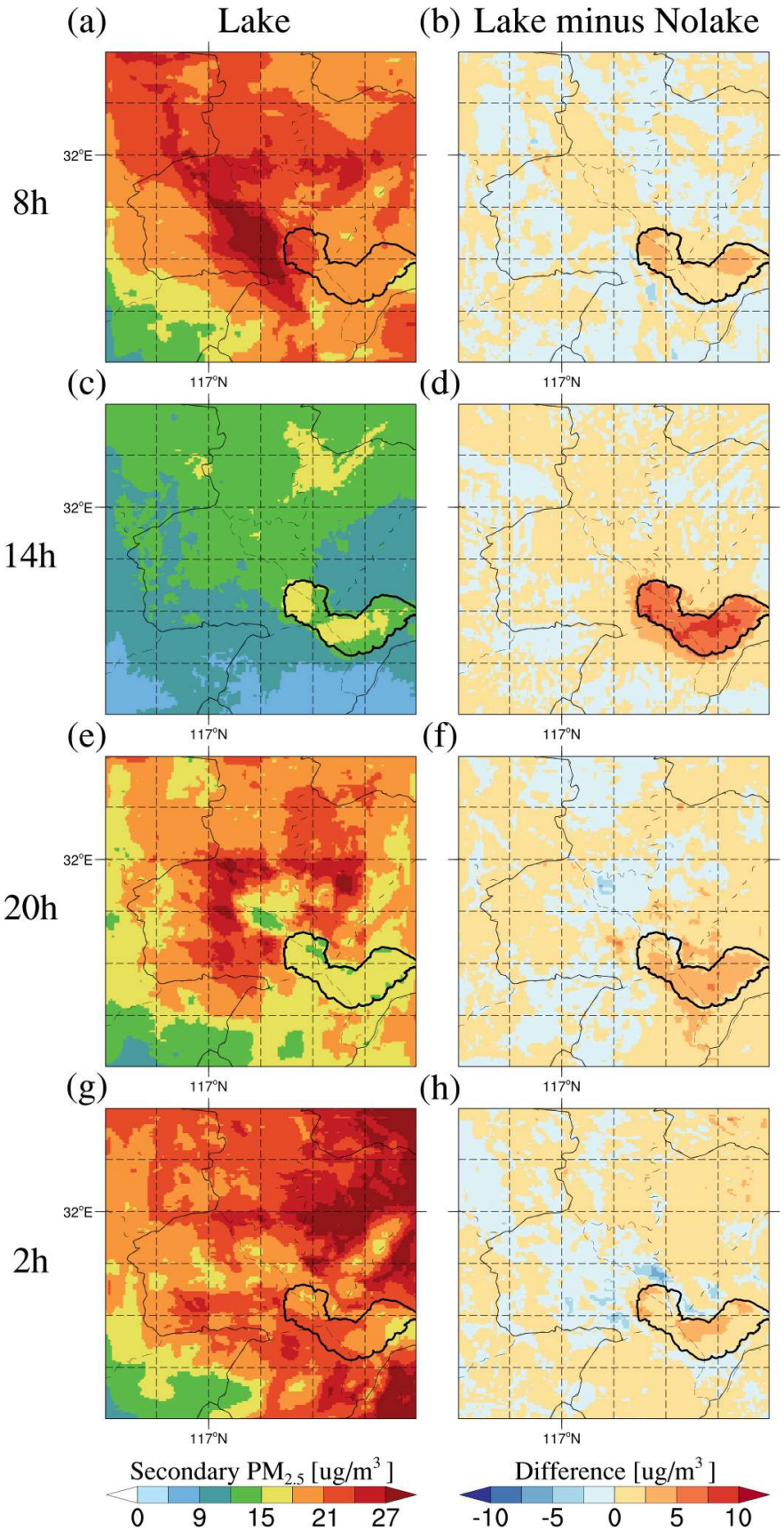

Figure 5B. The spatial distribution of secondary $PM_{2.5}$ surface concentrations (sum of sulfate (SO_4^{2-}), nitrate(NO_3^-), and ammonium (NH_4^+)) in the (a、c、e、g) Lake experiment and (b、d、f、h) the differences between Lake and Nolake experiments (Lake minus Nolake) at 08:00, 14:00, 20:00, and 02:00 LT across the study area, averaged over 10-20 March 2019.

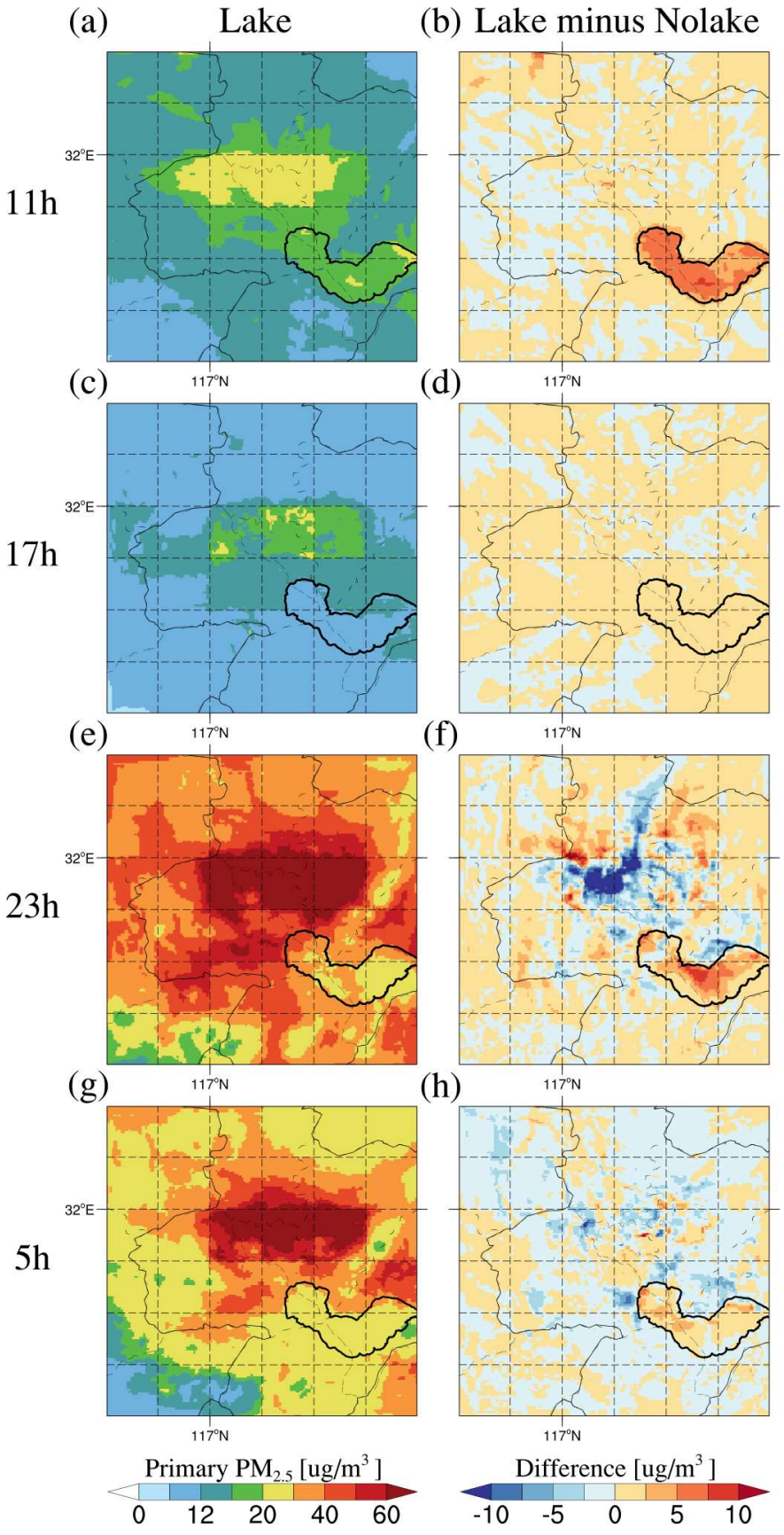

Figure S9A. The spatial distribution of primary $PM_{2.5}$ surface concentrations (sum of black carbon (BC), organic carbon (OC), and other inorganics (OIN)) in the (a、c、e、g) Lake experiment and (b、d、f、h) the differences between Lake and Nolake experiments (Lake minus Nolake) at 11:00, 17:00, 23:00, and 05:00 LT across the study area, averaged over 10-20 March 2019.

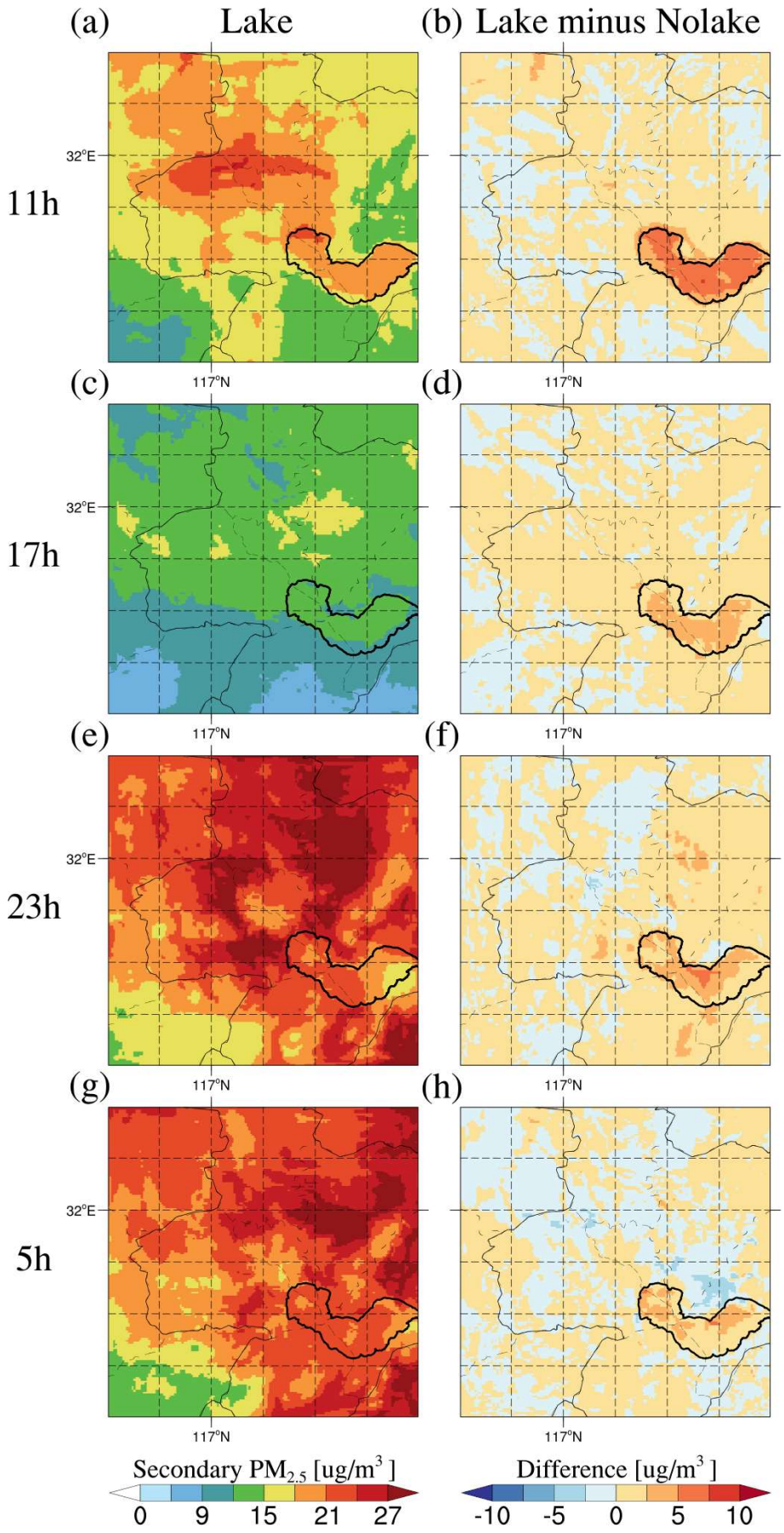

Figure S9B. The spatial distribution of secondary $PM_{2.5}$ surface concentrations (sum of sulfate (SO_4^{2-}), nitrate(NO_3^-), and ammonium (NH_4^+)) in the (a、c、e、g) Lake experiment and (b、d、f、h) the differences between Lake and Nolake experiments (Lake minus Nolake) at 11:00, 17:00, 23:00, and 05:00 LT across the study area, averaged over 10-20 March 2019.

- ***15. Line 461: Replace "degree" with "extent".***

Response: We thank the reviewer for this suggestion to improve the word choice. We agree that "extent" is more appropriate and commonly used in this context. Following the reviewer's recommendation, we have replaced "degree" with "extent" in the revised manuscript. This revision makes the expression more idiomatic and precise.

- ***16. Line 463: On what basis do the authors conclude that the differences between the lake and no-lake experiments stem from pollutant transport from the lake surface? It seems more appropriate to state "transport to the lake."***

Response: We sincerely thank the reviewer for this insightful comment, which has identified an important ambiguity in our original manuscript. The reviewer is absolutely correct that pollutants are first transported "to the lake" from urban source regions, and we greatly appreciate the opportunity to clarify this critical aspect of the pollutant transport mechanism.

We recognize that our original phrasing at Line 463 could be interpreted as suggesting that pollutants originate from the lake itself, which would be inconsistent with the fact that the lake surface has no local emissions. The reviewer's suggestion to use "transport to the lake" appropriately describes the initial movement of pollutants from source regions toward the lake. This is indeed an essential first step in the overall transport process, and we acknowledge that our original text did not adequately clarify this sequence.

Clarification of the complete transport mechanism:

However, we would like to emphasize that the complete pollutant transport process in our study actually involves two distinct and sequential stages, both of which are essential for understanding how the lake influences urban air quality:

Stage 1 - Initial transport to the lake and accumulation:

Pollutants are first transported from pollution source regions (such as urban areas) to the lake surface, which has no local emissions. Once over the lake, these pollutants accumulate substantially near the surface due to suppressed boundary layer height, weak turbulent mixing, and low dry deposition rates. This accumulation process leads to elevated pollutant concentrations over the lake that can even exceed those in urban source areas, forming an anomalous pollution pattern.

Stage 2 - Subsequent transport from the lake back to surrounding areas:

After accumulating over the lake, these concentrated pollutants are then transported back to urban and other surrounding areas through lake-breeze circulation and dispersion mechanisms. This return transport is the key mechanism that explains why urban areas in the Lake experiment show higher $PM_{2.5}$ concentrations compared to the Nolake experiment.

Focus of our analysis:

The reviewer's comment correctly identifies Stage 1 (transport "to the lake"), which we had not explicitly described in our original text. However, the primary focus of our discussion in this section is on Stage 2 (transport "from the lake" back to urban areas), which has been detailed in Section 3.2.3 of the manuscript. This is because Stage 2 is the critical process that

explains the differences observed between the Lake and Nolake experiments in urban areas (the central question of our study). Without the lake, pollutants would not accumulate and subsequently return to urban areas through lake-breeze circulation and dispersion, which is why the Nolake experiment shows lower urban $PM_{2.5}$ concentrations during daytime.

We acknowledge that by not explicitly describing Stage 1, our original text created ambiguity about the complete transport pathway and could lead readers to misunderstand the source of pollutants. The reviewer's comment has helped us recognize that both stages must be clearly articulated to provide a complete understanding of the lake-mediated pollutant transport mechanism.

Revisions made in the manuscript:

To address this issue comprehensively, we have substantially revised this section in the manuscript to explicitly describe both stages of the transport process. The revised text now provides a clear, sequential description of the complete pollutant pathway. In the revised manuscript (Section 3.1.2), we have updated the detailed explanation:

"Furthermore, $PM_{2.5}$ is composed of different components, yet the diurnal variation characteristics of these different components and their response mechanisms to lake influence remain unclear. To investigate the different impact of the lake on various $PM_{2.5}$ components, this study conducted an in-depth analysis of primary and secondary $PM_{2.5}$, as shown in Figures 5A and 5B. During daytime, the difference in secondary $PM_{2.5}$ concentrations between the Lake and Nolake experiments is substantially larger than that of primary $PM_{2.5}$. At 14:00 LT (Figures 5Ac, 5Ad, 5Bc and 5Bd), the increase of secondary $PM_{2.5}$ concentration over the lake is generally 5-10 μg/m³, which is not only significantly greater than the 0-5 μg/m³ increase in primary $PM_{2.5}$, but also extends over a broader range and extent of influence in surrounding areas. This spatial pattern reflects a sequential transport process. Particulate matter is first transported from pollution source regions (such as urban areas) to the lake surface, which has no local emissions. Subsequently, due to the suppressed boundary layer height, weak boundary layer mixing, and low dry deposition rates over the lake, particles accumulate substantially on the lake surface (detailed mechanism explained in Section 3.2.2). Notably, secondary $PM_{2.5}$ concentrations over the lake are 15-18 μg/m³, while secondary $PM_{2.5}$ concentrations in urban areas in the Lake experiment are approximately 12-15 μg/m³, forming an anomalous pollution pattern where lake surface concentrations exceed urban concentrations, which differs dramatically from conventional understanding. Moreover, because the accumulation of secondary $PM_{2.5}$ over the lake during daytime is much greater than that of primary $PM_{2.5}$ (detailed mechanism explained in Section 3.2.4), a greater amount of secondary particles is subsequently transported back to urban and other surrounding areas through lake-breeze circulation and dispersion (detailed mechanism explained in Section 3.2.3), becoming a key factor in exacerbating urban daytime $PM_{2.5}$ pollution. "

In the revised manuscript (Section 3.2.3), we have also added these detailed explanations:

"While physical mechanisms lead to pollutant accumulation, dynamic processes drive the transport and redistribution of these particles. Pollutants are first transported from source regions (such as urban areas) to the lake surface, which has no local emissions. Subsequently, the suppressed boundary layer height, weakened turbulent mixing, and reduced dry deposition

velocity over the lake cause $PM_{2.5}$ to accumulate substantially near the surface, resulting in near-surface $PM_{2.5}$ concentrations in the Lake experiment being substantially higher than in the Nolake experiment over the lake area. " and "The lake's influence extends to surrounding urban areas through complex dynamical transport processes that redistribute the accumulated pollutants both horizontally and vertically back to urban and other surrounding areas. These lake-induced perturbations to urban areas exhibit distinct mechanisms during daytime (horizontal convergence and frontal stagnation) and nighttime (enhanced vertical redistribution). "

Summary:

The bolded additions in the revised text clearly delineate the two-stage process: (1) initial transport to the lake and accumulation (Stage 1, which the reviewer correctly identified), and (2) subsequent transport from the lake back to urban areas (Stage 2, which is our primary analytical focus). This revision clarifies that while pollutants initially move "to the lake" as the reviewer suggested, the differences observed in urban areas between the Lake and Nolake experiments result from the complete two-stage process, with particular emphasis on the return transport "from the lake" that redistributes the accumulated pollutants back to urban areas. We believe this revised description resolves the ambiguity identified by the reviewer and provides readers with a complete and accurate understanding of the lake-mediated pollutant transport mechanism. We thank the reviewer again for this valuable comment, which has significantly improved the clarity and completeness of our manuscript.

- ***17. Line 472: Additionally, air masses transported over the lake may spend a longer time in the atmosphere, increasing the likelihood of secondary aerosol formation. This process could contribute to the larger differences between the lake and no-lake simulations for secondary aerosols.***

Response: We sincerely thank the reviewer for this valuable comment regarding the mechanisms behind the larger differences in secondary aerosol concentrations between the Lake and Nolake experiments. The reviewer suggests that "air masses transported over the lake may spend a longer time in the atmosphere, increasing the likelihood of secondary aerosol formation." We appreciate this consideration and would like to clarify the actual mechanisms that explain why secondary aerosols show substantially larger lake-induced differences compared to primary aerosols.

Stage 1 - Transport from urban source regions to the lake surface

As discussed in detail in our previous response (Response 16), both primary and secondary $PM_{2.5}$, along with their gaseous precursors, are transported from urban source regions to the lake surface by regional winds. However, the reviewer's suggested mechanism, that extended atmospheric residence time during transport increases secondary aerosol formation, is not the primary reason why secondary aerosols show larger concentration differences between Lake and Nolake experiments compared to primary aerosols. The true explanation lies in Stage 2 mechanisms, specifically the lake-induced chemical regulation that we detail below.

Stage 2 - Differential behavior over the lake surface due to chemical regulation

Once air masses reach the lake surface, the behavior of primary and secondary aerosols diverges dramatically due to fundamental differences in their chemical properties and formation mechanisms. This is where the lake's influence becomes critically important and component-specific, and this is the primary focus of our manuscript. For primary $PM_{2.5}$ (BC, OC, OIN), which originates mainly from direct emissions with no chemical transformations, the lake primarily acts as a physical accumulation zone. The suppressed boundary layer height, weak turbulent mixing, and low dry deposition rates (detailed in Section 3.2.2) lead to enhanced concentrations over the lake surface. However, because primary aerosols undergo minimal chemical transformations, their accumulation is relatively straightforward and primarily driven by physical processes alone.

For secondary $PM_{2.5}$, however, the situation becomes far more complex. Beyond the same physical accumulation mechanisms that affect primary aerosols, secondary aerosols experience additional and profound lake-induced chemical regulation that fundamentally alters their formation, stability, and distribution. This dual effect of physical accumulation plus chemical regulation is the key reason why secondary $PM_{2.5}$ shows substantially larger concentration differences between Lake and Nolake experiments compared to primary $PM_{2.5}$. In other words, for primary aerosols the lake effect equals physical accumulation only, whereas for secondary aerosols the lake effect equals physical accumulation plus chemical regulation that suppresses decomposition and enhances stability.

The critical role of lake-induced chemical regulation

As comprehensively analyzed in Section 3.2.4 of our manuscript, the lake's large heat capacity powerfully regulates local temperature and humidity, creating unique thermodynamic conditions that profoundly influence reversible chemical equilibria governing secondary aerosol formation. This regulation process is highly component-specific. Ammonium nitrate (NH_4NO_3) formation exhibits strong thermodynamic reversibility ($NH_3 + HNO_3 \rightleftharpoons NH_4NO_3$), where decreased temperature or increased humidity promotes particulate NH_4NO_3 formation, while high temperature and low humidity cause decomposition back to gaseous precursors. The lake's regulation creates dramatically different chemical environments between the two experiments. In the Nolake experiment at 14:00 LT, elevated near-surface temperatures and lower humidity over land promote NH_4NO_3 decomposition, producing strong negative chemical contributions to $PM_{2.5}$ (exceeding -16 μg/m³) near the surface. The decomposed gaseous precursors are then transported by strong turbulent mixing to higher altitudes (0.5-1.5 km), where lower temperatures and higher humidity favor re-condensation, creating strong positive chemical contributions (exceeding 16 μg/m³) aloft. In contrast, in the Lake experiment, the lake surface maintains higher humidity and slower temperature increases, which enhance NH_4NO_3 stability and suppress decomposition. This creates three synergistic effects. First, minimal near-surface decomposition preserves particulate nitrate and ammonium. Second, reduced decomposition cuts off precursor transport to higher altitudes. Third, weak vertical mixing further minimizes chemical redistribution. The result is near-zero chemical contributions throughout the lake area from surface to upper atmosphere, but with substantially higher absolute concentrations of nitrate and ammonium passively accumulating at the surface. These concentrations reach or exceed urban levels (Figures S15 and S16). In contrast, sulfate shows much smaller concentration differences between experiments (Figure S17) because

SO_2 oxidation to sulfate is nearly irreversible and demonstrates greater atmospheric stability, making it relatively insensitive to the lake's thermodynamic regulation.

Summary

In summary, the substantially larger differences in secondary aerosol concentrations between Lake and Nolake experiments compared to primary aerosols are not primarily explained by extended atmospheric residence time during transport, as the reviewer suggested. Instead, these differences are driven by Stage 2 mechanisms, specifically the lake's powerful regulation of local temperature and humidity, which profoundly influences reversible chemical equilibria governing secondary aerosol formation and stability. This component-specific chemical regulation mechanism, detailed in Section 3.2.4, transforms the lake from a passive surface into an efficient "storage zone" specifically for reversibly-formed secondary aerosols, creating the anomalous spatial pattern where secondary $PM_{2.5}$ concentrations over the lake exceed urban levels. We thank the reviewer for raising this important point, which has allowed us to clarify that the differential response between primary and secondary aerosols is driven by chemical regulation mechanisms rather than atmospheric residence time during transport, reinforcing the novel contributions of our lake-induced chemical regulation framework.

Regarding the modifications to the original manuscript text, these have been described in detail in our previous **Response 16** and will not be repeated here.

- ***18. Section 3.1.2: Could the authors comment on why secondary aerosols primarily contribute to the differences during daytime, while primary aerosols dominate during nighttime?***

Response: We sincerely appreciate the reviewer's insightful question regarding the contrasting roles of primary and secondary aerosols in daytime versus nighttime concentration differences. This is indeed a critical aspect of our findings that warrants detailed explanation.

Daytime dominance of secondary aerosols:

During daytime, secondary aerosols (particularly nitrate and ammonium) primarily contribute to the concentration differences between Lake and Nolake experiments, especially over the lake region. This phenomenon is fundamentally driven by the lake's strong regulation of atmospheric thermodynamic conditions and chemical processes, as comprehensively explained in Section 3.2.4 in the manuscript. The key mechanism lies in the thermodynamic reversibility of ammonium nitrate formation ($NH_3 + HNO_3 \rightleftharpoons NH_4NO_3$), which is highly sensitive to temperature and humidity, two parameters that lakes powerfully regulate through their large heat capacity. In the Nolake experiment, elevated daytime temperatures and lower humidity promote NH_4NO_3 decomposition near the surface, producing strong negative $PM_{2.5}$ chemical contributions (exceeding -16 μg/m³, as shown in Figure S14b). The decomposed gaseous precursors are then transported by vigorous thermal turbulence to higher altitudes (0.5-1.5 km) where cooler temperatures favor re-condensation, creating strong positive contributions (exceeding +16 μg/m³). In stark contrast, the Lake experiment maintains higher humidity and slower temperature

increases due to the lake's large heat capacity and evaporative cooling effects, which significantly enhances NH_4NO_3 stability and suppresses decomposition throughout the atmospheric column. As illustrated in Figures S14b and S14d, this results in near-zero chemical contributions over the lake area from surface to upper atmosphere at 14:00 LT. The suppressed decomposition eliminates the source of gaseous precursors and, combined with weakened vertical mixing over the lake, allows secondary aerosols to passively accumulate over the lake surface without the decomposition losses experienced over farmland. Consequently, nitrate and ammonium concentrations over the lake reach or even exceed urban levels at 14:00 LT (Figures S15 and S16), representing maximum concentration differences between experiments.

Importantly, primary $PM_{2.5}$ (BC, OC, OIN), which originates mainly from direct emissions with minimal chemical transformations, shows much weaker sensitivity to the lake's thermodynamic regulation. Primary aerosols are transported to the lake and accumulate due to suppressed boundary layer development and weakened dry deposition, but they do not undergo the dramatic chemical transformation cycles that amplify secondary aerosol differences. Among secondary aerosols, sulfate shows much smaller concentration differences than nitrate and ammonium (Figure S17) because sulfate formation is essentially irreversible and demonstrates greater atmospheric stability. Consequently, the maximum concentration differences at 14:00 are dominated by secondary aerosols, particularly nitrate and ammonium. This transforms the lake from a passive surface type into an efficient secondary pollutant "storage zone" during daytime.

Nighttime dominance of primary aerosols:

During nighttime, the pattern reverses, primary aerosols dominate the concentration reduction in urban areas, while secondary aerosols play a relatively minor role. This difference stems from distinct dynamic and chemical mechanisms operating at night, as detailed in Section 3.2.3.

The nighttime concentration reduction in urban areas results primarily from lake-enhanced vertical mixing driven by land breeze circulation. The lake-land thermal contrast induces additional dynamic turbulence and upward motion above the city, disrupting typical nighttime stable conditions. Figure 11c demonstrates that vertical mixing intensity in urban areas is substantially greater in the Lake experiment, with stronger negative contributions near the surface (indicating more efficient upward pollutant removal) and stronger positive contributions aloft. Figure S11c further validates this mechanism, showing lower $PM_{2.5}$ concentrations near the urban surface but higher concentrations at 100-300 m altitude in the Lake experiment during nighttime. However, this enhanced vertical redistribution mechanism affects primary and secondary aerosols very differently.

Primary aerosols respond immediately and efficiently to the enhanced vertical mixing, being readily transported upward from the surface. Since primary aerosols are chemically inert at night, they respond proportionally to the enhanced vertical mixing without significant concurrent source or sink processes. As illustrated in Figure S13, primary $PM_{2.5}$ (including BC, OC, and OIN) shows substantial and consistent reductions near the urban surface due to this efficient upward transport. In contrast, secondary aerosols exhibit a more complex behavior during nighttime due to the superposition of dynamic transport and active

chemical production processes. Despite enhanced vertical mixing, nighttime urban areas continue to experience active secondary aerosol formation through ongoing chemical reactions. Figure S13 shows the vertical profile of chemical contributions for secondary particles during nighttime. While primary $PM_{2.5}$ shows straightforward concentration reductions (negative differences between Lake and Nolake experiments), secondary $PM_{2.5}$ exhibits much smaller net differences. This occurs because the reduction effect from enhanced vertical mixing is partially offset by concurrent chemical production. The net change in secondary $PM_{2.5}$ thus represents the balance between removal by vertical mixing (which decreases concentrations) and local chemical formation (which increases concentrations).

Consequently, the nighttime decrease in near-surface urban $PM_{2.5}$ concentrations predominantly results from the efficient upward transport of primary particles via lake-enhanced vertical mixing, while the reduction of secondary $PM_{2.5}$ is partially offset by simultaneous chemical production. This component-specific response fundamentally explains why primary aerosols dominate the nighttime concentration differences in urban areas.

We acknowledge that in the original Section 3.1.2, we did not explicitly indicate that the mechanisms underlying these day-night contrasts would be thoroughly explained in subsequent sections. Following the reviewer's valuable suggestion, we have now revised Section 3.1.2 to clearly state:

"This spatial pattern reflects a sequential transport process. Particulate matter is first transported from pollution source regions (such as urban areas) to the lake surface, which has no local emissions. Subsequently, due to the suppressed boundary layer height, weak boundary layer mixing, and low dry deposition rates over the lake, particles accumulate substantially on the lake surface (detailed mechanism explained in Section 3.2.2). Notably, secondary $PM_{2.5}$ concentrations over the lake are 15-18 μg/m³, while secondary $PM_{2.5}$ concentrations in urban areas in the Lake experiment are approximately 12-15 μg/m³, forming an anomalous pollution pattern where lake surface concentrations exceed urban concentrations, which differs dramatically from conventional understanding. Moreover, because the accumulation of secondary $PM_{2.5}$ over the lake during daytime is much greater than that of primary $PM_{2.5}$ (detailed mechanism explained in Section 3.2.4), a greater amount of secondary particles is subsequently transported back to urban and other surrounding areas through lake-breeze circulation and dispersion (detailed mechanism explained in Section 3.2.3), becoming a key factor in exacerbating urban daytime $PM_{2.5}$ pollution. "

Additionally, we have substantially enhanced Section 3.2.3 by adding explicit discussion of the differential responses of primary versus secondary aerosols to nighttime vertical mixing processes. We have updated Section 3.2.3 in the revised manuscript accordingly:

"Notably, this nighttime concentration reduction in urban areas is primarily attributable to the vertical mixing of primary $PM_{2.5}$ rather than secondary aerosols. As illustrated in Figure S13, nighttime urban areas continue to experience active secondary aerosol formation through chemical reactions, which increases secondary $PM_{2.5}$ concentrations and counteracts the reduction effect from vertical mixing. Consequently, the net decrease in near-surface urban $PM_{2.5}$ concentrations during nighttime predominantly results from the efficient upward transport of primary particles via lake-enhanced vertical mixing,

while the reduction of secondary $PM_{2.5}$ is substantially offset by concurrent chemical production."

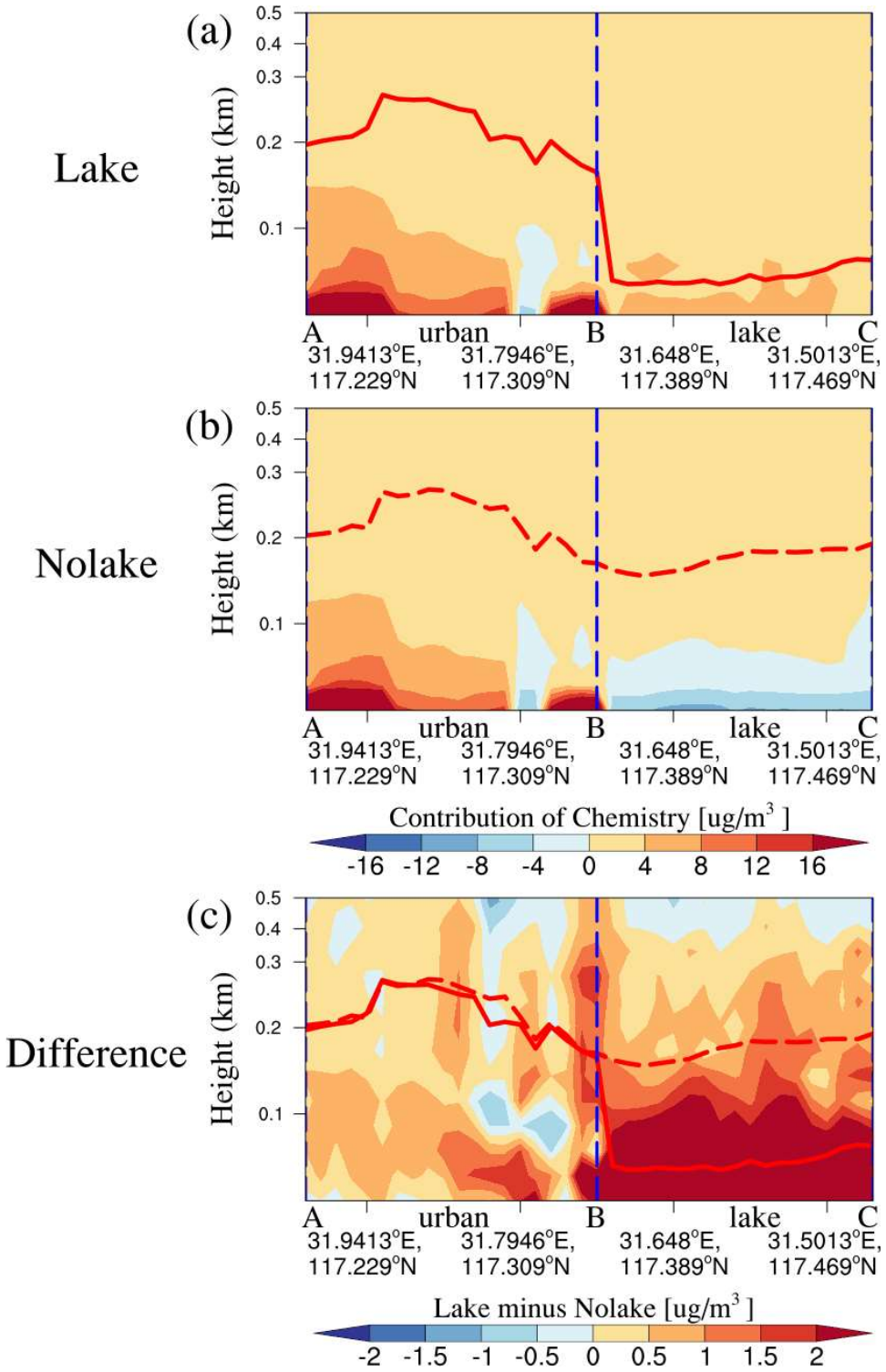

Figure S13. The vertical cross-section of chemical process contributions to $PM_{2.5}$ concentrations along the key path AC (indicated in Figure 2) for the (a) Lake experiment, (b) Nolake experiment, and (c) their differences (Lake minus Nolake) during nighttime, averaged over 10-20 March 2019. The shaded contours represent the contribution of chemical processes to surface $PM_{2.5}$ concentrations or their differences between the two experiments at each altitude. The red solid line represents the PBLH in the Lake experiment, and the red dashed line represents the PBLH in the Nolake experiment. The blue dashed line represents the lake-land boundary.

- ***19. Figure 7 caption: Replace "wind speed" with "wind vectors," as the figure shows both speed and direction. Also correct "thier" to "their." Are the wind fields and boundary layer height interpolated along the AC track? I suggest reducing the spatial density of the wind vectors for improved visualization. Also, I suggest changing their color to something brighter like cyan that would be easy to read over dark red color. Additionally, the resolution of the figure is low, making the labels difficult to read.***

Response: We thank the reviewer for the detailed suggestions and careful review. Regarding the caption of Figure 6, we have replaced "wind speed" with "wind vectors" to accurately reflect that the figure shows both wind speed and direction information. We have also corrected the spelling error from "thier" to "their." Regarding the reviewer's question about whether the wind fields and boundary layer height are interpolated along the AC track, yes, these variables are indeed interpolated along the AC cross-section to clearly display the spatial variations in the vertical profile. Regarding the suggestion to reduce the spatial density of wind vectors, we understand the reviewer's consideration for improved visualization. However, after careful consideration, we believe that reducing the wind vector density might result in the loss of important spatial details of the wind field, particularly in key regions such as the lake boundary and urban areas. These details are crucial for understanding the lake-induced circulation structure and pollutant transport processes. Therefore, we have maintained the original wind vector density. Regarding the suggestion to change the wind vector color, we greatly appreciate the reviewer's suggestion to use brighter colors such as cyan. During the revision process, we tested multiple color schemes, including cyan, white, and other bright colors. However, considering the complexity of the background colors in the figure (especially the dark red regions), we found that black wind vectors provide the best contrast and readability across all background colors, enabling readers to more clearly identify the wind field structure. Therefore, we have maintained the use of black wind vectors. Regarding the figure resolution issue, we completely agree with the reviewer's comment and have improved the resolution of all figures to ensure that labels and details are clearly readable. We have completed all the above-mentioned revisions in the revised manuscript.

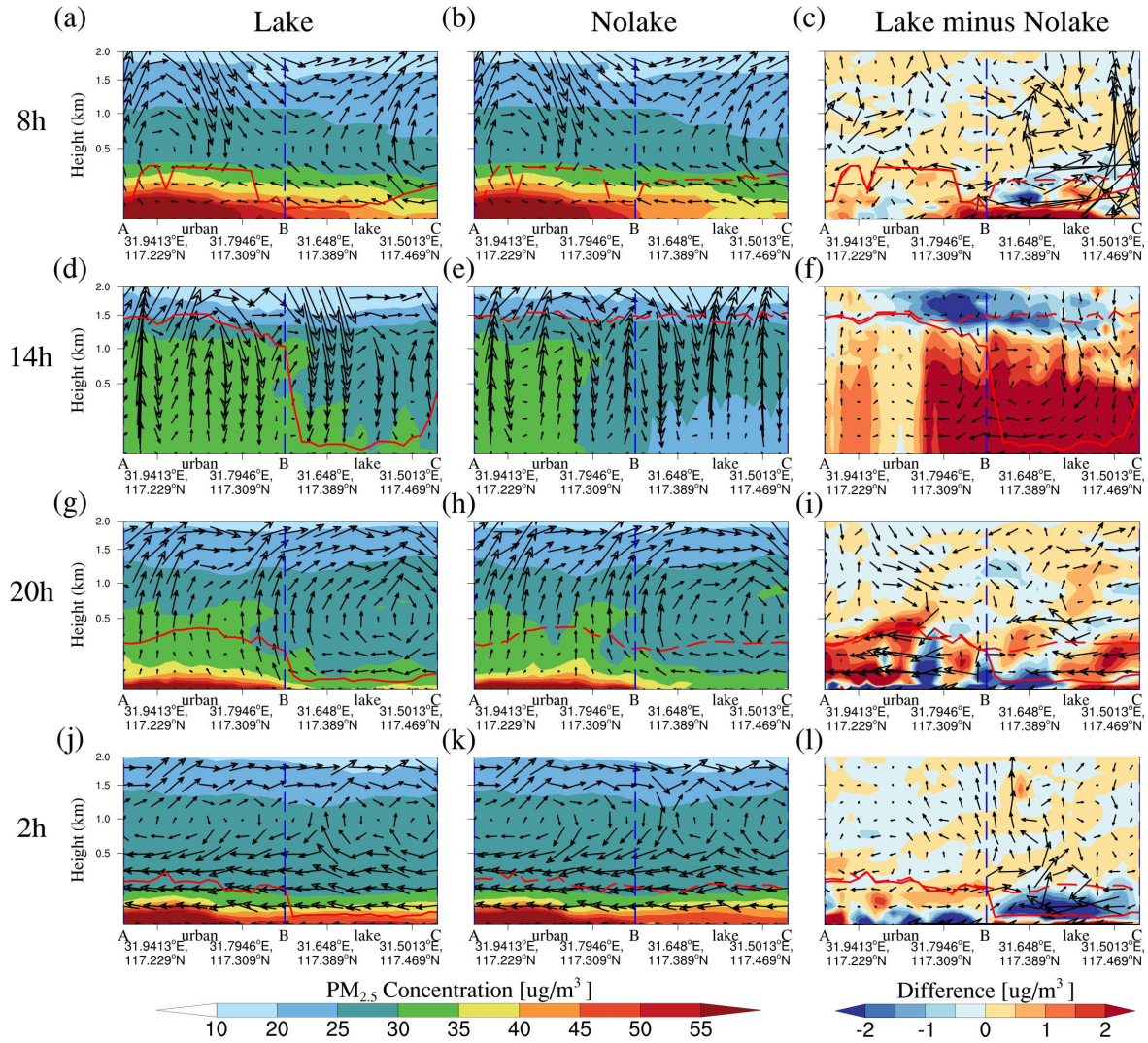

Figure 6. The vertical cross-section of $PM_{2.5}$ concentration and wind vectors along the key path AC (indicated in Figure 2) for the (a、d、g、j) Lake experiment, (b、e、h、k) Nolake experiment, and (c、f、i、l) their differences (Lake minus Nolake) at 08:00, 14:00, 20:00, and 02:00 LT, averaged over 10-20 March 2019. The shaded contours represent PM2.5 concentrations or their differences between the two experiments at each altitude. The black vector arrows indicate the superimposed vertical wind field (including horizontal and vertical wind components), with the vertical vector being multiplied by 50 for visibility. The red solid line represents the planetary boundary layer height (PBLH) in the Lake experiment, and the red dashed line represents the planetary boundary layer height in the Nolake experiment. The blue dashed line represents the lake-land boundary.

- ***20. Line 32: Replace "mechanisms which" with "mechanisms by which".***

Response: We thank the reviewer for this suggestion to improve the grammatical precision. We agree that "mechanisms by which" is more grammatically correct and clearer in expressing the causal relationship. Following the reviewer's recommendation, we have replaced "mechanisms which" with "mechanisms by which" in the revised manuscript. This revision enhances the grammatical accuracy and clarity of the sentence.

- ***21. Line 38: It would be clearer to use "column dry deposition velocities" instead of "dry deposition velocities".***

Response: We thank the reviewer for this suggestion. We appreciate the opportunity to clarify our terminology. In our study, "dry deposition velocities" specifically refers to the surface-level dry deposition velocities at the lowest model layer, not column-integrated values throughout the atmospheric column. We emphasize this distinction because dry deposition is fundamentally a surface boundary process that occurs exclusively at the interface between the atmosphere and the underlying surface, where pollutants are removed through direct contact. Unlike chemical processes or wet deposition that can occur throughout the atmospheric column, dry deposition only takes place at the near-surface layer (typically within the lowest few meters of the atmosphere) and is parameterized in atmospheric models as a lower boundary flux. Since our focus is on how lake surface characteristics (such as surface roughness, moisture availability, and thermal properties) influence this surface-level pollutant removal process, and given that dry deposition inherently occurs only at the surface rather than throughout the vertical column, we believe "dry deposition velocities" accurately describes the physical process we are analyzing. The term "column dry deposition velocities" might incorrectly imply vertical integration through multiple atmospheric layers, which would be inconsistent with the fundamental nature of the dry deposition process.