Response Letter

Dear Editors and Reviewers:

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

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 #1

General comments:

- ***1. I would like to thank the authors for preparing a compelling response to my initial review, in which they clearly demonstrate that my technical concern regarding the parametrization of vertical mixing strength only applies to gases, but not to aerosols. I believe that the modelling community will benefit from articles that point out these hidden model aspects as clearly as possible, which this article now does.***

Response: Thank you for your positive feedback and for acknowledging the improvements made in the manuscript. We are glad that our response effectively addressed your technical concern regarding the parametrization of vertical mixing strength and clarified that it applies to gases but not to aerosols. We also appreciate your recognition of the value of pointing out such model aspects clearly, and we are pleased that the revised version of the article meets that expectation.

- ***2. I have two more minor comments, and a few proposals for language improvements.***

Response: Thank you for your additional comments and suggestions. We appreciate your attention to detail and will carefully consider the minor comments and language improvement proposals. The necessary revisions will be made to enhance the clarity and quality of the manuscript.

Minor comments:

- ***1. I still wonder how the findings of this article should be interpreted in the context of Du et al. (2021). Figure 7 shows a range of nighttime mixing coefficients in the range of 0-3 m^2/s. As pointed out by the authors, the clipping of mixing strength does not apply to black carbon (BC), but Du et al. (2021) have nonetheless shown that perhaps it should, because their clipping at 5 m^2/s resulted in significantly more realistic surface BC. There is also no reason why trace gases should undergo a different mixing routine than aerosols. Based on these two arguments, it seems plausible that the exclusion of aerosols from the clipping procedure was an oversight. The authors need not concern with this question directly, but they should attempt to answer the following (perhaps in the discussion of Figure 7)::***

  Assuming it were appropriate to apply the clipping also to the mixing of aerosols, would this not potentially change the findings of this article? After all, the entire variance of the nighttime mixing values would vanish if the values were clipped to 5 m^2/s as suggested by Du et al. (2021). At daytime, where mixing coefficients are higher anyways, this logic would not apply.

Response: Thank you for your thoughtful comment and for bringing up the comparison with Du et al. (2021). We acknowledge the potential influence of clipping the mixing coefficients on the simulation results, especially with respect to the surface black carbon (BC) concentrations as demonstrated by Du et al. (2021). However, as we note in our response, the use of specific thresholds, such as the 5 m^2/s threshold proposed by Du et al., is primarily empirical in nature. These thresholds are designed to compensate for missing physical processes within the model, often by artificially enhancing mixing intensity. While such approaches may yield improvements in simulation results, they do not necessarily reflect the underlying physical mechanisms at play. Our study aims to understand the root causes of the underestimation of nighttime mixing intensity in the model, with a particular focus on the role of model resolution in turbulent mixing processes. Instead of relying on empirical thresholds to match the model output to observed data, we seek to address the fundamental physical processes that lead to discrepancies in mixing strength. We believe that threshold-based methods, while useful for aligning simulations with observations, do not provide a sufficient theoretical basis and may obscure a deeper understanding of the physical dynamics involved. Therefore, while we do not dispute the value of threshold adjustments in improving model performance, our approach deliberately avoids these artificial modifications in favor of investigating the underlying mechanisms that affect nighttime mixing intensity. In this context, we do not apply the clipping approach to aerosols in our analysis, as we believe it would detract from our goal of improving the model's representation of the physical processes driving turbulent mixing. We hope this clarifies our position and addresses the concern raised regarding the potential impact of applying clipping to aerosol mixing coefficients.

In the revised manuscript (Section 2.2.2), we have added these detailed explanation: "The publicly available version of WRF-Chem defines a default lower limit of 0.1 m^2/s for particulate matter mixing coefficients. We did not implement the adjustment proposed by Du et al. (2020), which suggest raising the lower limit of PBL mixing coefficient from 0.1 m^2/s to 5 m^2/s within the PBL. Although setting specific thresholds can improve simulation results, such thresholds are predominantly empirical in nature, whether based on CO and $PM_{2.5}$ emissions or the 5 m^2/s threshold suggested by Du et al. (2020). These threshold adjustments effectively compensate for missing physical processes in the model by artificially enhancing mixing intensity. Our approach focuses on understanding the physical mechanisms responsible for the model's underestimation of nighttime mixing intensity, with particular emphasis on how model resolution affects turbulent mixing processes. Rather than employing empirical thresholds to align model output with observations, we aim to investigate the fundamental causes of the discrepancies. We contend that threshold approaches rely heavily on empirical data, lack sufficient theoretical foundation, and may impede comprehensive understanding of the underlying physical processes. Consequently, this study utilizes the default particulate matter turbulent mixing coefficients in the model for our analyses."

In the revised manuscript (Section 3.2.2), we have also added another detailed explanation:

"We contend that threshold approaches are primarily based on empirical data and may impede comprehensive understanding of the underlying physical processes. In our study, particulate matter mixing coefficients are directly calculated through boundary layer parameterization without adjustments based on empirical settings."

- ***2. The authors discuss the correlation between surface types / land usage and mixing strength. In doing so, they mainly***

focus on how turbulence is affected by surface roughness. But there is another major mechanism to be discussed: The differences in radiative uptake among different surface types. Some surfaces absorb more of the actinic flux, and subsequently transfer this energy through sensible heat to the air above, which significantly contributes to the day-time convective boundary layer.

Response: Thank you for your detailed review and constructive comments on our manuscript. You highlighted an important aspect regarding the differences in radiative uptake among different surface types and their impact on the convective boundary layer formation, which we fully agree is a crucial mechanism. In response to your suggestion, we have expanded the discussion in Section 4 to include this important mechanism. The added text thoroughly addresses how different surface types affect the convective boundary layer (CBL) and turbulence mixing strength through variations in radiative flux absorption, reflection, and heat exchange. We discuss how urban areas with lower albedo absorb more shortwave radiation, leading to increased surface temperature and energy transfer to the atmosphere through sensible heat. In contrast, we explain how vegetated areas with higher albedo release more latent heat through transpiration while reducing sensible heat output. Furthermore, we elaborate on how the balance between sensible and latent heat fluxes across different surface types impacts turbulence intensity and CBL depth. For instance, we note that urban areas with stronger sensible heat flux generate more intense thermal convection, while vegetated areas dominated by latent heat flux may develop more stable atmospheric conditions. We conclude by emphasizing that future studies on land use impacts on turbulence mixing should consider not only surface roughness but also radiative flux differences, heat exchange mechanisms, and the comprehensive effects of surface albedo on turbulence development. In the revised manuscript (Section 4), we have added this detailed explanation: "Moreover, in addition to the influence of surface roughness on turbulence intensity, surface type significantly affects the CBL and turbulence mixing strength through differences in radiative flux absorption, reflection, and heat exchange. There are substantial variations in the absorption and reflection of shortwave radiation across different surface types. Urban areas typically have lower albedo, absorbing more shortwave radiation, which increases surface temperature and transfers energy to the atmosphere as sensible heat. In contrast, vegetated areas generally have higher albedo and, through transpiration, release more latent heat while reducing sensible heat output. These differences in energy exchange between surface and atmosphere directly influence turbulence strength. Furthermore, the varying balance between sensible and latent heat fluxes across different surface types impacts turbulence intensity and CBL depth. For instance, urban areas, with stronger sensible heat flux, tend to generate more intense thermal convection, often resulting in a shallower CBL, while vegetated areas, with predominant latent heat flux, may develop more stable atmospheric conditions, potentially leading to a deeper CBL with weaker turbulence. These mechanisms of radiative absorption and heat exchange are crucial in the formation of the diurnal CBL and determining turbulence intensity. Future studies on land use impacts on turbulence mixing should therefore consider not only surface roughness but also radiative flux differences, sensible and latent heat exchange mechanisms, and the comprehensive effects of surface albedo on turbulence development."

Comments on language / writing style:

- ***1. 101: change to "Previous research has indicated (...)". The use of the plural "researches" appears in other parts of the article as well and appears false to me.***

Response: Thank you for your valuable comment. I have carefully revised the manuscript as per your suggestion. Specifically, I have changed the plural form "researches" to the singular "research" throughout the manuscript, including the part you mentioned. I appreciate your attention to this detail.

- ***2. 101-106: This research summary is only helpful if the readers are made aware of the differences between the model code in the presented article and the referenced literature. For example, the study by Kuhlmann et al. (2003) is over 20 years old, and uses a different model at far lower resolutions. I would argue that such literature has limited relevance for the article's research question.***

Response: Thank you for your valuable comment. We understand your concern regarding the relevance of the literature referenced, particularly the study by Kuhlmann et al. (2003). As you pointed out, this study is over 20 years old and uses a different model at much lower resolutions, which may indeed limit its direct relevance to the research question presented in our article. In response to your feedback, we have removed references to outdated or less relevant studies, including Kuhlmann et al. (2003), to ensure that the literature cited is more aligned with the current modeling approaches, higher resolutions, and research objectives. We agree that it is crucial to reference studies that better match the scope and methods of our work, and we will now focus on more recent studies that are directly relevant to the model framework, resolution, and physical processes discussed in our paper. In the revised manuscript, we will explicitly clarify the differences between the model code used in this article and the referenced literature, especially in terms of model selection, resolution, and the physical processes involved, to avoid any potential confusion. We believe these revisions will enhance the relevance and scientific rigor of the paper.

- ***3. 163: Here and in other places, the term "multi-resolutions" is used, which I would recommend to replace by "multiple resolutions", "different resolutions", or "various resolutions".***

Response: Thank you for your suggestion regarding the use of the term "multi-resolutions." While we understand that terms like "multiple resolutions," "different resolutions," or "various resolutions" are commonly used, we have intentionally chosen the term "multi-resolutions" to more precisely convey the concept of utilizing several distinct resolution levels in our analysis. We believe this term captures the intended meaning more effectively in the context of our study. However, we appreciate your input and will consider your suggestion carefully as we finalize the manuscript.

- ***4. 177: The sentence with "contains some treatments" is not very informative. The authors could write instead: "WRF-Chem treats photochemistry of trace gases and aerosol-related processes with various different schemes (e.g. the Statewide Air Pollution Research Center (SAPRC99) photochemical mechanism and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC))."***

Response: Thank you for your constructive comment. Following your suggestion, we have revised the sentence to provide more specific and informative details. The revised sentence now reads: "WRF-Chem treats photochemistry of trace gases and aerosol-related processes with various different schemes (e.g., the Statewide Air Pollution Research Center (SAPRC99) photochemical mechanism and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC))." We believe this revision enhances the clarity of the manuscript.

- ***5. 190: remove "also".***

Response: Thank you for your suggestion. We have removed the word "also" as per your recommendation. We have updated Section 2.1.1 of the manuscript to include this modification:" The SAPRC99 photochemical mechanism (Carter, 2000) is chosen to simulate the gas-phase chemistry, and the MOSAIC is selected for aerosol processes (Zaveri and Peters, 1999; Zaveri et al., 2008). "

- ***6. 313: Split into two sentences: "This study primarily focuses on BC. The spatial distribution of BC emissions is shown in Figure 22."***

Response: Thank you for your suggestion. We have split the sentence into two as per your recommendation: "This study primarily focuses on BC. The spatial distribution of BC emissions is shown in Figure 2."

- ***7. 322: Is Zhang et al. (2021) a good reference to the MEGAN model? As far as I know, MEGAN is usually cited as: Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, https://doi.org/10.5194/acp-6-3181-2006, 2006.***

  This refers to the original version of MEGAN, and newer versions have been published. Regardless, Guenther et al. should be cited in the context of MEGAN.

Response: Thank you for your valuable feedback. We acknowledge the concern regarding the citation of Zhang et al. (2021) in relation to the MEGAN model. As you correctly pointed out, the appropriate reference for the MEGAN model is Guenther et al. (2006), which describes the original version of the model. We have revised the manuscript to include Guenther et al. (2006) as the primary reference for MEGAN and have adjusted the citation of Zhang et al. (2021) accordingly. Additionally, if newer versions of the MEGAN model are referenced in the manuscript, we will ensure that the relevant citations for those updates are properly included. In the revised manuscript (Section 4), we have added this cite: "Biogenic emissions were calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v3.0 model (Guenther, 2007; Zhang et al., 2021)."

Reference:

Guenther, A.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (vol 6, pg 3181, 2006), Atmospheric Chemistry and Physics, 7, 4327-4327, https://doi.org/10.5194/acp-7-4327-2007, 2007.

- ***8. 350: Change "derived" to "used".***

Response: Thank you for your suggestion. We have made the recommended change and revised the sentence to "We used the hourly BC observations from the air quality monitoring site on the campus of USTC during spring (March 10 to March 20, 2019)." We appreciate your input, which has improved the clarity of the text.

- ***9. 391: "We believe this approach (...)" should be rephrased to a more objective statement.***

Response: Thank you for your constructive feedback. We have revised the sentence as suggested, changing "We believe this approach (...)" to "This approach effectively captures daily variability patterns without losing essential detail." We also ensured that the revised text maintains clarity and objectivity. Your suggestion has improved the precision of the statement.

- ***10. 489: Change to "Figure S8b demonstrates (...)".***

Response: Thank you for your suggestion. We have made the recommended change and revised the sentence to "Figure S8b demonstrates (...)." Your input has helped improve the clarity and accuracy of the text.

- ***11. 647: It is unclear to me what "intension" means in this context.***

Response: Thank you for your suggestion. We have removed the term "intension," as it was unclear in this context, and revised the sentence to: "Figure S13 shows that the turbulent mixing coefficient parameterized at 5 km resolution is larger than that at 1 km resolution, which fails to explain the similar surface concentrations in these two higher-resolution (5 km and 1 km) simulations." We believe this revision clarifies the intended meaning.

- ***12. 671: Remove "vertically upward".***

Response: Thank you for your suggestion. We have removed the phrase "vertically upward" as per your recommendation. This revision has helped improve the clarity of the text.

- ***13. 863: Change to "dynamical framework of the model".***

Response: Thank you for your suggestion. We have made the recommended change and revised the text to "dynamical framework of the model." This adjustment has improved the accuracy and clarity of the statement.

- ***14. 865: Change to "The resolution of dynamic processes reduces (...)".***

Response: Thank you for your valuable feedback. We have revised the sentence as suggested, changing it to "The resolution of dynamic processes reduces reliance on traditional parameterization schemes, thereby decreasing the PBL mixing coefficient parameterized at finer resolutions." This modification has enhanced the clarity and precision of the statement.