

Dear reviewer,

Thank you for giving us the opportunity to revise our manuscript (Manuscript Number: egosphere-2023-1089). We appreciate your constructive comments and suggestions, which we have studied carefully while making appropriate revisions on the manuscript. We believe that under your guidance, our manuscript has been substantially improved.

In the following, the reviewer's comments/suggestions are highlighted by gray. The symbol ">>" quotes the original texts in the manuscript. Followed by the comments are our responses in plain text, as well as the respective revisions in the manuscript. Some important revisions are marked with red font.

Thank you again for your constructive comments and suggestions.

Yours sincerely,

Wen Lu, Bin Zhu, and all co-authors.

Replies to Reviewer

General Comments:

"Parameterized minimum eddy diffusivity in WRF-Chem(v3.9.1.1) for improving PM_{2.5} simulation in the stable boundary layer over eastern China" by Lu et al. proposed a parameterization formula for minimum turbulent diffusivity (Kzmin) and tested the simulations effects for PM_{2.5}. The results show that the revised Kzmin parameterization formula improved the PM_{2.5} simulation by improving turbulent diffusion under stable conditions. Weak turbulence in SBL is the key challenge in restricting progress of SBL theory and simulation, the topic of the manuscript is very important. However, the physical logic of the revised Kzmin parameterization formula is questionable, and numerical experiments need to be added. Therefore, I recommend major revision.

We feel great thanks for your professional review work on our article. As you are concerned, there are several problems that need to be addressed. According to your nice suggestions, we have made extensive corrections to our pervious draft, and the detailed corrections are listed below. We think your suggestions and comments have greatly improved our manuscript in science and technical details.

Specific comments:

1) Line 45, the SBL, weak turbulence and turbulence intermittency are hot topics in studies of atmospheric boundary layer with a lot of papers and progresses, I suggest the citations here keep up with the latest developments.

>>Line 45-46: Studies of the SBL remain insufficient; the SBL is often accompanied by intermittent turbulence and decoupling of the surface and free troposphere (Louis 1979; Grachev et al. 2005).

Thank you for this valuable suggestion. As you say, weak turbulence and turbulence intermittency have gotten a lot of attentions in recent years over the world. We have cited the latest research in the line 44-46 of the revised manuscript.

In the last of our revised manuscript in Line 44-46:

... While at present, the mesoscale meteorological numerical models cannot reasonably capture the weak turbulence and turbulence intermittency under SBL (Teixeira et al., 2008, Mahrt et al., 2020, Van der Linden et al., 2020, Jia et al., 2021, Allouche et al., 2022, Ren et al., 2023).

Reference:

- Allouche, M., Bou-Zeid, E., Ansorge, C., Katul, G. G., Chamecki, M., Acevedo, O., ... & Fuentes, J. D. (2022). The detection, genesis, and modeling of turbulence intermittency in the stable atmospheric surface layer. *Journal of the Atmospheric Sciences*, 79(4), 1171-1190.
- Jia, W., Zhang, X., Zhang, H., & Ren, Y. (2021). Application of turbulent diffusion term of aerosols in mesoscale model. *Geophysical Research Letters*, 48(11), e2021GL093199. In page2
- Mahrt, L., & Bou-Zeid, E. (2020). Non-stationary boundary layers. *Boundary-Layer Meteorology*, 177, 189-204.
- Ren, Y., Zhang, H., Zhang, L., & Liang, J. Quantitative description and characteristics of submeso motion and turbulence intermittency. *Quarterly Journal of the Royal Meteorological Society*.
- Teixeira, J., Stevens, B., Bretherton, C. S., Cederwall, R., Doyle, J. D., Golaz, J. C., ... & Soares, P. M. (2008). Parameterization of the atmospheric boundary layer: a view from just above the inversion. *Bulletin of the American Meteorological Society*, 89(4), 453-458.
- Van Der Linden, S. J., Van De Wiel, B. J., Petenko, I., Van Heerwaarden, C. C., Baas, P., & Jonker, H. J. (2020). A Businger mechanism for intermittent bursting in the stable boundary layer. *Journal of the Atmospheric Sciences*, 77(10), 3343-3360.

2) Line 49, "Huang et al. (2010)" did not show in Reference list.

>> ... Huang et al. (2010) improved the turbulent fluxes in the SBL by redefining the closure constants and modifying the sensible heat flux prognostic equation. ...

We apologize of the incorrect citation. The correct cited year of reference should be 2017. We have revised the text in the line 49-50 of the revised manuscript.

In the last of our revised manuscript in

Line 49-50:

... Huang et al. (2017) improved the turbulent fluxes in the SBL by redefining the closure constants and modifying the sensible heat flux prognostic equation. ...

Reference

Huang, Y., & Peng, X. (2017). Improvement of the Mellor–Yamada–Nakanishi–Niino planetary boundary-layer scheme based on observational data in China. *Boundary-Layer Meteorology*, 162(1), 171-188, 10.1007/s10546-016-0187-0.

3) Section 2.1, the basic information of the field experiment was missing. Readers cannot get anything about the field experiment in such simple description now. Which time periods of the first and second data sets used for model validation? All of this information should be added in detailed.

>> Three sets of data were used to evaluate model performance. The first set of data is the hourly ground-based observations of PM_{2.5} mass concentrations in 89 cities obtained from the China National Environmental Monitoring Center and published online (<http://106.37.208.233:20035>). The second set of data is the 3 h-hourly meteorological factors at 99 ground observation stations in eastern China. The meteorological factors contain 10 m wind speed, 10 m wind direction, and 2 m temperature. The third set of data is the vertical observations of PM_{2.5} and meteorological factors from field experiments by our group in Nanjing. The field experiment was carried out between 27 December 2016 and 31 December 2016 to obtain the 3 h-resolution vertical distribution data of PM_{2.5}.

Thank you for this valuable suggestion. We apologize that we miss enough information about our field experiment and data. The time periods of first and second data set that we used is both from December 1, 2016, to December 31, 2016. The third data set is the vertical observations of PM_{2.5} from field experiment. The observation site is located in the northern suburbs of Nanjing. The coordinates and altitude of the observation site are 32.0°N, 118.4°E and approximately 23 m asl, respectively. The PM_{2.5} concentrations were measured by a PDR-1500 fixed on an unmanned aerial vehicle (UAV) platform from December 27, 2016, to December 31, 2016. The details of field experiment please refer to the reference of Shi et al. (2021). 10 profiles (surface to ~1.0km) of PM_{2.5} in SBL were obtained in the field experiment for model evaluation in vertical. We have added the text in the line 75-80 of the revised manuscript.

In the last of our revised manuscript in

Line 75-80:

The time periods of first and second data set that we used is both from December 1, 2016, to December 31, 2016. The third set of data is the vertical observations of PM_{2.5} from field experiments. The observation site is located in the northern suburbs of Nanjing. The coordinates and altitude of the observation site are 32.0°N, 118.4°E and approximately 23 m asl, respectively. The PM_{2.5} concentrations were measured by a PDR-1500 fixed on an unmanned aerial vehicle (UAV) platform from December 27, 2016, to December 31, 2016. 10 profiles (surface to ~1.0km) of PM_{2.5} in SBL were obtained for model evaluation in vertical.

4) Section 2.3, how many haze cases did the numerical experiments choose? I did not see any introduction about the time periods or haze cases through the manuscript. Or only one case form 27 December 2016 to 31 December 2016? Can the reliability of the results be confirmed by more haze cases? The introductions on sensitivity experiments were also missing.

Thank you for this valuable suggestion. The observation of 8 cities was used to show the haze events in this study. There locations are Baoding (38.87°N, 115.48°E); Dezhou (37.45°N, 116.32°E), Jinan (36.61°N, 116.99°E), Zaozhuang (35.10°N, 117.45°E), Suqian (33.95°N, 118.29°E), Nanjing (32.0°N, 118.4°E); Liyang (31.4°N, 119.46°E), and Hangzhou (30.29°N, 120.16°E). Daily mean concentration statistics for each station was shown in Table R1. Three wider regional pollution events (the concentration of PM_{2.5} exceeds 115 $\mu\text{g}\cdot\text{m}^{-3}$ and lasts for more than 2 days) was occurred in December 2016. The time range of each pollution events are haze (December 1 to December 9), haze2 (December 16 to December 23), and haze3 (December 28 to December 31). We added the observed spatial and temporal variations of PM_{2.5} in Figure S3 of the revised supplement and line 97-100 of the revised manuscript. As for the reliability of the results about the haze cases, the time series of simulated and observed PM_{2.5} was compared in Figure S2 and the discussion is given in [Comment 10](#), and the new Kzmin has better performance in capturing the temporal evolution of the three haze events mentioned in Figure S3.

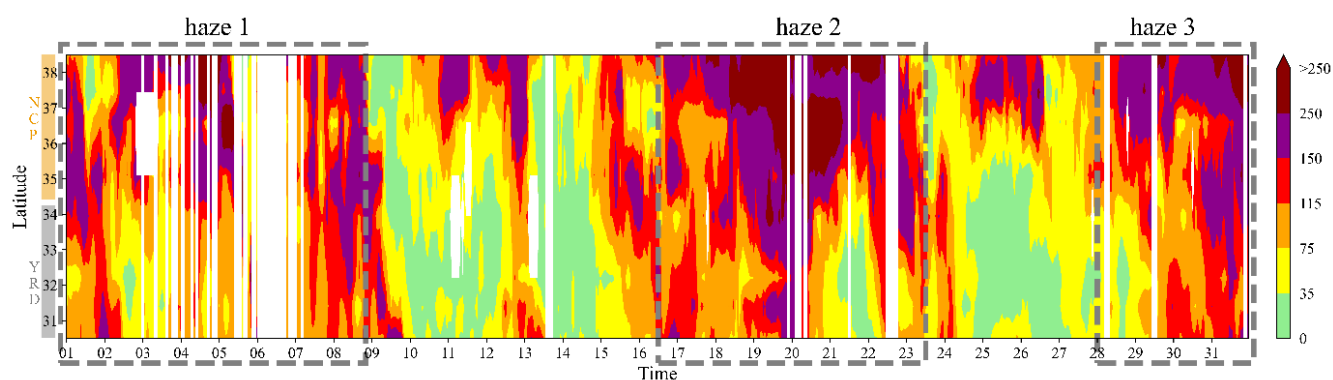


Figure S3. The observed spatial and temporal variations of PM_{2.5} mass concentration ($\mu\text{g}\cdot\text{m}^{-3}$) along the latitudes of the 8 observation sites from north to south. The grey boxes represent three haze events (concentration exceeds 115 $\mu\text{g}\cdot\text{m}^{-3}$ and lasts for more than 48 hours). The white color shaded is the missing data.

Table R1. Daily mean concentration statistics for each station

Standard	Baoding	Dezhou	Jinan	Zaozhuang	Suqian	Nanjing	Liyang	Hangzhou
>35 ug·m ⁻³	30	30	31	30	28	23	26	26
>75 ug·m ⁻³	27	28	22	22	18	17	18	19

As for the sensitivity experiments, the default Kzmin value in YSU scheme is 0.01 m²·s⁻¹. We set the fixed Kzmin value as 0.3, 0.5, 0.8, 1.0, 1.3, 1.5, 1.8, 2.0 in sensitivity experiments. The other setting is same as the description in Section 2.2. Then, we divided the sites in the simulation area into northern and southern sites for statistics, and the statistical results are shown in Table S1. The introductions on sensitivity experiments were added in the line 121-125 of the revised manuscript.

In the last of our revised manuscript

Line 97-100:

... The simulation started at 00:00 UTC on 28 November 2016 and ended at 00:00 UTC on 1 January 2017. To eliminate the effect of initial conditions, we set the first 3 d as the spin-up period (Napelenok et al., 2008). Three wider regional pollution events (the concentration of PM_{2.5} exceeds 115 ug·m⁻³ and lasts for more than 2 days) was occurred over eastern Chian in December 2016 (Figure S3).

Line 121-125:

... Therefore, we set the fixed Kzmin value as 0.3, 0.5, 0.8, 1.0, 1.3, 1.5, 1.8, 2.0 in sensitivity experiments (shown in Table S1, the available values are marked in red). We found that the reasonable Kzmin values for winter aerosol simulations have latitudinal difference in eastern China (the north of eastern China, NCP: 0.8 to 1.3 m²·s⁻¹, the Yangtze River Delta, YRD:1.0 to 1.5 m²·s⁻¹). ...

5) Line 105, some studies had revealed that the turbulent characteristics of PM_{2.5} are different with heat, this information should be clarified.

»Line 103-104: ... The turbulent mixing process of pollutants is considered to be similar to that of heat, which supposes the turbulent diffusion of particles and heat is identical.

Thank you for this valuable suggestion. It should be noted that here just states how the turbulent diffusion coefficients of PM_{2.5} in the WRF-Chem model are obtained. In recent, there have been observational study (Ren et al., 2021) showed that the relationship between turbulent difussion coefficient of PM_{2.5} (K_c) and turbulent difussion coefficient of heat (K_H) cannot be completely determined. In this study, we still treat particles as a scalar. To avoid ambiguity, we have revised the text in line 107-108 of revised manuscript.

In the last of our revised manuscript in

Line 107-108:

...The value of the diffusion coefficient of chemical compositionis is assumed to be equal to the value of the heat diffusion coefficient in Chem module (Jia et al, 2021b). ...

6) Line 111, () was missing in “Noh et al., 2003”.

»Line 111: ... According to the Prandtl number as in Noh et al., 2003:

Thank you for this valuable suggestion. We have revised the text in revised manuscript line 115.

In the last of our revised manuscript in

Line 115:

... According to the Prandtl number as in Noh et al. (2003):

7) The parameterization of the new value of K_{zmin} was proposed abruptly without sufficient physical discussion. Line 129, the authors described “We assume that the value of EF can be used to characterize K_{zmin} in different regions.” Why did you propose this assumption? In other words, what is the physical meaning behind the formula 5? Why is it set up in this form? Line 223, “After setting the adjustment factor value to 1.0 in formula 3”, why did you set an adjustment factor if you “assume that the value of EF can be used to characterize K_{zmin} in different regions”? and What is the basis for setting 1.0 as the adjustment factor? Very confusing, was it formula 5 or 3 in Line 223? Under stable conditions, because of the high value of EF, the values of K_{zmin} were at least 100 times larger than under unstable conditions, K_h might be close to 2, was it reasonable? I highly doubt the physical rationality. Based on your formula 5, K_h under stable conditions might larger than under unstable conditions. Anyway, the formula 5 you proposed was the core of this manuscript, more physical explanations are needed.

»Line 129: We assume that the value of EF can be used to characterize K_{zmin} in different regions.

»Line 223-225: ... As such, EF can reflect thermal flux features related to climate and the underlying surface in different regions. After setting the adjustment factor value to 1.0 in formula 3 ...

Thank you for your valuable suggestion. In the revised manuscript, we further analyze the relation between adjusted K_{zmin} (formula 5) and simulated $PM_{2.5}$ bias and try to give a reasonable physical proof.

Firstly, as shown in Figure 5, the new K_{zmin} scheme enhanced K_{zmin} values over eastern China, much larger than the default value of $0.01 \text{ m}^2 \cdot \text{s}^{-1}$. The distribution of the monthly averaged nocturnal K_{zmin} values exhibited a latitudinal difference with $0.88 \text{ m}^2 \cdot \text{s}^{-1}$ ($0.8\text{-}1.3 \text{ m}^2 \cdot \text{s}^{-1}$) in the NCP and $1.17 \text{ m}^2 \cdot \text{s}^{-1}$ ($1.0\text{-}1.5 \text{ m}^2 \cdot \text{s}^{-1}$) in the YRD, which is within the reasonable K_{zmin} ranges based on the sensitivity experiments in section 2.3 (Table S1). Also, in figure S6, the $PM_{2.5}$ bias shows a nonlinear positive correlation with the K_{zmin} in NCP and YRD. Most of k_{zmin} values (69%) in the NCP are less than 1, while most of K_{zmin} values (70%) in the YRD are greater than 1. The evidence indicated that the formula 5 is available to reflect the dependency of K_{zmin} on the landuse and the meteorology with a latitudinal effect. The latitudinal effect could relate to solar radiation, air temperature, cloud, precipitation and landuse in the 2 climate zones (Zhou et al., 2014, Jin et al, 2021) and in mostly extent can be expressed by 5. As such, the formula 5 can reasonable give the dynamic K_{zmin} values over east China in this study. We have added above sentences into the revise manuscript in line 231 to 236.

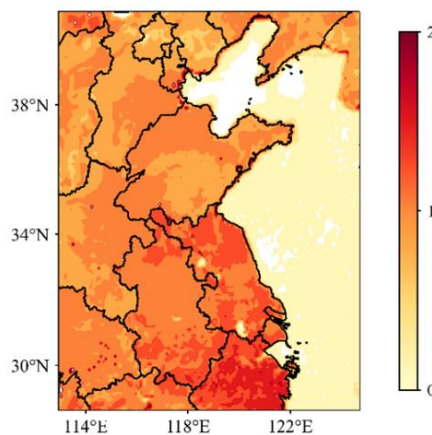


Figure 5. The distribution of K_{zmin} (unit: $\text{m}^2 \cdot \text{s}^{-1}$) in EXP_NEW.

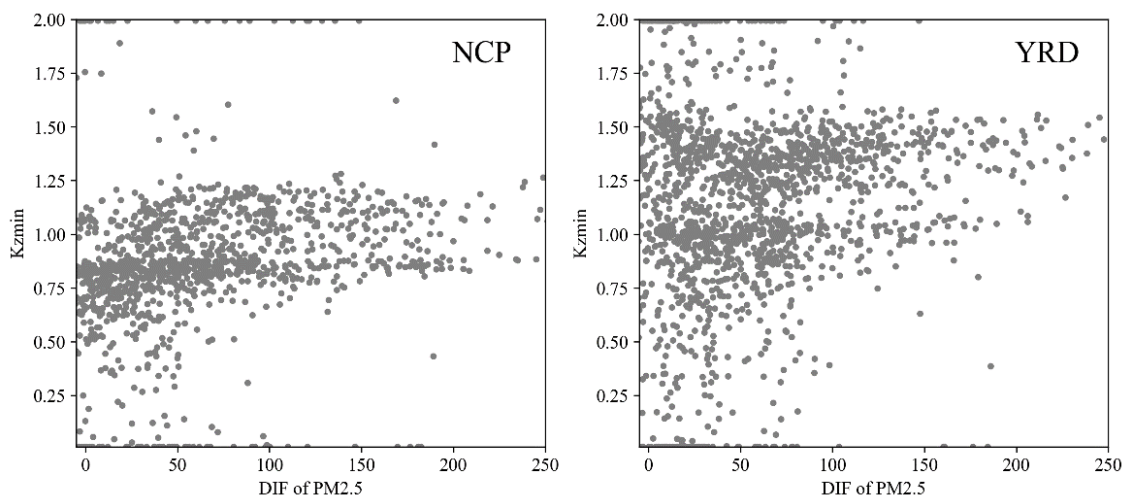


Figure S4. Scatter plots of the K_{zmin} and the bias of $PM_{2.5}$.

Secondly, The turbulent diffusion coefficient K_h was calculated following formula 3: $K_h = K_m/Pr + K_{zmin}$, where K_m is momentum mixing coefficient, Pr is the prandtl number.

The default value of the K_{zmin} in YSU PBL scheme is $0.01 \text{ m}^2 \cdot \text{s}^{-1}$. The aim of our work is to improve the simulation for the stable condition. Therefore, we only parameterized the K_{zmin} value during stable condition. While for the unstable, we let K_{zmin} consistent with the default settings ($0.01 \text{ m}^2 \cdot \text{s}^{-1}$). we find that the new adjusted K_h were $1.35 \text{ m}^2 \cdot \text{s}^{-1}$ in NCP and $2.03 \text{ m}^2 \cdot \text{s}^{-1}$ in YRD, which are much lower than the K_h in daytime. For example, the values of K_h in EXP_BASE and EXP_NEW in Nanjing ($32.0^\circ\text{N}, 118.4^\circ\text{E}$) was shown in Figure R1. There is no K_h value at stable condition larger than that in unstable condition. The simulation results of Du et al., (2020) and Jia et al., (2021) indicated that K_h great than 2.0 is a general value under SBL in eastern China.

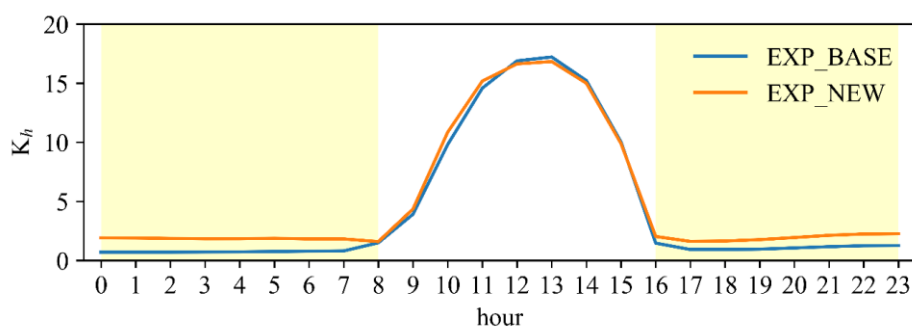


Figure R1. The turbulent diffusion coefficient in EXP_NEW and EXP_BASE (unit: $\text{m}^2 \cdot \text{s}^{-1}$) in Nanjing. The yellow shaded represent the stable condition (Richardson number > 0).

Reference

- Du, Q., Zhao, C., Zhang, M., Dong, X., Chen, Y., Liu, Z., ... & Miao, S. (2020). Modeling diurnal variation of surface $PM_{2.5}$ concentrations over East China with WRF-Chem: Impacts from boundary-layer mixing and anthropogenic emission. *Atmospheric Chemistry and Physics*, 20(5), 2839-2863, 10.5194/acp-20-2839-2020.
- Jia, W., & Zhang, X. (2021). Impact of modified turbulent diffusion of $PM_{2.5}$ aerosol in WRF-Chem simulations in eastern China. *Atmospheric Chemistry and Physics*, 21(22), 16827-16841, 10.5194/acp-21-16827-2021.
- Jin, H., Chen, X., Wu, P., Song, C., & Xia, W. (2021). Evaluation of spatial-temporal distribution of precipitation in mainland China by statistic and clustering methods. *Atmospheric Research*, 262, 105772.
- Zhou, L. T., & Huang, R. (2014). Regional differences in surface sensible and latent heat fluxes in China. *Theoretical and applied climatology*, 116, 625-637.

In the last of our revised manuscript

Line 231 to 236

... Also, in Figure S4, the $PM_{2.5}$ bias shows a nonlinear positive correlation with the $Kzmin$ calculated by fomular 5 in NCP and YRD. Most of $kzmin$ values (69%) in the NCP are less than 1, while most of $Kzmin$ values (70%) in the YRD are greater than 1. The evidence indicated that the formula 5 is available to reflect the dependency of $Kzmin$ on the landuse and the meteorology with a latitudinal effect. The latitudinal effect could relate to solar radiation, air temperature, cloud, precipitation and landuse in the 2 climate zones (Zhou et al., 2014, Jin et al., 2021) and in mostly extent can be expressed by fomular 5. As such, the formula 5 can reasonable give the dynamic $Kzmin$ values over east China in this study. ...

8) Line 134, “the value calculated by formula 1”, was it formula 5?

»Line 133-134: ... the $Kzmin$ value was set to the value calculated by formula 1. ...

We apologize for this mistake. We have revised the text in the line 138 of the revised manuscript.

In the last of our revised manuscript in

Line 138:

... the $Kzmin$ value was set to the value calculated by formula 5 ...

9) Line 140, the formulas of MB, IOA, RMSE, R, NMB and NME should be clarified at somewhere appropriate.

»Line 140-141: ... model performance metrics (MB, mean bias; IOA, index of agreement; RMSE, root mean square error; R: correlation coefficient, NMB: normalized mean bias, NME: normalized mean error) were used ...

The definition of the metrics that we used in our manuscript are added according to (revised supplement line 10-17)

In the last of our revised supplement

Line 10-17:

$$\text{Mean Bias: } MB = \frac{1}{N} \sum_{i=1}^N (M_i - O_i)$$

$$\text{Index Of Agreement: } IOA = 1 - \frac{\sum_{i=1}^N (M_i - O_i)^2}{\sum_{i=1}^N (|M_i - \bar{M}| + |O_i - \bar{O}|)^2}$$

$$\text{Root Mean Square Error: } RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2}$$

$$\text{Correlation Coefficient: } R = \frac{Cov(x,y)}{\sqrt{\bar{x}\bar{y}}}$$

$$\text{Normalized Mean Bias: } NMB = \frac{\sum_{i=1}^N (M_i - O_i)}{\sum_{i=1}^N O_i} \times 100\%$$

$$\text{Normalized Mean Error: } NME = \frac{\sum_{i=1}^N |M_i - O_i|}{\sum_{i=1}^N O_i} \times 100\%$$

10) The mean model performance in Table 1 and Table S1 means the mean performance from several cases or one case in whole domain? Another key issue is that I did not see any comparison of the simulated and observed $PM_{2.5}$ time series.

Thank you for this valuable suggestion. The mean model performance in Table1 and Table S1 is the mean performance in the whole month (December 2016). We added the time series comparison of observed and similated $PM_{2.5}$ into the supplement. The simulation of four citys (2 north city; 2 south city) was used to demonstrate the improvements. There locations are

Zhengzhou (34.75°N, 113.64°E); Jining (35.43°N, 116.63°E) in north of China and Hefei (31.94°N, 117.27°E); Nanjing (32.2°N, 118.8°E) in YRD. Three to four pollution events (the concentration of PM_{2.5} exceeds 115 ug·m⁻³ and lasts for more than 2 days, shadow in yellow) occurred in each city in December 2016. The overestimation of EXP_BASE is obvious, especially during stable condition, e.g., at night, and during heavy pollution events. The model result in EXP_NEW is improved by using our parameterized Kzmin and close to the observations. The metrics such as MB and IOA has significant improvement. It's worth noting that both two schemes underestimate extreme high peak concentrations of PM_{2.5} (such as Zhengzhou on 19 December, the concentration greater than 600 ug·m⁻³), which may be due to the poor ability to extremely heavy pollution simulation in existing mesoscale models. The hourly time-series was added in the Figure S2 of the revised supplement.

In the last of our revised supplement

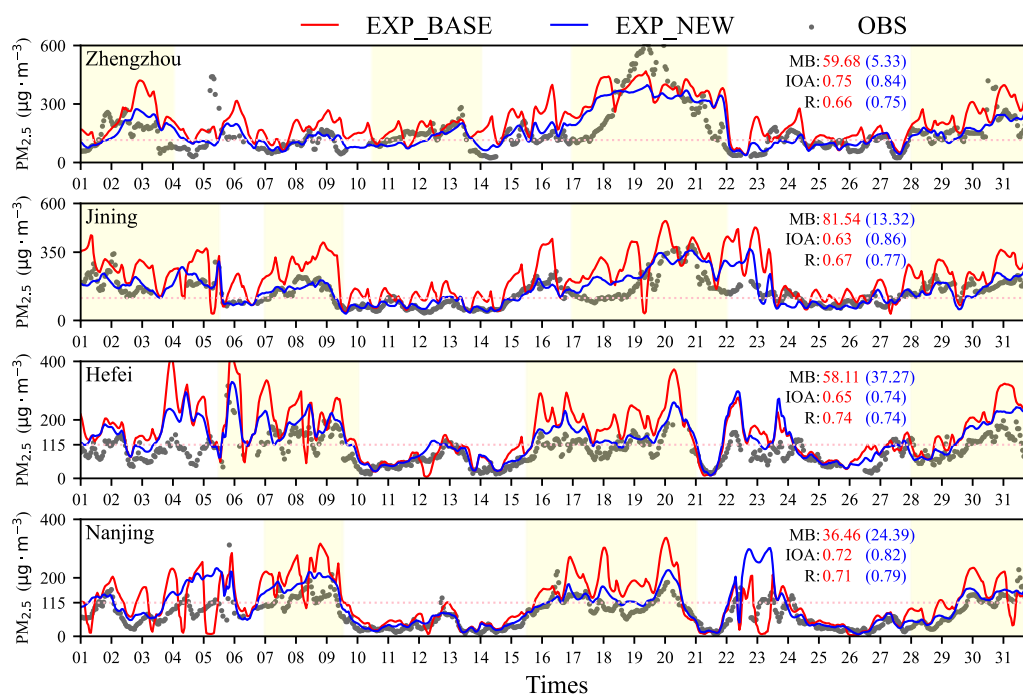


Figure S2. Time series of PM_{2.5} concentration in Zhengzhou, Jining, Hefei and Nanjing in December 2016. The grey dots, red lines and blue lines represent the results of observation, EXP_BASE and EXP_NEW, respectively. The yellow shaded represent haze events (the concentration of PM_{2.5} exceeds 115 ug·m⁻³ and lasts for more than 48 hours) in each city. The metrics (MB, IOA and R) was calculated by the full day (daytime and nighttime) data.

11) Line 238, “Figure 5d, 5e” means “Figure 6d, 6e”? same mistake in line 239, Figure 5 f.

>>Line 238: ... between EXP_BASE and EXP_NEW (Figure 5d, 5e). ...

>>Line 239: ... Figure 5f shows that the difference ...

We apologize for the mistakes. We have revised the text accordingly in the revised manuscript line 250, line 251.

In the last of our revised manuscript in

Line 250:

... between EXP_BASE and EXP_NEW (Figure 6d, 6e). ...

Line 251:

... Figure 6f shows that the difference ...

12) Line 265, “Figure 7. The distribution of difference ...”, you mean the PM_{2.5} concentrations difference?

»Line 265: Figure 7. The distribution of difference in difference process contribution.

We apologize for the unclear description in the figure caption for Figure 7. It is the distribution of process contribution differences of VMIX, ADV, AERO, and NET in model simulations. In section 3.3, we use process analysis to determine the key process of the improvement. For example, the vertical mixing (VMIX) value in EXP_BASE is represent the contribution of turbulent diffusion process to the change of PM_{2.5}. In Figure 6a, it is obvious that VMIX contribution on the surface is negative, which means turbulence diffusion process reduces the surface PM_{2.5} concentrations. The VMIX value of EXP_BASE minus the VMIX value of EXP_NEW is negative on the surface and positive on the upper BL. It indicates that EXP_NEW enhanced surface turbulence diffusion process compared to the EXP_BASE and diffused more PM_{2.5} into the high altitude and thus increasing the concentration of PM_{2.5} in the high altitude. We have revised the description between line 276-277 in the revised manuscript.

In the last of our revised manuscript in

Line 276-277:

Figure 7. The distribution of **PM_{2.5} process contribution** difference between EXP_NEW and EXP_BASE (unit: $\mu\text{g}\cdot\text{m}^{-3}\cdot\text{h}^{-1}$). a: VMIX, b: ADV, c: AERO d: NET=VMIX+ADV+AERO.

13) Writing needs to be further improved.

Thank you for this valuable suggestion. We made revisions to the paper and received help from the English rewriting Agency AJE. We hope that our revised paper will be approved by you!