
Response to Editor

The author sincerely thanks the editor for giving us an opportunity to revise the manuscript again, as well as the reviewers' professional evaluation and valuable suggestions. According to suggestions from reviewers, we have made extensive corrections to our previous manuscript, and the detailed point-by-point responses are listed below.

Response to Referee #1

RE: Impact of modified turbulent diffusion of PM_{2.5} aerosol in WRF-Chem simulations in Eastern China

Author(s): Wenxing Jia and Xiaoye Zhang

MS No.: acp-2021-435: MS type: Research article; Iteration: Revised submission

General Comments - Peter Taylor

Question1:

The authors have changed the title, as suggested, and made a series of changes but I still have concerns about some missing details. There are also sections, including the abstract, where some careful language edits could clarify the text.

Response1:

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 manuscript, and the detailed point-by-point responses are listed below. We have reorganized the abstract, please see [Lines 13-40](#) in the revised manuscript.

Question2:

The Richardson number dependent eddy diffusivity (Eq2, 5) for particles in stable conditions ($Ri > 0$) was developed and discussed in Jia et al (2021b). In that paper Ri is said to be a gradient Richardson number "where $f(Ri)$ is empirical stability function of gradient Richardson number (Ri). " but is not defined. The definition given in the present paper (Eq 1) is a bulk Richardson number based on differences relative to "surface level values". For winds are these 0 or U_{10} ? Tracking down Ri use by different authors is always a problem. Esau and Byrkjedal (2007) use finite difference approximations to the local gradient Ri , and discuss issues associated with their accuracy, " Another important aspect of the vertical resolution is a numerical

approximation of strongly-curved vertical profiles on coarse meshes using finite-difference numerical schemes." The current paper seems to use Esau and Byrkjedal's TDC functions for heat and momentum, but with a bulk Ri.

Response2:

We are very sorry that the description of data and methods in Section 2 are not clear enough, and even puzzles you. Actually, we have been using gradient Richardson number in previous and current papers, and the gradient Richardson number is approximated in finite difference form and the resulting is sometimes referred to as the bulk Richardson number (Garratt, 1992). For example, Louis et al. (1982) shows the Ri is the bulk Richardson number, but the expression is the form of gradient Richardson number (Eq .(5) in Louis et al., 1982). Moreover, our calculation method is consistent with that in the model. Equation (1) introduces the calculation method of PBL height, which leads to many doubts. Because this method has been widely used (Guo et al., 2016; Miao et al., 2018; Seidel et al., 2012; Zhang et al., 2013), it has been deleted to avoid confusion in the revised manuscript. In the revised manuscript, we will reorganize the content of Section 2 and add a flow chart (Figure 2) to show more details. In the original ACM2 scheme (in WRFv3.9.1), the Esau and Byrkjedal's TDC functions are used to calculate heat and momentum, and the Ri in the model is calculated as follows:

$$DZF = ZA(i, k + 1) - ZA(i, k) \quad (1)$$

$$SS = \frac{(US(i, k + 1) - US(i, k))^2 + (VS(i, k + 1) - VS(i, k))^2}{DZF^2} \quad (2)$$

$$GOTH = \frac{2G}{\theta_v(i, k + 1) + \theta_v(i, k)} \quad (3)$$

$$Ri = \frac{GOTH \cdot (\theta_v(i, k + 1) - \theta_v(i, k))}{DZF \cdot SS} \quad (4)$$

where Z_A is the height of each level in the model, which is related to the setting of the model. To resolve the PBL structure, 21 vertical layers were set below 2 km in this study (i.e., the specific setting of vertical levels is $\sigma = 1.000, 0.997, 0.994, 0.991, 0.988, 0.985, 0.980, 0.975, 0.970, 0.960, 0.950, 0.940, 0.930, 0.920, 0.910, 0.895, 0.880, 0.865, 0.850, 0.825, 0.800$). i and k represent grid points in horizontal and vertical direction respectively. US and VS are the component of wind. G is the gravity, θ_v is the virtual potential temperature. Then, substitute equations (1)-(3) into the equation (4) to calculate the Ri . Finally, Ri is used to calculate the turbulent diffusion coefficient.

We also need to note that the version of the model is very important, and Easun and Byrkijedal's TDC functions are updated only in the version after WRFv3.6.1. The improvement shows the importance of turbulent diffusion in the PBL scheme.

Please see [Section 2](#) for details in the revised manuscript.

Question3:

Fig 3 of Jia et al (2021b) shows the data used to develop Eq 5. There is a lot of scatter, near zero values of fc for several data points with $Ri < 0.2$. For $Ri > 0.2$ there are some large (~ 0.6) experimental fc values. Bottom line is that I would have very little confidence in the fc formulation proposed by Jia et al (2021b).

Response3:

We quite agree with you. We cannot rashly adopt a new functional form for fitting, because of the scarcity of data. However, these only data selected through field observation and strict quality control (Ren et al., 2020). Therefore, we still use a series of function forms proposed by predecessors for fitting, and finally select a better equation. For $Ri > \sim 0.2$, there are some large experimental fc values, there are four points that do not follow the tail behavior, and account for 8% of the number of scattered points in the figure. We check these abnormal points, finding that these points correspond to a larger turbulent flux. We have also speculated whether the turbulent diffusion of particles may be larger than what is fitted now. Although the current

function coefficient needs to be discussed, the ideas and techniques are worthy of deliberation. After more observation data are obtained, we will have a more accurate coefficient. The change of function coefficient will only make our future results better, and the continuous development and progress of the model also needs everyone's joint efforts to improve step by step. We are also doing our best to get more observation data (in progress) and further improve our fitting function.

Question4:

If implementation in WRF-CHEM as a tuning exercise produced well documented, convincing results that could certainly be of interest but I am not convinced by the material here. We are given information on the impacts of changes to the TDC for particles on model results. They do appear to improve overall comparisons with observed average values but I would like to see much more detailed discussion. One set of daily PBL height values (Fig 6) are given as a time series but it would be informative to see more sample time series data, of PM_{2.5} concentrations and fluxes at observation points, and with hourly resolution to see day-night differences. Start with some time series comparisons and then worry about the overall statistics. It is hard to take output from a large, complex meteorological model like WRF-CHEM in order to see the impact of tuning one of the internal equations but we should be shown more of the details.

Response4:

In fact, numerous scholars have been debugging a parameter in the mesoscale model for a long time, so as to change the simulation results of meteorological parameters and pollutants. For example, numerical weather prediction (NWP) usually adopts stability functions with so-called "enhanced mixing" to improve the unrealistic surface cooling (Bejaars and Holtslag, 1991; Derbyshire, 1999). In addition, the simulation of meteorological parameters and pollution concentration can be improved by specifying a minimum background turbulent diffusion under the stable conditions (Du et al., 2020; Savijärvi and Kauhanen, 2002). For the ACM2 scheme itself, with the continuous

updating of the version, the turbulent diffusion coefficient is gradually improved. Because the turbulent diffusion coefficient is extremely important, it controls the vertical mixing of momentum, heat, water vapor and pollutants within the PBL. Therefore, changes in turbulent diffusion coefficient can significantly affect the change of meteorological parameters and aerosol pollutant concentration.

We have described the improvement of the PBL scheme in further detail in the [Section 2](#) and [Figure 2](#) in the revised manuscript. We have made the comparison of pollutant concentration time series in the early stage. Since we need to simulate for a long time and compare many stations, finally, we show the monthly average results of regional distribution. Here we take three typical cities (i.e., Hefei, Nanjing and Shanghai) in Eastern China as an example to show the comparison of simulated and observed pollutant concentration (Fig. R1).

It can be seen from Figure R1 that the new scheme significantly improves the pollutant concentration is overestimated during heavy pollution episodes. At the same time, for the underestimated periods, the new scheme does not further worsen the underestimated situation. This is also an advantage of the new scheme. There must be some difference in the simulation results of individual cases, because of the difference of pollution cases. In short, the overall average results are excepted (Fig. 5).

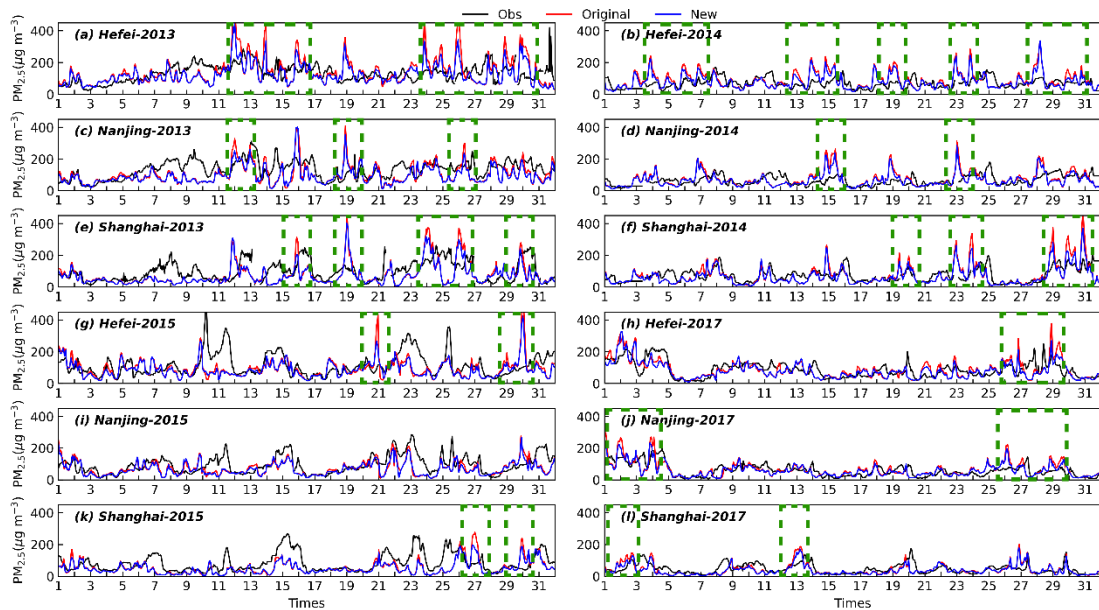


Figure R1. Time series of PM_{2.5} concentration in Hefei, Nanjing and Shanghai from 2013 to 2017. The black, red and blue lines represent the results of observation, original scheme and scheme, respectively. The green dashed box indicates the period when the new scheme has significantly improved.

Question5:

The confusion about exactly what is used as Ri is concerning. Does the model Ri correspond to the value used from the measurement data from Jia et al (2021b)?

Response5:

The Ri we used is definitely the same as Jia et al (2021), and we can provide time series of Ri (Fig. R2).

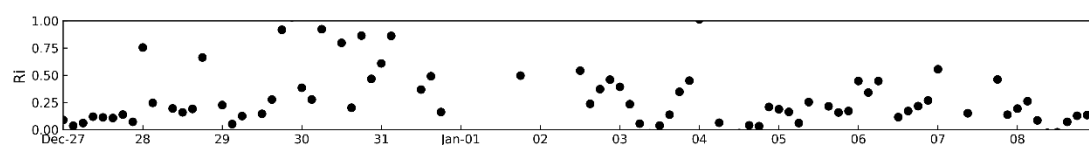


Figure R2. Time series of Richardson number from 27 December 2018 to 8 January 2019.

Question6:

Many of my concerns relate to that previous GRL paper but it would not be good for ACP to further encourage the use of a dubious result.

I am also concerned about the sources of PM_{2.5}. We are simply told to believe their inventory. Are we looking at PM_{2.5} from smoke stacks and subsequent chemical transformation, or is some of it road and other dust? Is the ground surface always a sink or can it be a source? How can we be sure that the differences between model prediction and observations are not related to the source inventory?

Response6:

We fully understand your concerns. In fact, the data used in this study have undergone strict quality control and many turbulence information characteristics have been

described in Ren et al (2020). Except that the sample size may be a little small. However, for the problem of sample size, we are also continuing to carry out field observation experiments, and we will obtain more data for our follow-up research in the future.

We very much agree with your question about the inventory. At present, the inventory we used is Multi-resolution Emission Inventory for China (MEIC) provided by Tsinghua University. The inventory published on the website has been updated to 2017 (<http://meicmodel.org/>), which is also the latest inventory published so far. Indeed, the accuracy of emission sources is very important for the simulation of pollutant concentration. In the WRF-Chem model, the processes of emission source, turbulent vertical mixing, dry deposition, advection transport and chemical conversion have a significant impact on the simulation of pollutants (Du et al., 2020; Chen et al., 2019; Gao et al., 2018). We also want to quantitatively prove the uncertainty of emission source, but this idea is difficult to realize at present. The focus of this study is to understand the impact of turbulent diffusion on pollutant concentration by adding turbulent diffusion of pollutants to the mesoscale model. It can be seen from the results of this paper that the simulated pollutant concentration is not completely consistent with the observation by improving the turbulent diffusion process. Therefore, the simulation results still have some room for improvement, and more efforts are needed in each process in the future.

Detailed comments

Question1:

line 105 Were these PM_{2.5} flux measurements compared with model predictions?

Response1:

These PM_{2.5} flux measurements are used to calculate the Richardson function of particles, i.e., $f_c(Ri)$.

Question2:

line 114 Good to calculate observed vertical fluxes, but are the results used or compared with the modelled fluxes. Is the ground a source or sink of PM_{2.5} in these simulations?

Response2:

Vertical flux is used for the calculation of the function of Richardson. In all simulation cases, PM_{2.5} may be both source and sink. We did not simulate a specific pollution process, but a long-time simulation. Therefore, the situation of each case is different. When there is a transport stage of a case, PM_{2.5} is a sink, while if it is a stable stratification with small wind, PM_{2.5} is more likely to be a source. If we want to know the source or sink of pollutants, we can only conduct simulation analysis for individual cases. In addition, the pollution process is complex. One pollution process is not only caused by source or sink, but is likely to have different results in different time periods (Zhong et al., 2018; 2019).

Question3:

line 123 Note that Eq(1) is a bulk Ri, and explain what are the surface level values, u_s , v_s , θ_s .

Response3:

We are sorry to bother you with this equation. This equation is a method for calculating the PBL height and has been widely used in models and observations (Guo et al., 2016; Miao et al., 2018; Seidel et al., 2012; Zhang et al., 2013). The surface level means the observation height is about 10 m (Miao et al., 2018). We have rewritten this section, please see [Section 2](#) in the revised manuscript.

Question4:

line 141 .. horizontal grid resolution.... and the factor 5 is rather larger than usually applied.

Response4:

Since we focus on a large area, the simulation time is relatively long, and used a two-way coupled meteorological-chemistry model, we do not use other horizontal grid resolutions. This ratio is more suitable for our simulation process. Of course, the ratio of 3 and 5 have been used by scholars, and the nesting of different ratio has little effect on the simulation results of the original scheme. Because other scholars can also find the pollutant concentration is overestimated in Eastern China (Du et al., 2020; Wang et al., 2021).

Question5:

line 147 ? Single layer UCM , and explain UCM. If measurement sites are urban then this could be an important issue.

Response5:

There are three urban surface physical schemes, namely, UCM, BEP and BEM.

UCM: Urban canopy model: 3-category UCM option with surface effects for roofs, walls, and streets.

BEP: Building Environment Parameterization: Multi-layer urban canopy model that allows for buildings higher than the lowest model levels.

BEM: Building Energy Model. Adds to BEP, building energy budget with heating and cooling systems.

The reason why we use UCM scheme here is that UCM can match any PBL schemes. However, BEP and BEM schemes can only match MYJ and BouLac PBL schemes. Therefore, we chose the UCM schemes.

Question6:

line 165 A 64 h spin up seems long for WRF (typically 12 h for meteorology) but may be needed for WRF-CHEM if initial concentrations are unknown, also why 91? $64 + 24 = 88$? Can you explain reasons for the long spin up time. Are results sensitive to this?

Response6:

Indeed, the spin-up time can be shorter in WRF, as you said, 12 hours. However, the variables have increased a lot in the WRF-Chem, and the simulation area is large. Therefore, we lengthened the spin-up time to obtain more stable calculation results. We are very sorry that we didn't clarify the simulation times.

*To reduce the systematic model errors, 91-hour simulation is conducted beginning from 0000UTC of three days ago for each day. The first 64-h of each simulation is considered as the spin-up period, the next 24-h is used for further analysis and the remaining 3-h is discarded (e.g., run one simulation from December 29, 0000 UTC (0800 BJT) to January 01, 1800 UTC (January 02, 0200 BJT), and in total 91 hours. We need the results from the January 01, 0000 BJT to 2300 BJT. From December 29, 0800 BJT to December 31, 2300 BJT is considered as the spin-up period (in total 64-h), and the results from January 02, 0000 BJT to 0200 BJT is discarded). This has no effect on the results. Please see **Lines 200-206** in the revised manuscript.*

The initial and boundary conditions of chemical fields were configured using the global model output of Model for Ozone and Related Chemical Tracers (MOZART), which has been widely used (<http://www/acom.ucar.edu/wrf-chem/mozart.shtml>).

For the initial and boundary conditions of the model, please see **Lines 183-189** in the revised manuscript.

Question7:

line 175 Is the 0.01 minimum value common to the original and new schemes?

Response7:

The minimum value is the same in the original scheme and the new scheme. Please see **Lines 223-224** and **Figure 2** in the revised manuscript.

Question8:

line 176 If one wants $u = 0$ at $z = 0$ then one can set $l = k(z+z_0)/(1 + kz/\lambda)$, where z_0 is a roughness length. There is no mention of roughness length until line 353, but it can

be an important parameter and should be different for momentum, heat and PM_{2.5}. It is present in WRF and should be discussed. It could be linked to the 0.01 minimum in Eq (2)?

Response8:

We agree with you that the roughness length is an important parameter and should be different for momentum, heat, moisture and PM_{2.5}. Moreover, the roughness length of momentum, heat and moisture do exist in the WRF model. We did not discuss this, here, mainly consider the following reasons. 1. The experiment station is in the southern suburbs of Dezhou city (37.15°N, 116.47°E), and flat farmland around this station (Fig. R3). The underlying surface is flat, and the roughness length is considered to be zero. 2. The roughness length is in the surface layer scheme in the WRF, not in the PBL scheme. In this study, we focus on the turbulent diffusion coefficient in the PBL scheme. Therefore, we default the roughness length to zero in the calculation of mixing length, which is consistent with pervious (Blackadar, 1962; Louis, 1979; Lin et al., 2008; Pleim, 2016). We have added this information in the revised manuscript (Lines 113-116, Figure S1). The setting of the minimum value assumes that the turbulence will not disappear completely in the model.

According to your opinion, it can be said that the roughness length is considered in the setting of the minimum value. This opinion inspired me, and it may be an extremely good innovation and we need to try in the further. Because we need to observe different underlying surface to obtain different roughness length, and use these observation data to do some interesting work combined with numerical simulation.

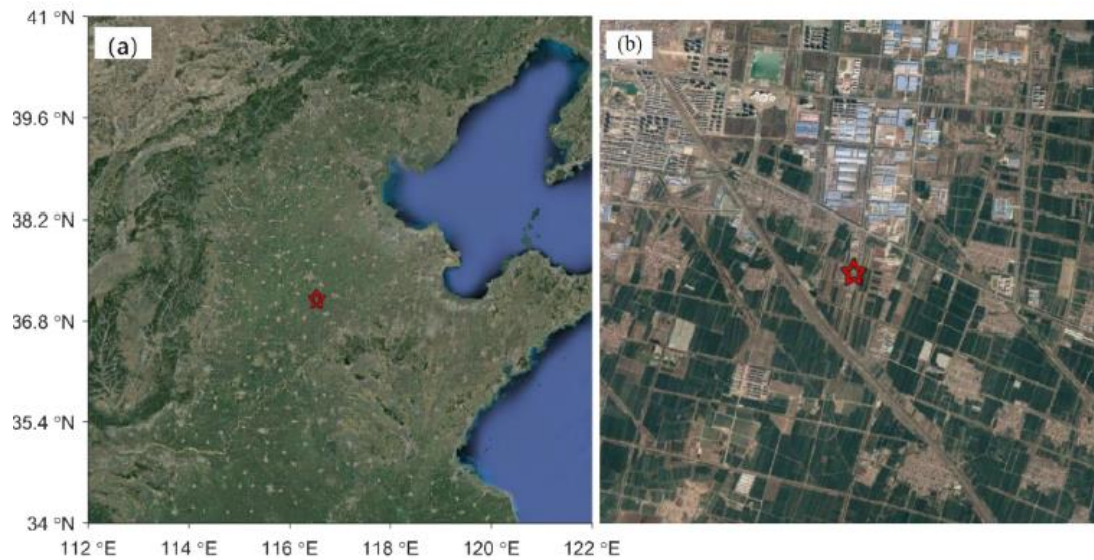
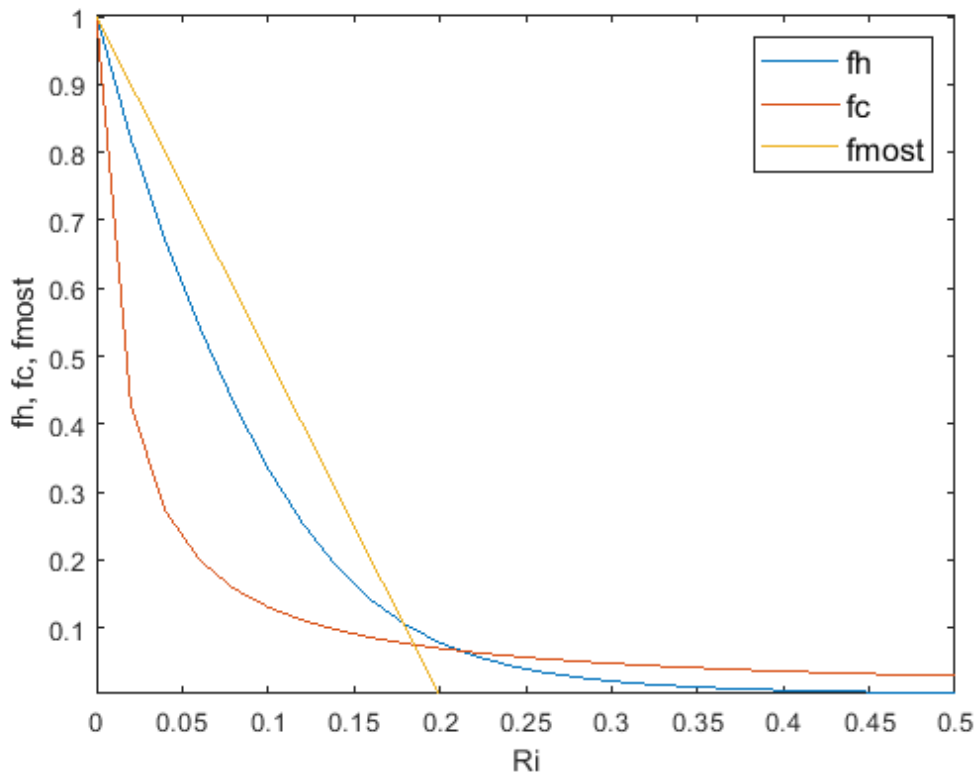


Figure R3. Google Earthmap of the Pingyuan observational site (marked as the red pentagram). The surrounding terrain (within a range of 5 km) is shown in (b) (adapted from Ren et al., 2020).

Question9:

line 186 It is worth comparing the current f_h and f_c , with the expression one would get using MOST, if we use the standard form $1/f_h = \phi_h = 1 + 5 z/L$, and relationship with Ri from Garratt (p52), and assuming $L = 0$ for $Ri > 0.2$. A quick plot is below with $f_{most} = 1/\phi_h$.



Response9:

Your suggestion is very good! Actually, the fitting function of particles (i.e., φ_c) has been obtained in Ren et al (2020). It is found that the turbulent diffusion of particles is greater than that of the original turbulent diffusion in Easter China (Figure 9), which should correspond to the cases where $Ri > \sim 0.2$. If we use the function you mentioned (i.e., f_{most}), there is no doubt that when $Ri > 0.2$, the turbulent diffusion coefficient is equal to the minimum value (i.e., 0.01). We will get the smaller turbulent diffusion than the original scheme, which will lead to worse simulation of pollutant concentration in Easter China. The function you mentioned is similar to the cut-off function in Jia et al (2021), and when Ri exceeds a certain critical value, the turbulent diffusion is the minimum value.

Question10:

For moderate values (<0.2) of Ri , MOST and f_h are both significantly larger than f_c so that the modified TDC is significantly reduced relative to MOST or earlier assumptions with $K_c = K_h$.

Response10:

Your understanding is absolutely correct. We found that the turbulent diffusion of particles is smaller than that of heat in the original scheme when $Ri < \sim 0.2$. And discuss and analyze what we have mentioned in the manuscript. Please see [Section 4.3](#) and [Figure 9](#) for details.

Question11:

For $Ri > 0.25$, $f_c > f_h$ as noted on line 188, but how often does this occur - in reality and in the model. In the model there may be confusion between gradient and bulk Ri . Can we see a pdf of Ri values?

Response11:

We have added the Ri in [Figure R2](#), and the [Section 2](#) has been modified accordingly in the revised manuscript. [Figure R2](#) is a time series of Ri during the observation period.

Question12:

line 199 "avoids the inapplicability of MOST". This "inapplicability" should be explained, maybe it is because we are not necessarily in a constant flux layer? MOST need not require $f_c = f_h$.

Response12:

We should describe it in more detail here, and we have reorganized the language of this part. Please see [Lines 253-259](#) in the revised manuscript.

Question13:

line 208 How is "night" defined in forming these averages?

Response13:

We take the time when the shortwave radiation is zero as the dividing line, and night is from 18:00 on the first day to 07:00 on the second day. This part has been supplemented in the revised manuscript. Please see [Lines 266-267](#) in the revised manuscript.

Question14:

line 223 Why the big difference in 2014? I am puzzled by why "relative bias" and "absolute bias" % differences are different. I assume that these are night-time values? Based on hourly data?

Response14:

Firstly, the pollution situation is different every year, so there must be differences in different cases, and the model cannot show a unified deviation in all cases. Secondly, we want to use a coordinate axis to explain the relative bias and absolute bias, which may be more helpful to your understanding. Figure R4 show the relative bias and absolute bias. In simple words, relative bias refers to the deviation between the original (or new) simulations and observations (i.e., the values of A or B), and the absolute bias refers to the absolute value difference between two relative biases (i.e., the value of C). And all simulated and observed pollutant concentrations are based on hourly data.

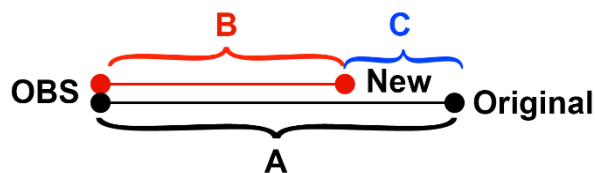


Figure R4. Schematic diagram of relative bias and absolute bias. OBS represents observation, Original and New indicate the simulation results of the original and new schemes, respectively. A and B represent the relative bias of the two schemes compared with the observation. C indicates absolute bias.

Question15:

From Fig 3 the relative and absolute bias values seem to be of opposite sign at some locations, A detailed definition of these quantities should be provided. I can guess but a few equations would help.

Response15:

The relative bias refers to the difference between the simulated value of the new scheme and observation in Fig. 4 (in the revised manuscript). The absolute bias decreases, indicating that the simulation results are improved. We believe that Figure R4 should be of great help to you.

Question16:

line 235 So n in Fig 4 is 31×24 ? I am not familiar with Taylor diagrams. Is the vertical axis from the model and the horizontal the observations?

Response16:

The number of points in Fig. 5 (in the revised manuscript) represents the observation stations used for comparison. The abscissa and ordinate represent the normalized standard deviation, and the arc represents the index of agreement. We can see that an “OBS” is marked on the abscissa. When the simulated statistical parameters can reach this point, indicating that the simulation is completely consistent with the observations.

Question17:

line 261 Can you say anything about deposition to the surface, which could be a critical removal process, or a source?

Response17:

The deposition process is very important for the evolution of pollutants, and we agree with you. We have supplemented the information about deposition in the model in [Section 2](#) in the revised manuscript, please see [Lines 212-219](#) in the revised manuscript. However, in the process of improving the model, we only changed the turbulent diffusion and did not change the dry deposition. Therefore, the change in Figure 6 is

only represents the change in pollutant concentration caused by the change in turbulent diffusion. We think it is best to use the process analysis method to quantify the contribution of each process, but this is a huge project, which is beyond the scope of this study. We can find a way to decompose each process in the model and then quantify the contribution of each process. We will do this work in the future, and we hope it can arouse your interest.

Question18:

line 287 There is a lot to be said about keeping models as simple as possible. What exactly do you have in mind as a " turbulence-aerosol two-way feedback module". But not needed here.

Response18:

Revised as suggested. Please see [Lines 350-354](#) in the revised manuscript.

Question19:

line 344 This seems to be the first mention of "source" and we are expected to accept that " there is no way to use other more elaborate inventories to quantify the uncertainty caused by emissions".

Response19:

In Section 4, even if we cannot quantify the uncertainty caused by emission sources, we should also mention it. The emission sources process is very key to the simulation of pollutants. We are unable to make further analysis, due to the limited of the current inventory. Once we have these resources, we will further improve the current work. In fact, there are many processes affecting the evolution of pollutants, and we only make an in-depth analysis of one of the very important processes, hoping to contribute to the development of the model.

Question20:

line 395 Maybe tell us what the mean absolute errors in hourly or daily PM_{2.5} values ($\mu\text{g m}^{-3}$) are in order to see how significant these bias improvements are.

Response20:

Here we express the regional average of absolute bias, which is shown in Fig. 4i-l.

Question21:

line 398 " Therefore, the pollutant concentration is reduced near the surface and better mixed in the whole layer, increasing the pollutant concentration in the upper level." Are there any upper level measurement to validate this effect?

Response21:

What we actually want to express here is that when the pollutant concentration is decreased near the surface, where does the pollutant go. We want to explain this phenomenon through models. However, we do not have vertical observations of particles for verification during all simulation periods. In recent years, we will have some vertical observation data of particles, but the latest emission source inventory has not been published, so we cannot simulate more periods. We will continue to do it in this direction and supplement our work step by step. After reviewing your professional advice, we found that there are still many ideas to be done, but not all problems can be solved at one time, we will continue to improve in the future.

Reference.

Garratt, J.R., (1992) The atmospheric boundary layer, Cambridge, UK

References

Beljaars, A. C. M., and Holtslag, A. A. M.: Flux parameterization over land surfaces for atmospheric models. *Journal of Applied Meteorology*, 30(3), 327–341. [https://doi.org/10.1175/1520-0450\(1991\)030<0327:FPOLSF>2.0.CO;2](https://doi.org/10.1175/1520-0450(1991)030<0327:FPOLSF>2.0.CO;2), 1991.

-
- Blackadar, A. K.: The vertical distribution of wind and turbulent exchange in a neutral atmosphere, *J. Geophys. Res.*, *67*, 3095–3102, <https://doi.org/10.1029/JZ067i008p03095>, 1962.
- Chen, L., Zhu, J., Liao, H., Gao, Y., Qiu, Y., Zhang, M., Liu, Z., Li, N., and Wang, Y.: Assessing the formation and evolution mechanisms of severe haze pollution in the Beijing–Tianjin–Hebei region using process analysis, *Atmos. Chem. Phys.*, *19*, 10845–10864, <https://doi.org/10.5194/acp-19-10845-2019>, 2019.
- Derbyshire, S. H.: Boundary-layer decoupling over cold surfaces as a physical boundary-instability. *Boundary-Layer Meteorology*, *90*, 297–325. <https://doi.org/10.1023/A:1001710014316>, 1999.
- Du, Q., Zhao, C., Zhang, M., Dong, X., Cheng, Y., Liu, Z., Hu, Z., Zhang, Q., Li, Y., Yuan, R., and Miao, S.: Modeling diurnal variation of surface PM_{2.5} concentrations over East China with WRF-Chem: impacts from boundary-layer mixing and anthropogenic emission, *Atmos. Chem. Phys.*, *20*, 2839–2863, <https://doi.org/10.5194/acp-20-2839-2020>, 2020.
- Gao, J., Zhu, B., Xiao, H., Kang, H., Pan, C., and Wang, D., and Wang, H.: Effects of black carbon and boundary layer interaction on surface ozone in Nanjing, China, *Atmos. Chem. Phys.*, *18*, 7081–7094, <https://doi.org/10.5194/acp-18-7081-2018>, 2018.
- Garratt, J.: *The atmospheric boundary layer*, Cambridge, UK, 37, 1992.
- Guo, J., Miao, Y., Zhang, Y., Liu, H., Li, Z., Zhang, W., He, J., Lou, M., Yan, Y., Bian, L., and Zhai, P.: The climatology of planetary boundary layer height in China derived from radiosonde and reanalysis data, *Atmos. Chem. Phys.* *16*, 13309–13319. <https://doi.org/10.5194/acp-16-13309-2016>, 2016.
- Jia, W., Zhang, X., Zhang, H., and Ren, Y.: Application of turbulent diffusion term of aerosols in mesoscale model, *Geophys. Res. Lett.*, *48*, e2021GL093199, <https://doi.org/10.1029/2021GL093199>, 2021.
- Lin, J., Youn, D., Liang, X., and Wuebbles, D. J.: Global model simulation of summertime U.S. ozone diurnal cycle and its sensitivity to PBL mixing, spatial resolution, and emissions, *Atmos. Environ.*, *42*, 8470–8483, <https://doi.org/10.1016/j.atmosenv.2008.08.012>, 2008.

-
- Louis, J.: A parametric model of vertical eddy fluxes in the atmosphere, *Bound.-Lay. Meteorol.*, 17, 187–202, <https://doi.org/10.1007/BF00117978>, 1979.
- Louis, J., Tiedtke, M., and Geleyn, J.: A short history of the PBL parameterization at ECMWF, *ECMWF*, 59–79, <https://www.ecmwf.int/node/10845>, 1982.
- Miao, Y., Liu, S., Guo, J., Huang, S., Yan, Y., and Lou, M.: Unraveling the relationships between boundary layer height and PM_{2.5} pollution in China based on four-year radiosonde measurements, *Environ. Pollut.*, 243, 1186–1195, <https://doi.org/10.1016/j.envpol.2018.09.070>, 2018.
- Pleim, J. E., Gilliam, R., Appel, W., and Ran, L.: Recent advances in modeling of the atmospheric boundary layer and land surface in the coupled WRF-CMAQ model, *Air Pollution Modeling and its Application XXIV*, 391–396, https://doi.org/10.1007/978-3-319-24478-5_64, 2016.
- Ren, Y., Zhang, H., Wei, W., Cai, X., and Song, Y.: Determining the fluctuation of PM_{2.5} mass concentration and its applicability to Monin–Obukhov similarity, *Sci. Total Environ.*, 710, 136398, <https://doi.org/10.1016/j.scitotenv.2019.136398>, 2020.
- Savijärvi, H., and Kauhanen, J.: High resolution numerical simulations of temporal and vertical variability in the stable wintertime boreal boundary layer: a case study, *Theor. Appl. Climatol.*, 70, 97–103, <https://doi.org/10.1007/s007040170008>, 2002.
- Seidel, D.J., Zhang, Y., Beljaars, A., Golaz, J.C., Jacobson, A.R., and Medeiros, B.: Climatology of the planetary boundary layer over the continental United States and Europe, *J. Geophys. Res. Atmos.* 117, D17106. <https://doi.org/10.1029/2012JD018143>, 2012.
- Wang, A., Li, Y., Zhao, C., Du, Q., Wang, X., and Gao, Z.: Influence of different boundary layer schemes on PM_{2.5} concentration simulation in Nanjing, *China Environmental Sciences*, 41, 2977–2992, <https://doi.org/10.19674/j.cnki.issn1000-6923.2021.0301>, 2021 (in Chinese).
- Zhang, Y., Seidel, D.J., and Zhang, S.: Trends in planetary boundary layer height over Europe, *J. Clim.* 26, 10071e10076. <https://doi.org/10.1175/JCLI-D-13-00108.1>, 2013.

Zhong, J., Zhang, X., Dong, Y., Wang, Y., Liu, C., Wang, J., Zhang, Y., and Che, H.: Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of PM_{2.5} during winter heavy pollution episodes in Beijing from 2013 to 2016, *Atmospheric Chemistry and Physics*, 18, 247–258. <https://doi.org/10.5194/acp-18-247-2018>, 2018.

Zhong, J., Zhang, X., Wang, Y., Wang, J., Shen, X., Zhang, H., Wang, T., Xie, Z., Liu, C., Zhang, H., Zhao, T., Sun, J., Fan, S., Gao, Z., Liu, Y., Wang, L.: The two-way feedback mechanism between unfavorable meteorological conditions and cumulative aerosol pollution in various haze regions of China. *Atmospheric Chemistry and Physics*, 19, 3287–3306. <https://doi.org/10.5194/acp-19-3287-2019>, 2019.

Response to Referee #2

General Comments:

I fell that my major concerns are appropriately addressed in the revised version. But I think the manuscript can be further improved. So my recommendation is publication in ACP after minor revisions.

Response:

Thank you again for your positive comments and valuable suggestions to improve the quality of our manuscript. Based on these comments and suggestions, we have made careful modifications to our pervious manuscript, and the detailed point-by-point responses are listed below.

Specific Comments:

1) For the value of TDC, the authors state in line 186-188 that “When R_i is greater than ~ 0.2 , the TDC of particles is greater than that of heat, which may reduce pollutant concentration”. This statement actually tell us that the TDC of particles is greater than that of heat when R_i is greater than ~ 0.2 . However, the TDC of particles is smaller than that of heat when R_i is smaller than ~ 0.2 . Therefore the result that the new scheme of TDC for particles can reduce the overestimated PM_{2.5} concentration implies that R_i is greater than ~ 0.2 in the most part of nighttime. So I suggest that the authors should add a paragraph to discuss this issue (it’s better to give the information about the statistics of the value of R_i), which will help the readers to understand the results more easily.

Response:

We agree with your understanding. According to the fitting results of our previous observation data, the TDC of particles is greater than that of heat in the original scheme when R_i is greater than ~ 0.2 . Although the R_i is not an input/output quantity in the model, the change in TDC is considered to be caused by the modification of $f(R_i)$ when other physical quantities remain unchanged.

According to your suggestion, we have added discussion in the revised manuscript (Lines 390-393 and Lines 407-409).

Once again, we have substantially revised the content of data and methods (i.e., including information about R_i), and provide a flow chart (Figure 2), so that readers can understand our results more clearly. Please see Section 2 in the revised manuscript.

Technical Comments:

1) Line 287: What is “turbulence barrier effect”? Please give the explanation.

Response:

Revised as suggested, we have added this concept to the revised draft (Lines 59-61).

“It means turbulence may disappear at certain heights forming a laminar flow as if there is a barrier layer hindering the transmission up and down during the heavy pollution episodes”. This phenomenon as the turbulent barrier effect, for detailed discussion, please refer to Ren et al. (2021).

2) Line 288: What is “HPEs”? Please give the specification.

Response:

Revised as suggested (Lines 354).

HPEs refers to “Heavy Pollution episodes”.

3) Fig. 5: Does each panel show the result for one day, or monthly mean? Does each panel show the result for multi-point mean, or area mean? Please add the specifications in the figure caption.

Response:

Revised as suggested (Fig. 6).

Figure 6 shows the situation in Hefei in the previous manuscript (Fig. R1). To be consistent with the statement of PBL height, we still selected Anqing (Fig. R2) and Fuyang (Fig. R3) stations. In addition, we selected the results of three typical cities (i.e.,

Hefei, Nanjing (Fig. R4) and Shanghai (Fig. R5)) in Eastern China as auxiliary verification, and added these results to the supplementary materials.

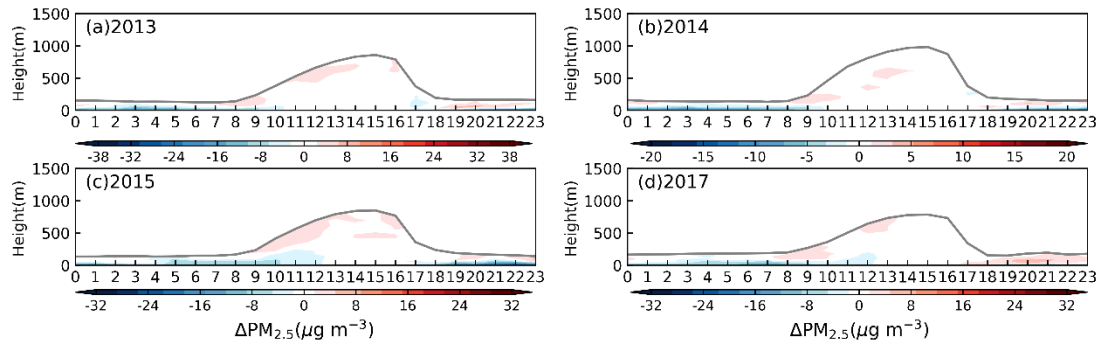


Figure R1. Time-height cross sections for the difference of $\text{PM}_{2.5}$ concentration between original and new schemes (i.e., the new scheme minus the original scheme) within the PBL in Hefei from 2013 to 2017. The gray line indicates the PBLH.

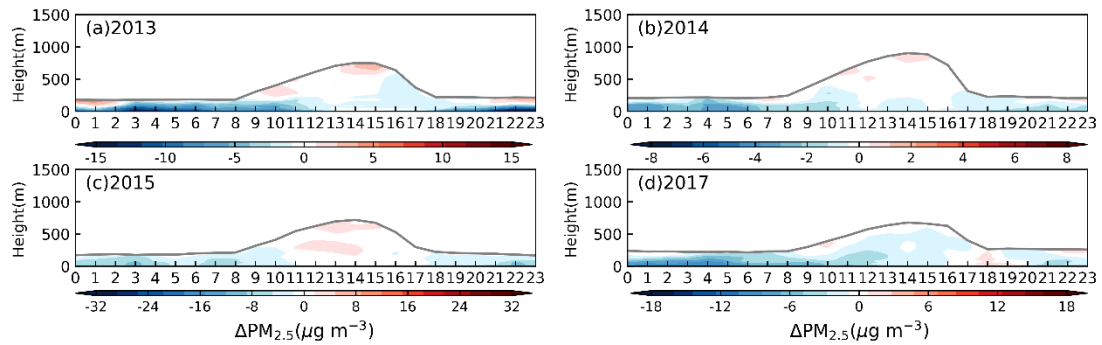


Figure R2. Similar to Figure R1, but in Anqing.

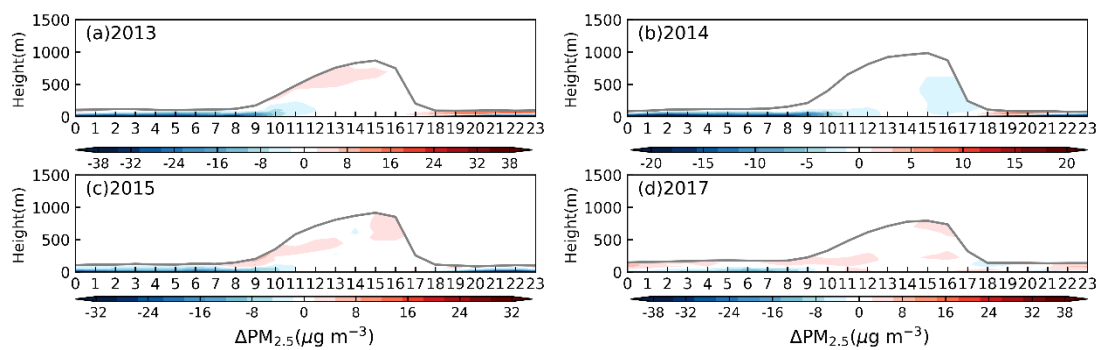


Figure R3. Similar to Figure R1, but in Fuyang.

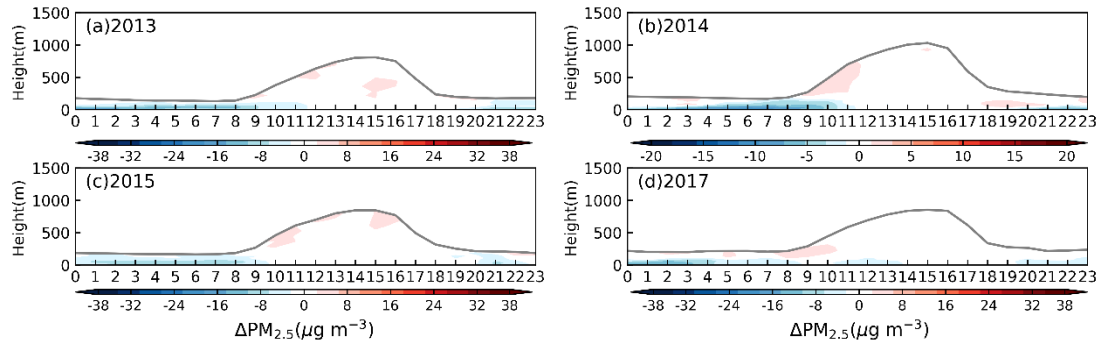


Figure R4. Similar to Figure R1, but in Nanjing.

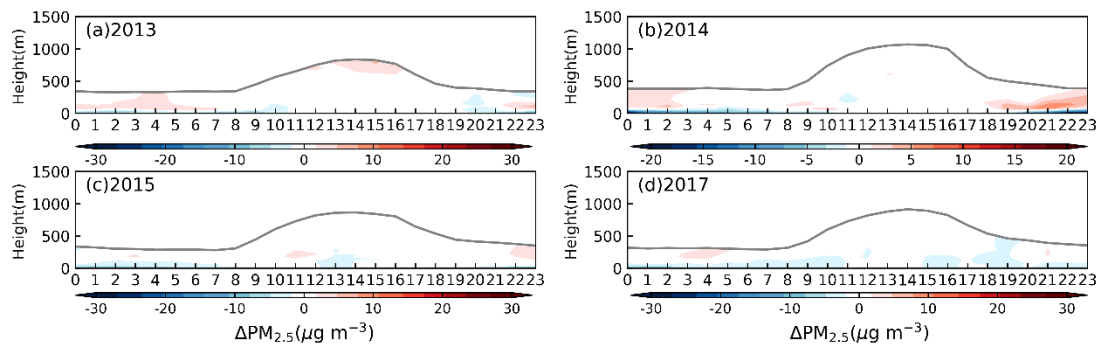


Figure R5. Similar to Figure R1, but in Shanghai.

References

Ren, Y., Zhang, H., Zhang, X., Wei, W., Li, Q., Wu, B., Cai, X., Song, Y., Kang, L., and Zhu, T.: Turbulence barrier effect during heavy haze pollution events, *Sci. Total Environ.*, 753, 142286, <https://doi.org/10.1016/j.scitotenv.2020.142286>, 2021.