Referee report for acp-2021-435

General Comments:

The scheme of turbulent mixing process of particles in the atmospheric boundary layer directly influences the particle concentration predicted by the numerical models. In this paper, the authors introduce a new scheme of turbulent diffusion coefficient, which is different from that for scalars, to describe the turbulent mixing process of particles in WRF-Chem model. The new scheme is only for stable conditions, while under unstable conditions the scheme is not changed. The results show that the new scheme can improve the prediction of particle mass concentration when compared to the output of model using the original scheme. However, the physics behind the results seems problematic. The numerical simulations show that the model using the new scheme reduces the overestimated PM2.5 concentration simulated using the original scheme. But the new scheme has a smaller turbulent diffusion coefficient (TDC) than the original one. It means that the model using the new scheme should predict larger PM2.5 concentration than using the original scheme. I cannot understand why a smaller TDC will lead to smaller PM2.5 concentration in the stable atmospheric boundary layer. So I think the authors should make additional analysis on their simulation results and tell us why a smaller TDC can result in smaller PM2.5 concentration under stable conditions (I mean, the authors should tell us the real reason for the reduced PM2.5 concentration simulated using the new scheme). Additionally, in this paper the authors provide some evidence to argue that the new scheme is reasonable for describing the turbulent mixing process of particles. I think the evidence is not convincing. In Section 3 the analyses of observational data show that the behaviors of transport efficiencies for heat and particle are different. But this result does not support the use of a smaller TDC for turbulent mixing of particle. As for the discussions in Section 5, the provided evidence does not tell us the physics that the model using the new scheme can have the better performance in predicting the PM2.5 concentration under stable conditions. So I think the presentation of this paper is not well done and the conclusions are unconvincing. My recommendation is major revision.

Specific Comments:

1) One of my major concerns is why the model using a smaller TDC can predict smaller PM2.5 concentration under stable conditions. Eqs. (2)–(5) give the new scheme of eddy diffusivities (the authors call eddy diffusivity as TDC) for heat, momentum and particle under stable conditions, in which the eddy diffusivity for particle K_c is different to that for heat K_h. Actually, the difference between K_c and K_h is embodied by the different Ri-dependant functions, f_c and f_h, as expressed in Eq. (5) and Eq. (3) respectively. The two equations indicate that in the new scheme f_c is smaller than f_h (in the original scheme f_c is equal to f_h, as expressed in Eq. (3)). Therefore the new scheme has smaller eddy diffusivity for particle than the original scheme. To our knowledge, the smaller eddy diffusivity means the weaker turbulent transport, which results in higher concentration of air pollutant when other

conditions are the same. However, the simulations in this paper show that the model using the new scheme predicts smaller PM2.5 concentration than the model using the original scheme. My question is why. I think it is the most important issue in this paper. The authors should do additional analyses on the simulation results to find the reasons and tell us what the reasons are.

2) In Section 3, the authors present the analytical results of correlation coefficients R_{wt}, R_{wc} and $R_{wt,wc}$ from observational data. They point out that the behaviors of turbulent transport of heat and particle are different, as shown in Figs. 2b–2d. Thus they use these results to support their choice of a different scheme of turbulent transport for particle from that for heat. In think these results do not make sense. On the one hand, Figs. 2b and 2c show that R_{wt} and R_{wc} are quite different under unstable conditions (i.e., in the daytime), but the authors choose the same scheme of turbulent transport for heat and particle. On the other hand, the behavior of R_{wc} is not changed under both stable and unstable conditions—the values of R_{wc} fluctuate around zero in the whole day as shown in Fig. 2c, but the authors choose different schemes of turbulent transport for particle under stable and unstable conditions respectively. As for the results show in Fig. 2d, I do not know what the results mean and how to interpret them. In my opinion, Section 3 is not a necessary part for this paper, because the results in this section cannot provide evidence to support the choice of a different scheme of turbulent transport for particle from that for heat under stable conditions. Actually, the authors choose the new scheme according to the results in Jia et al. (2021). They have cited the literature in this paper. That's enough. So I suggest the authors to delete this part, as well as Fig. S1.

3) For the results shown in Fig. 6, the authors state in lines 217-222 "Theoretically, increasing turbulent diffusion will reduce the pollutant concentrations near the surface-layer, and the pollutants will be more fully mixing in the vertical direction, which results in lower concentrations of pollutants in the near surface-layer and higher concentrations of pollutants in the upper layer. Actually, the pollutant concentration is reduced in the surface-layer and it is increased in the upper layer at night (Fig. 6), which is consistent with the theory". If the new scheme has a larger f_c, it is likely that these statements are reasonable. However, the new scheme actually has a smaller f_c. How to interpret the results shown in Fig. 6? Of course, a smaller f_c unnecessarily means a smaller eddy diffusivity K_c. As expressed in Eq. (2), if f_c becomes smaller, while the wind shear becomes much larger, we can still obtain a larger K_c. Does the model using the new scheme predict larger near-surface wind shear?

4) In subsections 5.1 and 5.2 the authors emphasize that the meteorological parameters and PBL height simulated by the new scheme are not changed when compared to those simulated by the original scheme (They provide evidence shown in Figs. S2–S5). They declare in lines 232-233 "noting that the new scheme does not alter the performance of meteorological fields, which is an advantage of the new scheme". They also declare in lines 264-266 "The results of the simulation of pollutant concentration are improved under the similar PBLH, which further demonstrates that the simulation of pollutant concentration is not only controlled by the

PBLH". I think these evidence cannot help us to know why the new scheme can reduce the overestimated PM2.5 concentration simulated by the original scheme. Given the unchanged meteorological field as well as PBLH, it seems that the reduced PM2.5 concentration can only be attributed to the new scheme of TDC. However, if the meteorological field and PBLH are really unchanged, the new scheme will have a smaller TDC and should predict larger PM2.5 concentration. So I suggest the authors to provide additional information about the simulation results. These information should tell us what are changed, as well as the relation between these changes and changed PM2.5 concentration.

5) Following the above comment, the same situation exists in subsection 5.3. I think the discussion about the influence of mountain terrain on the simulated PM2.5 concentration also cannot help us to know why the new scheme can reduce the overestimated PM2.5 concentration simulated by the original scheme. Furthermore, I do not understand the purpose of presenting the CO results in this subsection. Is the TDC for CO the same as that for particle in the new scheme? If not, why can the CO concentration be improved by the new scheme?

I have to say that the paper should be revised substantially. So I think there is no need to give the technical comments. Compared to my major concerns listed above, the technical problems are not important in the present stage.