Interactive comment on “The Contributions to the Explosive Growth of PM2:5 Mass due to Aerosols-Radiation Feedback and Further Decrease in Turbulent Diffusion during a Red-alert Heavy Haze in JING-JIN-JI in China” by Hong Wang et al.

Anonymous Referee #2

Received and published: 31 July 2018

This paper investigated the impact of aerosol radiation feedback and decreased turbulent diffusion on PM2.5 during a heavy polluted episode in China. The objectives of this research might be interesting and potentially important; however, I have a number of concerns with the manuscript.

Response:

We would like to heartily thank the reviewer for his serious review and so detailed comments on our work. We carefully considered comments of the reviewer and tried our best to revise the paper accordingly, one by one of the following:

General comments:

Comment 1:

First, the lack of description about the GRPAES_CUACE model is troubling. What are the basic physical parameterizing schemes and chemical mechanism used in this study? How the model treat those crucial processes, such as SOA formation, two-way coupling, BC mixing states, aging processes. More important, how the model calculate the diffusion mixing? Any deficiency that can explain the supposed underestimation in diffusion coefficient, beside the lack of the aerosol radiative effect?

Response:

Thanks for this valuable comment. The section 2.1 (line 84-125) is rewritten in the revised paper according to this comment.

First, the model description GRPAES_CUACE including dynamic, physical and chemical processes is given in section 2.1. The parameterizing schemes and chemical mechanism used in this study and the related references are summarized in new Table 1 in the revised paper. Beside the lack of the aerosol radiative effect, the under estimation of turbulences diffusion coefficient of chemical tracers calculated by PBL is supposed an important reason (DTD experiment) and it is mainly discussed in this paper

Second, CAUCE, two-way coupling and the calculation method of diffusion mixing in PBL scheme are closely related with the aim of this study, so an introduction of this is given in section 2.1 (line 107-126) and the references are also added in the revised manuscript.

Yes, chemical processes involving such as SOA formation, BC mixing states, aging processes are very important to $PM_{2.5}$ concentration, considering this content is not our major focus in this study and it had been introduced and evaluated in previous studies (Gong and Zhang, 2007; Gong et al., 2012; Wang et al., 2010, 2015a,; Zhou et al., 2008, 2012, 2016).

We add a brief introduction in section 2.1 to explain this and the offered the related references are added in the revised paper.

Comment 2:

Second, I suggest the authors to provide additional validation of the model performance. How was the model performance in simulating the meteorological variables, PM chemical components and precursors? Does the underestimation apply to all PM components? It is also very important to exam that how the change in diffusion influence on the model performance in simulating species including both PM chemical components and precursor, since the mixing process is critical in determining the concentrations of all species.

Response:

Considering the comment, some meteorology parameters close related with diffusion turbulence, such as wind speed, temperature and downward short wave fluxes are added to provide the model performance (figure 3 and figure 5 and the related discussion on the figures are added in the revised manuscript). The three sensitive experiments are applied to all PM components.

Yes, mixing process is also critical in determining the concentrations of all aerosols species and precursor, but the discussion on PM chemical components and precursors are complex and will take up a great deal of space in the manuscript, considering the existing studies of the chemical processes by CUACE model (Gong and Zhang, 2007; Gong et al., 2012; Wang et al., 2010, 2015a,; Zhou et al., 2008, 2012, 2016) the focus of this study, observational aerosol optical depth (AOD) and single scattering albedo (SSA) are the related with chemical components (absorbing and scattering features) and direct impact on aerosols radiative feedback directly, so the two are added to evaluate the model performance (added table 4 in the revised manuscript).

Anyway, the we are grateful for this valuable comment and will try our best to collect more observational data to focus on how the change in diffusion influence on the model performance in simulating species including both PM chemical components and precursor, since the mixing process is critical in determining the concentrations of all species in the following study.

Comment 3:

Third, the description about scenario design need be elaborated. In EXP_td_af, how the dynamic field is updated by the aerosol feedback, and is there any nudging processed? In EXP_td20_af, how was the 80% reduction in turbulent diffusion implemented in the model. Did the change apply to all simulated domains? Is there any evidence or references which can support such modification? Based on the results (overestimation is found for clean days and areas outside JJJ), I don't think the DTD is applicable for all grid cells and days and can explain the underestimation of $PM_{2.5}$.

Response:

The mechanism of aerosols feedback on the dynamic is added in table 1 and the introduction of aerosol feedback and related references are added on line 107-117. There isn't nesting domain in the experiments.

The 80% reduction in turbulent diffusion coefficient (DC) is implemented in the chemical tracers (gas and particles) in the chemical module CUCAE. DC outside the CAUCE is not changed in the other parts of the model. Yes, The 80% reduction is applied to all simulated domain, but JING-JIN-JI region is mainly discussed in this study.

The solar radiation is the major cause of turbulence diffusion and PBLH diurnal changing during daytime. The observation study showed that the direct solar radiation on severe haze days is reduced 89% comparing with clear day in Beijing during the same period with this study (the following figure if from the result by Zhong, J.T., et al., 2018). The 80% reduction of turbulence diffusion is mainly according to this study. This reason is also added in section 2.4, Line 162-178; The wind speed changing (also an indicator of turbulence diffusion) from clear to haze days is added (figure 3 in the revised manuscript) in the revised paper, which also support the supposing of 80% reduction of DC.

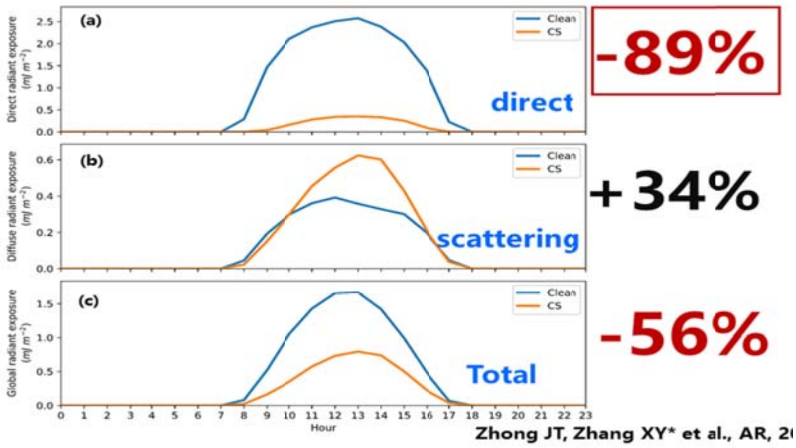

Fig. 4. Daily radiant exposure of all selected clean days before 9 HPEs with CSs and all selected polluted days during the CSs of the HPEs. (a). Daily direct radiant exposure; (b). Daily diffuse radiant exposure; (b). Daily global radiant exposure

Yes, we agree that 80% reduction of DC is not simple applicable for all grid cells and days and can accurately explain all the underestimation of $PM_{2.5}$ and out study did show DTD experiment is meaningful on $PM_{2.5}$ EG in the Jing-Jin-Ji region. Anyway, we know even in Jing-Jin-Ji region, this study is only a sensitive experiment to explain the possible huge deficiency in the description of the extreme weak turbulence of the PBL scheme and 80% reduction may be not an accurate value in every grid point. It is very difficult at present to offer the direct proof of the truth turbulence diffusion condition leading to severe haze episode due to the lack of vertical PBL observations during daytime in this region. We added a short paragraph in the end in the conclusion section to explain the limitations of this study.

Specific comments

Title: need provide some description about “Red-alert” in introduction section

Response:

The description about “Red-alert” is added in introduction section (Line 21-23) in the revised manuscript.

Line 83: “GRAPES_CUACE”, provide the full name and some references about the model.

Response:

The related content (line 84-90) and references (line 398,404,413) was added in the revised manuscript.

Line 89: How to get the boundary conditions?

Response:

No boundary conditions or related text is discussed in this line, so we don't know what the meaning of this comment is.

Line92: "The model horizontal resolution is adopted as 0.15*0.15". Is it high enough to capture the strong inversion during the episode? What about the vertical resolution?

Response:

The horizontal is optional in our model. Considering the resolution of emission inventory in China mainland obtained at present, 0.15*0.15 horizontal resolution is adopted in this study. If the model horizontal resolution is much higher than the resolution of emission data, model produces certain misleading results according to our experience. There are 33 vertical layers from surface to about 30 kilometers of the model top. Some introduction is added in line 96-97 in the revised manuscript. Our previous studies (Wang et al., 2015a; 2015b) showed that 0.15*0.15 horizontal resolution and the vertical layers used in this study had not much impact on the capturing of the strong temperature inversion.

Line 100: I would suggest the authors to elaborate the section 2.2. Is the emission data open to the public? What's the accuracy of the data? How does it compare to the others inventories, such as MEIC, EDGAR, etc? How was the spatial / temporal allocation processed?

Response

Yes, we couldn't give the complete and accurate description of the emission used in this study. The introduction of emission data including spatial and temporal information is rewrritten in section 2.2 in the revised manuscript.

In fact, we have long-term cooperation with MEIC team and may obtain the latest emission data from them. However, the emission condition in Jing-Jin-Ji region in China changed so rapidly, and our model is an operational haze forecast model in Chinese Meteorology Administration and we often find the MEIC emission data is time-lag for the real time forecasting, we had to do some corrections to MEIC emission data according to the latest emission reduction information in this region before using it.

The emission data used in this study may be opened to the editor and reviewer, even to the public if this is required. We didn't use EDGAR emission data in our model also considering the rapid changes of emission in this region.

Line 101: "human life", is it "domestic"?

Response:

Yes, "human life" is replaced by "domestic" in this line.

Line 105-106: need provide full names for the VOC species

Response:

17 VOCs species listed in table 2 and the full names are also given in table 2.

Line 121: "a further 80% decrease in turbulent diffusion (DTD) of chemical tracers based on EXP_td_af representing a compensation for the insufficient description of extremely weak turbulent diffusion by PBL scheme in atmospheric chemical model". how the 80% decreased DTD was determined? Was the overestimation of vertical mixing is due to the coarse resolution, or underestimation of aerosol feedback?

Response:

The 80% reduction of turbulence diffusion is according to the reference by Zhong, J.T., et al., 2018 and the wind speed changing from clear to haze day (added figure 3 in the revised manuscript). In his study, the observation of direct downward short wave fluxes decrease about 89% in Beijing at the same period (This is the base of 80% DTD in section 2.4, the related explanation is added in section 2.4 in the revised paper). Even though, we know that 80% DTD is only a sensitive test and not a definite value in every grid point.

Even if the he coarse resolution do has some impacts on the vertical mixing, the impacts could not be so greatly only during the EG stage of $PM_{2.5}$. We had been used a model 0.1*0.1 horizontal resolution and the results is basically same with the original. Aerosol feedback is one important reason, but not the all according to the results of the three experiments in this study.

Line 134: in section 3.1, what about PM chemical component? The mixing basically can revolve the total PM mass. However, if the chemical profile doesn't agree well the observation, it still cannot solve the issue.

Response:

Yes, PM chemical component is important, we can't find proper observational PM chemical components to compare with model outputs, considering observational aerosol optical depth (AOD) and single scattering albedo (SSA) are the important parameters related with chemical components (absorbing and scattering features) and impacting aerosols radiative feedback, so the two are added to evaluate the model performance (added table 4 and related text in the revised manuscript).

Line 155: "Some studies offline and online", is it "some offline/online modeling studies"?

Response:

Yes，this is revised in the manuscript.

Line 157: "AF of composite aerosols from black carbon, organic carbon, sulfate, nitrate, dust, ammonium, and sea salt aerosols had been online coupled into the in GRAPES_CAUCE model." how does the model treat mixing states and aging process? How is the model performance in simulating the PM components and AF?

Response:

The mixing method of black carbon, organic carbon, sulfate, nitrate, dust, ammonium, and sea salt aerosols was mainly introduced in previous study (Wang et al., 2015a). A brief introduction is also added in the 109-112.

Observational aerosol optical depth (AOD) and single scattering albedo (SSA) are the important parameters close related with chemical components (absorbing and scattering features) and they are also define the AF effects directly, so the two are added to evaluate the model performance (added table 4 and the related text in the revised manuscript).

Line 173: "the temperature inversion layer pre-existed during the haze event", it is not easy to see the temperature inversion in the plots.

Response:

It is easy to see in figure 7a and figure 7b, not in figure 6 in the revised manuscript. There is similar phrase in the discussion on figure 7a and figure 7b, so, this phrase is deleted in the revised manuscript.

Line 182: "Figure 4b shows that the observed temperature inversions were obvious stronger and the inversion depth thicker on 18 to 19 (during EGS of PM2.5) than those on 15 to 16 Dec (CS of PM2.5" But the PBL height seems opposite, lower on 18 to 19 but higher on 15 to 16 Dec.

Response:

No PBL height was displayed in this study. We are not sure where the reviewer drew the conclusion "But the PBL height seems opposite, lower on 18 to 19 but higher on 15 to 16 Dec"

According our previous studies (Wang et al., 2015a, 2015b), when the temperature inversion is stronger, the corresponding PBL height is lower and $PM_{2.5}$ is higher.

Line 191: "The contributions to PM2.5 EG due to AF and DTD". Since AF also contributes to DTD, how to separate these two effects.

Response:

The contribution to $PM_{2.5}$ due to AF means the $PM_{2.5}$ changing due to aerosols feedback online (EXP2 in the revised paper), only including the diffusions reduction by AF, but not including 80% reduction of DC; The results of DTD (EXP3 in the revised paper) means the differences between EXP3-EXP2, it does not include the AF's contribution, but only the decrease of turbulence diffusion of coefficient of chemical tracers. In EXP3, The DTD is implemented in the chemical tracers (gas and particles) in the chemical module domain. DC outside the CAUCE is not changed in the model run.

Line 207: "Exp_bk under underestimated the PM2.5", "under" should be deleted

Response:

"under" is deleted in the text.

Line 224: "the overestimation of turbulent DC", is there any observation data to prove the overestimation of DC?

Response:

There isn't direct observation data of DC, but the daytime direct shortwave downward fluxes reduced 89% during EG stage in Beijing in the reference (Zhong et al., 2018), solar radiation is the major cause if turbulence diffusion when the wind speed is small. The wind changing analysis (in added figure 2) from clear to haze days also support this supposing. This is the base of our sensitive experiment (the corresponding explanation is added in section 2.4)

Figure 2: The PM2.5 in area outside JJJ seems all overestimated. The td_af cases make it even worse. Seems like it is not proper to apply the 80% DTD to all grid cells.

Response:

$PM_{2.5}$_obs is the station observation data and the each color dot represents the value in the station, the white color stands for lack of observation data not the lower PM2.5 value < $35ug/m^3$, which is not completely same with the modeled $PM_{2.5}$ on grid points with high resolution. Excluding this reason, $PM_{2.5}$ by EXP3 (td20_af in initial manuscript) is still the best in general, then EXP2 (td_af), and EXP1 is the worst in Jing-Jin-Ji comparing with observation $PM_{2.5}$. Outside JJJ, td_af cases make it worse in the area with low $PM_{2.5}$, make it better in the area with higher $PM_{2.5}$. Anyway, this study mainly focus on Jing-Jin-Ji region.

Certainly, we agree 80% DTD may be not accurate to all grid cells even in Jinh-Jin-Ji region. Our study area is Jing-Jin-Ji and even in this area the 80% DTD can't represents the exact condition of turbulence diffusion in all grid cells. Our study is sensitive experiment and we hope the underestimation of high $PM_{2.5}$ due to the distinct deficiency of PBL scheme in the description of the extreme weak turbulence diffusion in Jing-Jin-Ji in east China may cause attention by this sensitive experiment. The final solution for this underestimation depends on the improving of PBL algorithm base on more detailed observation of PBL meteorology scales, not the simple decreasing of DC.

A paragraph is added in the last in section 4 to explain all above limitations and the other possible reasons leading to the underestimation in this study.

Figure 3: please clarify that the data is regional average in JJJ.

Response:

This is revised in the caption of figure 3.

Figure 4: what about the days when PM reach peak for Dec 20-22 in Beijing.

Response:

The inversion and the impacts on it due to AF is similar in 20-22 with that in EG stage. The explanation about this is added in the text after this figure.

Figure 5: PM2.5_td_af seems more reasonable than $PM_{2.5}$_td20_af, in consideration of the possible missing heterogeneous chemistry. What's the reason for the underestimation of the peak on Dec 21, even though the DC is already very low.

Response:

CUACE model includes a simple scheme of heterogeneous chemistry of SO_2 and the related explanation is added in section 2.1 in the revised model.

Yes, "heterogeneous chemistry" is a very important influencing factor to $PM_{2.5}$ concentrations, but there are also many uncertainties of this influence due to a series of complex chemical processes and species. At present, it is very difficult to offer a quantitative estimation of the impacts of heterogeneous chemistry on $PM_{2.5}$ either in observation or in model.

There are several causes impacting local $PM_{2.5}$ concentration involving in emission, meteorology, atmospheric chemical processes in including gas-particles and "heterogeneous chemistry" and etc. Some studies emphasize the impacts of meteorology condition including the feedback from AF. Some studies stressed the impacts of heterogeneous chemistry on $PM_{2.5}$. It's a controversial issue. This study mainly focuses on meteorology impacts from turbulence diffusion and aerosols feedback. Anyway, this is a limitation of this study and it is explained in the last paragraph in section 4 in the revised paper.

The $PM_{2.5}$ and DC condition on December 21 mainly related with changing of meteorology condition such as the inversions and wind fields, a sort explanation is added in this paragraph to explain it.

Figure 6: the figure is misleading. Since the reduced error in td20_af is because that the overestimation on Dec 18 compensates the underestimation on Dec 21 in Beijing.

Response:

The result of this figure is calculated by the model result from 00 UTC 17 to 00 UTC on 21 December, the data in 21 December is not included in the calculation. The description of CS and EG is not accurate and it is corrected in section 3.1 in the revised paper.