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Discussions

The Contributions to the Explosive Growth of $PM_{2.5}$ Mass due to Aerosols-Radiation Feedback and Further Decrease in Turbulent Diffusion during a Red-alert Heavy Haze in JING-JIN-JI in China

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Abstract. The explosive growth (EG) of $PM_{2.5}$ mass usually resulted in $PM_{2.5}$ extreme levels and severe haze pollution in east China and they were generally underestimated by current atmospheric chemical models. Based on the atmospheric chemical model GRPAES_CUACE, three experiments of background (EXP_bk), normal turbulent diffusion and aerosols feedback (EXP_td_af), and retaining 20% of normal turbulent diffusion of chemical tracers of EXP_td_af (EXP_td20_af) are designed to study the contributions to the EG of $PM_{2.5}$ due to aerosols-radiation feedback (AF) and further decrease in turbulent diffusion (DTD) focusing on a red-alert heavy haze in JING-JIN-JI of China. The study results showed that turbulent diffusion coefficient (DC) calculated by EXP_bk is about 60-70m^2/s on clear day and 30-35m^2/s on haze day. This difference of DC was not enough to discriminate the unstable atmosphere on clear day and extreme stable atmosphere during EG stage of $PM_{2.5}$, and the inversion calculated by EXP_bk was obviously weaker than the actual atmosphere of sounding observation on haze day. This led to 40-51% underestimation of $PM_{2.5}$ EG by EXP_bk; AF reduced about 43-57% of DC during EG stage of $PM_{2.5}$, which strengthened the local inversion obviously on haze day and local inversion by EXP_td_af was much

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closer to the sounding observation than that by EXP_bk. This resulted in 20-25% reduction of model errors of $PM_{2.5}$ and it was as low as -16 to -11%. However, the inversion by EXP_td_af was still weaker than the actual observation and AF could not solve all the problems of $PM_{2.5}$ underestimation. Based on EXP_td_af, 80% DTD of chemical tracers resulted in a near-zero turbulent diffusion named as "turbulent intermittent " atmosphere state in EXP_td20_af resulting in a further 14-20% reduction of $PM_{2.5}$ underestimation and the negative $PM_{2.5}$ errors of was reduced to -11 to 2% during the EG stage of $PM_{2.5}$. The combined effects of AF and DTD solved over 79% underestimation of $PM_{2.5}$ EG in this case study. The results showed that the online calculation of aerosol-radiation feedback and a further improving arithmetic of PBL scheme focusing on extreme stable atmosphere stratification are indispensable for reasonable description of local "turbulent intermittent" and more accurate prediction of $PM_{2.5}$ EG and high levels during the severe haze in Jing-Jin-Ji in China.

Keywords: Aerosols-Radiation Feedback; Turbulent Diffusion; PBL Scheme; Temperature Inversion; $PM_{2.5}$

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1 Introduction

East china experienced unprecedented intrusions of severe hazes accompanied by high level of particulate matter less than 2.5 micron in aerodynamic diameter ($PM_{2.5}$) caused wide public concern since 2013 until now (Ding et al., 2013; Wang et al. 2013; Huang et al., 2014; Wang et al., 2014; Sun et al., 2014; Hua et al., 2015; Yang et al., 2015; Yang et al., 2016; Zhong et al., 2017, 2018). Instant $PM_{2.5}$ concentration usually reached hundreds, or even one thousand ug/m^3 occasionally, in the metropolitans in Beijing (JING), Tianjin (JIN), Hebei province (alias JI) and their near surroundings of East Shanxi, West Shandong, and North Henan in east China (abbreviated this region as JING-JIN-JI in this study) during severe haze episodes (Wang et al., 2014; Quan et al., 2014; Sun et al., 2014; Yang et al., 2015; Zheng et al., 2016). Studies showed that models generally underestimated the explosive growth (EG) and peak values of $PM_{2.5}$ during the severe hazes in Jing-Jin-Ji in China (Wang et al., 2013; Wang et al., 2014; Li et al., 2016).

The causes of $PM_{2.5}$ EG and its underestimation by atmosphere chemical models are complex and uncertain at present, which may involve in local emission, reginal transportation, aerosol physicochemical processes, gases-particles conversion, meteorology condition, and so on. However, the actual atmospheric stability and how accurate it is described by atmospheric models is a fundamental problem that can't be ignored among others. Local or regional meteorology condition dictates whether the haze occurs and what the $PM_{2.5}$ level may be (Zhang et al., 2013; Zheng et al., 2015; Gao et al., 2016) when source emissions are unchanged for a short period of time. The meteorology condition of planetary boundary layer (PBL) is the key and direct trigger for touching off a haze event (Wang et al., 2014; Li et al., 2016; Zhong et al., 2017). Turbulent diffusion is an important factor to characterize PBL meteorology when the atmosphere is stable. It is also the major way of particles and gas pollutants exchanging from surface to upper atmosphere and further cleaned by the upper winds when haze occurs accompanied by calm surface wind and weak vertical motion of air in surface and PBL. The intensity of turbulent diffusion largely determines the severity of haze pollution. Reasonable description of turbulent diffusion by PBL schemes in atmospheric chemical models is determinant for severe pollution prediction (Hong et al., 2006; Wang et al., 2015; Hu et al., 2012, 2013a, 2013b; Li et al., 2016). The latest studies showed (Wang et al., 2015; Li et al., 2016) that current PBL schemes may be insufficient enough for describing the extreme weak turbulent diffusion condition when extremely severe hazes occurred in JING-JIN-JI, which may be one important reason for the underestimating of $PM_{2.5}$ peaks by model. There may be two independent reasons resulting in this deficiency description of extreme weak turbulent diffusion in atmospheric models. One is that aerosols radiation feedback (AF) is not calculated online in the model run. AF can restrain turbulence by cooling surface and PBL while heating the atmosphere above it (Wang et al., 2010; Forkel et al., 2012; Gao et al., 2014, 2015; Wang et al., 2015; Ding et al., 2016; Li et al., 2016; Miao et al., 2106; Petaja et al., 2016; Gao et al., 2017; Qiu et al., 2017; Zhong et al., 2018). Ignoring AF is likely to lead to obvious overestimation of turbulent diffusion when $PM_{2.5}$ exceeds certain value, which is worthy of further study. Another possible reason is that the extreme weak turbulence resulting to extremely severe hazes is not fully described by the atmospheric chemical model (Li et al., 2016). A Red-alert Heavy Haze occurred on 15 to 17 December, 2016 in JING-JIN-JI in China was elected to study the contributions to $PM_{2.5}$ EG and peaks during severe haze due to AF and the possible deficiency in description of the extreme weak turbulent diffusion of atmosphere models in this study.

2 Model, Data and Methodology

2.1 GRAPES_CUACE Model

The double way atmospheric chemical model GRAPES_CUACE was established focusing on simulation and prediction of dust and haze pollutions in China and East Asia. Trans-city and regional transportation of $PM_{2.5}$, aerosols-radiation-PBL-meteorology interactions, and aerosols-cloud interactions etc. had been widely simulated and studied by using it (Wang et al., 2009, 2010, 2015a, 2015b; Zhou et al., 2012, 2016; Jiang et al., 2015; Zhang et al., 2018). GRAPES_CUACE is also used in this study. Considering interregional transport of gas and particle pollutants in the main polluted areas in eastern China, the model domain includes the whole east China (100-140°E, 20-60°N) (figure 1a), but our study mainly focuses on Jing-Jin-Ji region (the red box in figure 1a). Figure 1b shows the detailed geographical location and topography of JING-Jin-Ji. The black dots in Figure1a are the locations of $PM_{2.5}$ observation stations. The model horizontal resolution is adopted as 0.15°×0.15° to match the resolution of emission source data used in this study. There are two balloon sounding stations, Xingtai and Beijing (Figure 1b) in our study area. Xingtai, located in southern Hebei province, the eastern foot of Taihang Mountains and it is influenced by the sinking airflow from Taihang Mountains in winter, is the most polluted city and the $PM_{2.5}$

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concentrations usually ranked the first in China in recently years. The topography of Xingtai and the serious haze pollution closely related to it are the typical representative of the southern plain of Jing-Jin-Ji. Beijing lies in the transitional zone from Yan Mountain to its southern plain, next to Tianjin and surrounded by Hebei, representing the polluted areas in the central part of Jing-Jin-Ji.

2.2 Emission Inventory

5 kinds of emission sources of industrial, human life, agricultural, natural and traffic are obtained by the data statistics of China national industry factories, energy consumption, road net and motor vehicles, population information, land use, vegetation cover and etc. in 2015. The 32 kinds of monthly gridded emission inventories of 0.15°×0.15° horizontal resolution required by GRAPES_CUACE model, including 5 reactive gases, i.e. SO_2, NO, NO_2, CO, NH_3, 20 VOCs, i.e. ALD, CH_4, CSL, ETH, HC_3, HC_5, HC_8, HCHO, ISOP, KET, NR, OL_2, OLE, OLI, OLT, ORA_2, PAR, TERPB, TOL, XYL and 5 aerosols species, i.e. black carbon, organic carbon, sulfate, nitrate and fugitive dust, are based on above five emission sources according to the emission mode and VOCs partition scheme by CAO (Cao et al.,2016, 2010).

2.3 Data Used

Hourly averaged observation $PM_{2.5}$ data for more than 1440 surface observational stations from China National Environmental Monitoring Centre (CNEMC) (http://www.cnemc.cn) from 15 to 23 December 2016 were used to evaluate the model results. The meteorological balloon sounding data at 00UTC (early morning) and 12UTC (and dusk in local time) in Xingtai and Beijing from China Meteorology Administration (CMA) during the same period were also used compare with the modeled results. NCEP 0.25×0.25° global analysis grids data (https://rda.ucar.edu/datasets/ds083.3) were used as the model initial and every 6-hour lateral boundary meteorology input fields. The initial values of chemical tracers were obtained according to the five-year mean climatic values. The results of the first 120 hours of model start are split out to eliminate the effects of chemical initial fields.

2.4 Experiments Design

Three experiments of EXP_bk, EXP_td_af, and EXP_td20_af were designed to discuss the relative contributions to $PM_{2.5}$ EG due to AF and 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 (the detailed descriptions of the experiments listed in Table 1). All other model dynamic process, physical options and initial input data of meteorology and chemical tracers are same for the three experiments except for the differences shown in Table 1.

3 Results and Discussions

This haze episode began on 15 December, 2016. $PM_{2.5}$ began to gather and climb slowly from 15 to 16, but were below 150 ug/m^3 in most JING-Jin-Ji region and we name this period as the climbing stage (CS) of $PM_{2.5}$; From 17 to 20 December, $PM_{2.5}$ increased sharply and most of the study area reached the $PM_{2.5}$ peaks of 400-600 ug/m^3 rapidly during this period, which is named as the explosive growth (EG) stage (EGS) of $PM_{2.5}$. This section mainly focuses on the contributions to the $PM_{2.5}$ EG due to AF and further DTD.

3.1 The Comparison study of observation and three experiments

Figure 2 displays the averaged observed $PM_{2.5}$ ($PM_{2.5}$_OBS) and simulated $PM_{2.5}$ of Exp_bk ($PM_{2.5}$_bk), EXP_td_af ($PM_{2.5}$_td_af) and EXP_td20_tf ($PM_{2.5}$_td20_af) experiments during EGS. It can be seen from $PM_{2.5}$_OBS that the averaged $PM_{2.5}$ values were generally over 100μg/m^3 in east China and JING-JIN-JI covered the most polluted areas and $PM_{2.5}$ reached up to 300 to 400μg/m^3 in parts of Beijing, Tianjin, Middle-south Hebei province, western frontier region of Shandong province and north Henan province. The $PM_{2.5}$ center of 500-700μg/m^3 appeared in south Hebei and North Henan province and the $PM_{2.5}$ maximum of 700μg/m^3 was found in south Hebei. The comparison study of $PM_{2.5}$_bk and $PM_{2.5}$_OBS shows that $PM_{2.5}$_bk is obvious lower than $PM_{2.5}$_OBS on the whole. It is noteworthy that EXP_bk fail to simulate the $PM_{2.5}$ over 300μg/m^3. $PM_{2.5}$_OBS is about 200 to 300μg/m^3 over most Shandong province while the $PM_{2.5}$_bk is only 100 to 200μg/m^3 in this region. Compared with $PM_{2.5}$_bk, $PM_{2.5}$_td_af values are significantly improved by AF and they are much closer to the $PM_{2.5}$_OBS. High $PM_{2.5}$_OBS centers of 300 to 400, 400 to 500, 500 to 600μg/m^3 are almost simulated by EXP_td_af, indicating the important effects of AF on the model simulation of $PM_{2.5}$ high values. However, the areas of the simulated $PM_{2.5}$ values of 300 to 400, 400 to 500, 500 to 600μg/m^3 are still smaller than that of the $PM_{2.5}$_OBS. EXP_td_af also fails to simulate the maximum $PM_{2.5}$ values over 600μg/m^3 observed in south

Hebei province. $PM_{2.5}$_td20_af just makes up for this shortage, comparing with $PM_{2.5}$_bk and $PM_{2.5}$_td_af, $PM_{2.5}$_td20_af is undoubtedly the closest to $PM_{2.5}$_OBS both in $PM_{2.5}$ extreme and its influence area. This study result illustrates that both AF and DTD in atmospheric chemical models are required for the effective prediction of $PM_{2.5}$ EG during the severe haze in JING-JIN-JI in China.

3.2 The aerosols reform on local atmosphere temperature profiles

Some studies offline and online indicated the reforming of atmosphere temperature profile due to aerosols direct radiation (Wang et al., 2010, 2015b; Forkel et al., 2012; Gao et al., 2014, 2015; Wang et al., 2014; Gao et al., 2016; Ding et al., 2016). In our previous works (Wang et al., 2015a, 2015b), 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. On this basis, the changes of mean temperature profile of Jing-Jin-Ji region of daytime due to aerosols radiation were calculated from 15 to 20 December, 2016 in this work. It can be seen from Figure 3 that AF cooled the atmosphere below 750 to 800 hPa while warmed the atmosphere above this height. Considering planetary boundary Layer (PBL) height may be as low as several hundreds to one thousand meters when severe hazes occurs in Jing-Jin-Ji (Wang et al., 2015a, Zhong et al., 2017), it may be concluded that whole PBL and its near upper atmosphere was cooled by AF to a different extent during the different stage of this haze. The aerosols' warming effects above 750-850hPa height were very weak and the temperature changes among different days were also small. However, the aerosols' cooling effects shows the most differences from surface to 975 hPa height on different day. The surface daytime cooling is about 2.2 K on 19, 1.5K on 18 and 20, 1K on 17, and 0.5-0.6 K on 15 to 16 December. This aerosols' cooling effect decreased rapidly with the height. The difference of cooling rates between surface and 850hPa is 1.8 K on 19, 1.3K on 18 and 20, 1K on 17, and 0.3-0.4 K on 15 and 16 December. It can be seen that the AF cooling difference between surface and upper PBL during EGS are much bigger than those during CS. Such obvious difference of cooling effect on surface to upper PBL due to AF may result in the further intensification of the temperature inversion layer pre-existed during the haze event.

The vertical sounding meteorology data in Beijing and Xingtai in JING-JIN-JI can be used to prove if this change of the temperature profile by AF is correct. Figure 4 shows the vertical temperature profiles of sounding observation and the modeled temperature profiles of EXP_bk and EXP_td_af during CS (Figure 4a) and EGS (Figure 4b) at the two stations. The temperature profiles (Figure 4a) shows that both modeled results by EXP_bk and EXP_td_af partly simulated the observed temperature inversion in Beijing and Xingtai on 15 to 16. The very little difference between the temperature profiles of EXP_bk and EXP_td_af indicated that aerosols radiation had very little impacts on the temperature profiles and local inversion during CS. Nevertheless, Figure 4b shows that the observed temperature inversions were obvious stronger and the inversion depth thicker on 18 to 19 (during EGS of $PM_{2.5}$) than those on 15 to 16 Dec (CS of $PM_{2.5}$) both in Xingtai and Beijing. The temperate profiles by EXP_td_af were much closer to the observation results than that by EXP_bk, and especially, the temperature inversions were much stronger and also closer to the observation than that by EXP_bk. This result proved the effective correction of local inversions by AF during the EGS of $PM_{2.5}$. However, it also can be seen, that the inversions by EXP_td_af, which included online AF, are still weaker than the truth observed inversion in the two stations, suggesting that except for AF, there must be other causes that the observed extreme strong inversion was not simulated sufficiently by the model. This will be discussed in detail in the following sections.

3.3 The contributions to $PM_{2.5}$ EG due to AF and DTD

Turbulent diffusion process is the main way of gas and particles exchanging from near ground to upper atmosphere and then removed by the high altitude transport, which is usually described by turbulent diffusion coefficient (DC) in the chemical atmospheric models. Firstly, the inversion and weak turbulent diffusion, which generates from atmosphere dynamic process, leads to atmosphere stabilization and determines the occurrence of haze and its strength (Zheng et al., 2017). Once the haze occurs, the aerosols radiation may reinforce the inversion in turn when aerosols exceeds certain critical value and lead to more $PM_{2.5}$ gathering near the ground (Figure 4). The relative importance of the two aspects on $PM_{2.5}$ EG may vary with the $PM_{2.5}$ values and meteorology conditions, but they are irreplaceable for the reasonable prediction and simulation of $PM_{2.5}$ peaks by atmospheric models.

Figure 5 displays the hourly changing of observed $PM_{2.5}$ ($PM_{2.5}$_OBS) and modeled $PM_{2.5}$ of Exp_bk, EXP_td_af, and EXP_td20_tf experiments ($PM_{2.5}$_bk, $PM_{2.5}$_td_af, and $PM_{2.5}$_td20_af), together with the modeled turbulent DC of the three experiments (DC_bk, DC_bk_af, and DC_td20_bf) from 15 to 23

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December in Beijing (Figure5a) and Xingtai (Figure 5b). Comparison of the $PM_{2.5}$_bk, $PM_{2.5}$_td_af, and $PM_{2.5}$_td20_af with $PM_{2.5}$_OBS in Beijing (Figure 5a) shows that the modeled $PM_{2.5}$_td20_af was the closest to $PM_{2.5}$_OBS during the whole haze episode, which was agreed with the results of regional distribution during EGS in Figure 2. Exp_bk under underestimated the $PM_{2.5}$ obviously from 17 to 22 December and this underestimation enlarged rapidly with the increasing of $PM_{2.5}$ values and the difference between the modeled and observed $PM_{2.5}$ was the largest during the EGS of $PM_{2.5}$. AF shortened this difference to a great extent and $PM_{2.5}$_td_af was much closer to $PM_{2.5}$_OBS than $PM_{2.5}$_bk during $PM_{2.5}$ EGS. However, it can be seen that there was still certain differences between $PM_{2.5}$_OBS and $PM_{2.5}$_td_af, illustrating that AF can't completely fill the gap between $PM_{2.5}$_OBS and $PM_{2.5}$_td_af. $PM_{2.5}$_td20_tf shortened this gap further and shows the best agreement with the $PM_{2.5}$_OBS, especially during the EGS.

It also can be seen from figure 5a that the DC_bk was about 30-40 m^2/s during the EGS of $PM_{2.5}$, which was about 50% of the 60-70 m^2/s on the clear day on 15 and 22 December. Obviously, the 50% DC differences between the clear and haze days may be not enough to discriminate the difference of turbulent diffusion intensity between extreme stable atmosphere on haze day and unstable atmosphere on clear day, which may be the important reason for underestimation of $PM_{2.5}$ EG by Exp_bk. AF led to notable enhancement of temperature inversion (Figure 4b), significant decrease in turbulent diffusion on $PM_{2.5}$ during EGS and DC_td_af was as low as $14m^2/s$ on 20 December, which decreased about 50% comparing with DC_bk. DC_td_af on haze day was only about 20% of that on clear day. The DC_td20_af was lower than $5m^2/s$ on 20 December and at the same time $PM_{2.5}$_td20_af was further increased and it was also much further closer to the $PM_{2.5}$_OBS than $PM_{2.5}$_td_af.

It can be seen from the comparative study of the temporal changing between DC and $PM_{2.5}$ of Exp_bk, Exp_td_af, Exp_td20_af in Beijing that the overestimation of turbulent DC owning to lack of online calculation of AF and deficient description of the extreme stable stratification by PBL schemes in atmospheric model led to distinct underestimation of $PM_{2.5}$ EG and peaks when severe haze occurred in Jing-Jin-Ji in China.

The changing trends of DC and $PM_{2.5}$ of the three sensitive experiments in Xingtai (Figure 5b) shows the similar results with those in Beijing. The $PM_{2.5}$ _td20_tf was also the closest to $PM_{2.5}$_OBS, followed by $PM_{2.5}$_td_af and $PM_{2.5}$_bk was the worst during the whole haze episode. However during the EGS of $PM_{2.5}$, the relative contributions on the $PM_{2.5}$ peak values due to AF and DTD showed some difference with those in Beijing. The contributions to $PM_{2.5}$ peaks due to DTD were more important than that by AF in Xingtai. Located at the east foot of the east side of Taihang Mountains, Xingtai is usually affected by the downhill airflow and temperature inversion in this area is easy to form and strengthened, leading to stronger inversion, weaker turbulent diffusion and more stable atmospheric stratification. This kind of inversion and weak turbulent diffusion derived from local terrain is more difficult to described and likely underestimated by PBL scheme in atmospheric chemical models.

Figure 6 shows the diagrammatic sketch of the contributions to the $PM_{2.5}$ of EGS due to AF and DTD. It can be seen that the DC_bk was 30-35m^2/s, DC_td_af was 15-17 m^2/s, means that AF reduces about 43-57% DC based on EXP_bk, which led to the a rise in simulated $PM_{2.5}$ from 144 by EXP_bk to 205 ug/m^3 by EXP_td_af in Beijing, 280 by EXP_bk to 360 ug/m^3 EXP_td_af in Xingtai. This means that AF reduced 20% in Beijing and 25% in Xingtai of simulated $PM_{2.5}$ negative errors. DC_td20_af was as low as 4-6 m^2/s during EGS of $PM_{2.5}$, showing the joint effects of AF and DTD reduced DC value to less than 4-6 m^2/s, near-zero, we name it as "turbulent intermittent". The direct results of this "turbulent intermittent" is the further increasing of simulated $PM_{2.5}$ based on EXP_td_af. DTD decreases 14% to 20% underestimation of simulated $PM_{2.5}$ and the errors of $PM_{2.5}$_td20_af were reduced as low as -11% to 2%.

4 Conclusions

Using atmospheric chemical model GRAPES_CUACE, three experiments EXP_bk, EXP_td_af and EXP_td20_af were designed to study the reason for the explosive growth of $PM_{2.5}$ mass during a red-alert heavy haze occurred on 15 to 23 December, 2016 in JING-JIN-JI in China. The contributions to the $PM_{2.5}$ due to aerosols feedback and a further decrease in turbulent diffusion coefficient of chemical tracers, representing a compensation for the deficient description of extreme weak turbulent diffusion by PBL scheme in atmospheric models, are studied by analysing the changes of $PM_{2.5}$, temperature profiles, diffusion coefficient and the relationship between them.

The study shows that the diffusion coefficient by EXP_bk is about 60-70m^2/s on clear day and 30-35m^2/s on haze day. The 50% difference of the two was not considered enough to discriminate the unstable atmosphere on clear day and extreme stable atmosphere on severe haze day, which is proved by the weaker inversion calculated by EXP_bk than that of the actual sounding observation. This led to 40-51% underestimation of the $PM_{2.5}$ by EXP_bk during the explosive growth stage of $PM_{2.5}$.The surface daytime cooling due to aerosols was 1.5-2.2 K during explosive growth stage of $PM_{2.5}$ and 0.5-0.6 K during climbing stage of $PM_{2.5}$. The impacts on $PM_{2.5}$ due to AF was distinct during the explosive growth stage of $PM_{2.5}$ while very little during climbing stage of $PM_{2.5}$ in the model run, indicating a critical value of 150 ug/m^3 of $PM_{2.5}$ leading to an effective AF in online atmospheric chemical model. This aerosols' cooling effect decreased rapidly with the height and this is the reason for the strengthening of the temperature inversion during the explosive growth stage of $PM_{2.5}$. The local inversion by EXP_td_af was strengthened and closer to the actual sounding observation than it by EXP_bk. This resulted in a 20-25% reduction of $PM_{2.5}$ underestimation and $PM_{2.5}$ errors by EXP_td_af was as low as -16 to -11% during the explosive growth stage of $PM_{2.5}$. However, the local inversion simulated by EXP_td_af was still weaker the actual observation and the $PM_{2.5}$_td_af was still smaller than $PM_{2.5}$ observation, illustrating that AF could not solve all the $PM_{2.5}$ underestimation problems. Further DTD of particles and gas resulted in another 14-20% lessening of $PM_{2.5}$ underestimation based on EXP_td_af and the $PM_{2.5}$ errors of EXP_td20_af was reduced to -11 to 2%.

This study result illustrated that the PBL scheme in current atmospheric chemical models is probably insufficient for describing the extremely stable atmosphere resulting in explosive growth of $PM_{2.5}$ and severe haze in JING-JIN-JI in China, which may involve in two important reasons: One is the absence of online calculation of AF, another is the deficient description of the extreme weak turbulent diffusion by PBL scheme in the atmospheric chemical model. Our study suggests that online calculation of AF and an improvement in arithmetic of turbulent diffusion in PBL schemes focusing on extreme stable atmosphere stratification in atmospheric chemical model are indispensable for reasonable description of local "turbulent intermittent" and accurate prediction the explosive growth and peaks of $PM_{2.5}$ of severe haze in Jing-Jin-Ji in China.

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References

Ding, A.J., Fu, C.B., Yang, X.Q., Sun, J.N., Petäjä, T., Kerminen, V.M., Wang, T., Xie, Y., Herrmann, E., Zheng, L.F., Nie, W., Liu, Q., Wei, X.L., Kulmala, M., 2013. Intense atmospheric pollution modifies weather: a case of mixed biomass burning with fossil fuel combustion pollution in eastern China. Atmos. Chem. Phys. 13 (20), 10545-10554.

Ding, A.J., Huang, X., Nie, W., Sun, J.N., Kerminen, V.M., Petäjä, T., Su, H., Cheng, Y.F., Yang, X.Q., Wang, M.H., Chi, X.G., Wang, J.P., Virkkula, A., Guo, W.D., Yuan, J., Wang, S.Y., Zhang, R.J., Wu, Y.F., Song, Y., Zhu, T., Zilitinkevich, S., Kulmala, M., Fu, C.B., 2016. Enhanced haze pollution by black carbon in megacities in China. Geophys. Res. Lett. 43 (6), 2873-2879.

Forkel, R., Werhahn, J., Hansen, A.B., McKeen, S., Peckham, S., Grell, G., Suppan, P., 2012. Effect of aerosol-radiation feedback on regional air quality – A case study with WRF/Chem. Atmos. Environ. 53, 202-211.

Gao, M., Carmichael, G.R., Saide, P.E., Lu, Z., Yu, M., Streets, D.G., Wang, Z., 2016. Response of winter fine particulate matter concentrations to emission and meteorology changes in North China. Atmos. Chem. Phys. 16 (18), 11837-11851.

Gao, M., Saide, P.E., Xin, J., Wang, Y., Liu, Z., Wang, Y., Wang, Z., Pagowski, M., Guttikunda, S.K., Carmichael, G.R., 2017. Estimates of Health Impacts and Radiative Forcing in Winter Haze in Eastern China through Constraints of Surface PM2.5 Predictions. Environ. Sci. Technol. 51 (4), 2178-2185.

Gao, Y., Zhao, C., Liu, X., Zhang, M., Leung, L.R., 2014. WRF-Chem simulations of aerosols and anthropogenic aerosol radiative forcing in East Asia. Atmos. Environ. 92, 250-266.

Gao, Y., Zhang, M., Liu, Z., Wang, L., Wang, P., Xia, X., Tao, M., Zhu, L., 2015. Modeling the feedback between aerosol and meteorological variables in the atmospheric boundary layer during a severe fog–

haze event over the North China Plain. Atmos. Chem. Phys. 15 (8), 4279-4295.

Gong, S.L., Zhang, X.Y., 2007. CUACE/Dust – an integrated system of observation and modeling systems for operational dust forecasting in Asia. Atmos. Chem. Phys. Discuss. 7 (4), 1061-1067.

Hong, S.Y., Noh, Y., Dudhia, J., 2006. A New Vertical Diffusion Package with an Explicit Treatment of Entrainment Processes. Mon. Weather Rev. 134 (9), 2318-2341.

Hu, X.M., Doughty, D.C., Sanchez, K.J., Joseph, E., Fuentes, J.D., 2012. Ozone variability in the atmospheric boundary layer in Maryland and its implications for vertical transport model. Atmos. Environ. 46, 354-364.

Hu, X.M., Klein, P.M., Xue, M., 2013. Evaluation of the updated YSU planetary boundary layer scheme within WRF for wind resource and air quality assessments. J. Geophys. Res. Atmos. 118 (18), 10490-10505.

Hu, X.M., Klein, P.M., Xue, M., Zhang, F., Doughty, D.C., Forkel, R., Joseph, E., Fuentes, J.D., 2013. Impact of the vertical mixing induced by low-level jets on boundary layer ozone concentration. Atmos. Environ. 70, 123-130.

Hua, Y., Wang, S., Wang, J., Jiang, J., Zhang, T., Song, Y., Kang, L., Zhou, W., Cai, R., Wu, D., Fan, S., Wang, T., Tang, X., Wei, Q., Sun, F., Xiao, Z., 2016. Investigating the impact of regional transport on PM2.5 formation using vertical observation during APEC 2014 Summit in Beijing. Atmos. Chem. Phys. 16, 15451–15460.

Huang, R.J., Zhang, Y., Bozzetti, C., Ho, K.F., Cao, J.J., Han, Y., Daellenbach, K.R., Slowik, J.G., Platt, S.M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S.M., Bruns, E.A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., Prevot, A.S., 2014. High secondary aerosol contribution to particulate pollution during haze events in China. Nature. 514 (7521), 218-222.

Jiang, C., Wang, H., Zhao, T., Li, T., Che, H., 2015. Modeling study of PM2.5 pollutant transport across cities in China's Jing–Jin–Ji region during a severe haze episode in December 2013. Atmos. Chem. Phys. 15 (10), 5803-5814.

Li, K., Liao, H., Zhu, J., Moch, J.M., 2016. Implications of RCP emissions on future PM2.5 air quality and

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-512
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 12 June 2018

Atmospheric Chemistry and Physics Discussions

direct radiative forcing over China. J. Geophys. Res. Atmos. 121 (21), 12985-13008.

Li, T., Wang, H., Zhao, T., Xue, M., Wang, Y., Che, H., Jiang, C., 2016. The Impacts of Different PBL Schemes on the Simulation of PM2.5 during Severe Haze Episodes in the Jing-Jin-Ji Region and Its Surroundings in China. Adv. Meteorol. 2016, 1-15.

Miao, Y., Liu, S., Zheng, Y., Wang, S., 2016. Modeling the feedback between aerosol and boundary layer processes: a case study in Beijing, China. Environ. Sci. Pollut R. 23 (4), 3342-3357.

Petäjä, T., Järvi, L., Kerminen, V.M., Ding, A.J., Sun, J.N., Nie, W., Kujansuu, J., Virkkula, A., Yang, X.Q., Fu, C.B., Zilitinkevich, S., Kulmala, M., 2016. Enhanced air pollution via aerosol-boundary layer feedback in China. Sci. Rep. 6, 18998.

Qiu, Y., Liao, H., Zhang, R., Hu, J., 2017. Simulated impacts of direct radiative effects of scattering and absorbing aerosols on surface layer aerosol concentrations in China during a heavily polluted event in February 2014. J. Geophys. Res. Atmos. 122 (11), 5955-5975.

Quan, J., Tie, X., Zhang, Q., Liu, Q., Li, X., Gao, Y., Zhao, D., 2014. Characteristics of heavy aerosol pollution during the 2012–2013 winter in Beijing, China. Atmos. Environ. 88, 83-89.

Sun, Y., Jiang, Q., Wang, Z., Fu, P., Li, J., Yang, T., Yin, Y., 2014. Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013. J. Geophys. Res. Atmos. 119 (7), 4380-4398.

Wang, H., Gong, S., Zhang, H., Chen, Y., Shen, X., Chen, D., Xue, J., Shen, Y., Wu, X., Jin, Z., 2009. A new-generation sand and dust storm forecasting system GRAPES_CUACE/Dust: Model development, verification and numerical simulation. Chinese Sci. Bull. 55 (7), 635-649.

Wang, H., Zhang, X., Gong, S., Chen, Y., Shi, G., Li, W., 2010. Radiative feedback of dust aerosols on the East Asian dust storms. J. Geophys. Res. 115 , D23214.

Wang, H., Tan, S.C., Wang, Y., Jiang, C., Shi, G.Y., Zhang, M.X., Che, H.Z., 2014a. A multi sources observation study of the severe prolonged regional haze episode over eastern China in January 2013. Atmos. Environ. 89, 807-815.

Wang, H., Xu, J., Zhang, M., Yang, Y., Shen, X., Wang, Y., Chen, D., Guo, J., 2014b. A study of the meteorological causes of a prolonged and severe haze episode in January 2013 over central-eastern

China. Atmos. Environ. 98, 146-157.

Wang, H., Xue, M., Zhang, X.Y., Liu, H.L., Zhou, C.H., Tan, S.C., Che, H.Z., Chen, B., Li, T., 2015a. Mesoscale modeling study of the interactions between aerosols and PBL meteorology during a haze episode in Jing–Jin–Ji (China) and its nearby surrounding region – Part 1: Aerosol distributions and meteorological features. Atmos. Chem. Phys. 15 (6), 3257-3275.

Wang, H., Shi, G.Y., Zhang, X.Y., Gong, S.L., Tan, S.C., Chen, B., Che, H.Z., Li, T., 2015b. Mesoscale modelling study of the interactions between aerosols and PBL meteorology during a haze episode in China Jing–Jin–Ji and its near surrounding region - Part 2: Aerosols' radiative feedback effects. Atmos. Chem. Phys. 15 (6), 3277-3287.

Wang, J., Wang, S., Jiang, J., Ding, A., Zheng, M., Zhao, B., Wong, D.C., Zhou, W., Zheng, G., Wang, L., Pleim, J.E., Hao, J., 2014. Impact of aerosol–meteorology interactions on fine particle pollution during China's severe haze episode in January 2013. Environ. Res. Lett. 9 (9), 094002.

Wang, Y., Zhang, Q.Q., He, K., Zhang, Q., Chai, L., 2013. Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia. Atmos. Chem. Phys. 13 (5), 2635-2652.

Wang, Z., Li, J., Wang, Z., Yang, W., Tang, X., Ge, B., Yan, P., Zhu, L., Chen, X., Chen, H., Wand, W., Li, J., Liu, B., Wang, X., Wand, W., Zhao, Y., Lu, N., Su, D., 2013. Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control. Sci. China Earth Sci. 57 (1), 3-13.

Yang, Y., Liao, H., Lou, S., 2016. Increase in winter haze over eastern China in recent decades: Roles of variations in meteorological parameters and anthropogenic emissions. J. Geophys. Res. Atmos. 121 (21), 13050-13065.

Yang, Y.R., Liu, X.G., Qu, Y., An, J.L., Jiang, R., Zhang, Y.H., Sun, Y.L., Wu, Z.J., Zhang, F., Xu, W.Q., Ma, Q.X., 2015. Characteristics and formation mechanism of continuous hazes in China: a case study during the autumn of 2014 in the North China Plain. Atmos. Chem. Phys. 15 (14), 8165-8178.

Zhang, M., H. Wang, X. Y. Zhang, et al., 2018. Applying the WRF double-moment six-class microphysics scheme in the GRAPES_Meso model: A case study. J. Meteor. Res., 32(2), 246–264, doi:

10.1007/s13351018-7066-1.

Zhang, R.H., Li, Q., Zhang, R., 2013. Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013. Sci. China Earth Sci. 57 (1), 26-35.

Zheng, G., Duan, F., Ma, Y., Zhang, Q., Huang, T., Kimoto, T., Cheng, Y., Su, H., He, K., 2016. Episode-Based Evolution Pattern Analysis of Haze Pollution: Method Development and Results from Beijing, China. Environ. Sci. Technol. 50 (9), 4632-4641.

Zheng, G.J., Duan, F.K., Su, H., Ma, Y.L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y.F., He, K.B., 2015. Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions. Atmos. Chem. Phys. 15 (6), 2969-2983.

Zhong, J., Zhang, X., Wang, Y., Sun, J., Zhang, Y., Wang, J., Tan, K., Shen, X., Che, H., Zhang, L., Zhang, Z., Qi, X., Zhao, H., Ren, S., Li, Y., 2017. Relative contributions of boundary-layer meteorological factors to the explosive growth of PM2.5 during the red-alert heavy pollution episodes in Beijing in December 2016. J. Meteorol. Res. 31 (5), 809-819.

Zhong, J.T., Zhang, X.Y., Dong, Y.S., Wang, Y.Q., Liu, C., Wang, J.Z., Zhang, Y.M., Che, H.C., 2018. Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of PM2.5 during winter heavy pollution episodes in Beijing from 2013 to 2016. Atmos. Chem. Phys. 18, 247–258.

Zhou, C.H., Gong, S., Zhang, X.Y., Liu, H.L., Xue, M., Cao, G.L., An, X.Q., Che, H.Z., Zhang, Y.M., Niu, T., 2012. Towards the improvements of simulating the chemical and optical properties of Chinese aerosols using an online coupled model – CUACE/Aero. Tellus B. 64 (1), 91-102.

Zhou, C., Zhang, X., Gong, S., Wang, Y., Xue, M., 2016. Improving aerosol interaction with clouds and precipitation in a regional chemical weather modeling system. Atmos. Chem. Phys. 16 (1), 145-160.

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Table 1 Experiments Design

EXP_bk	Background model experiment: ignoring aerosol radiation feedback, normal turbulent diffusion of chemical tracers
EXP_td_af	Model run with aerosols radiation feedback online, normal turbulent diffusion of chemical tracers
EXP_td20_af	Model run with aerosols radiation feedback online, retaining 20% (reducing 80%) of normal turbulent diffusion of chemical tracers, representing a supposed compensation for the deficient description of extreme weak turbulent diffusion by PBL scheme

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Figure

Fig.1 Model domain (a), cities locations and the topography features in Jing-Jin-Ji (b)

Fig.2 Observed $PM_{2.5}$ (OBS_$PM_{2.5}$) and simulated $PM_{2.5}$ (μg/m^3) of EGS by Exp_bk ($PM_{2.5}$_bk) EXP_td_af ($PM_{2.5}$_ td_af), and EXP_td20_tf ($PM_{2.5}$_ td20_af).

Fig.3 Variation of temperature profiles due to aerosol radiation (K) from 15 to 20 December, 2016.

Fig.4 Sounding observed and modeled temperature profiles by EXP_bk and EXP_af_td during CS (a) and EGS (b) in Beijing and Xingtai.

Fig.5 Hourly changing of $PM_{2.5}$_OBS, $PM_{2.5}$_bk, $PM_{2.5}$_td_af, and $PM_{2.5}$_td20_tf (μg/m^3), together with the turbulent diffusion coefficient (DC_bk, DC_ td_af, and DC_ td20_af) of the three experiments from 15 to 22 December, 2016 in Beijing (a) and Xingtai (b)

Fig.6 The diagrammatic sketch of the contributions to the $PM_{2.5}$ EG due to ARF and DTD

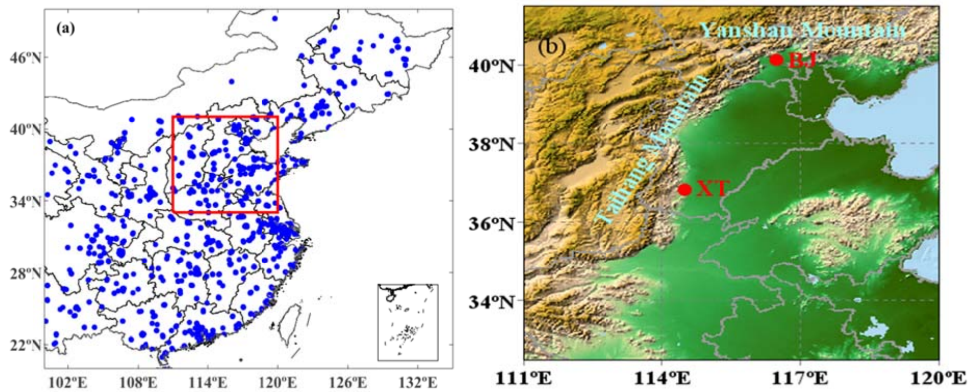

Fig. 1 Model domain (a), cities locations and the topography features in Jing-Jin-Ji (b)

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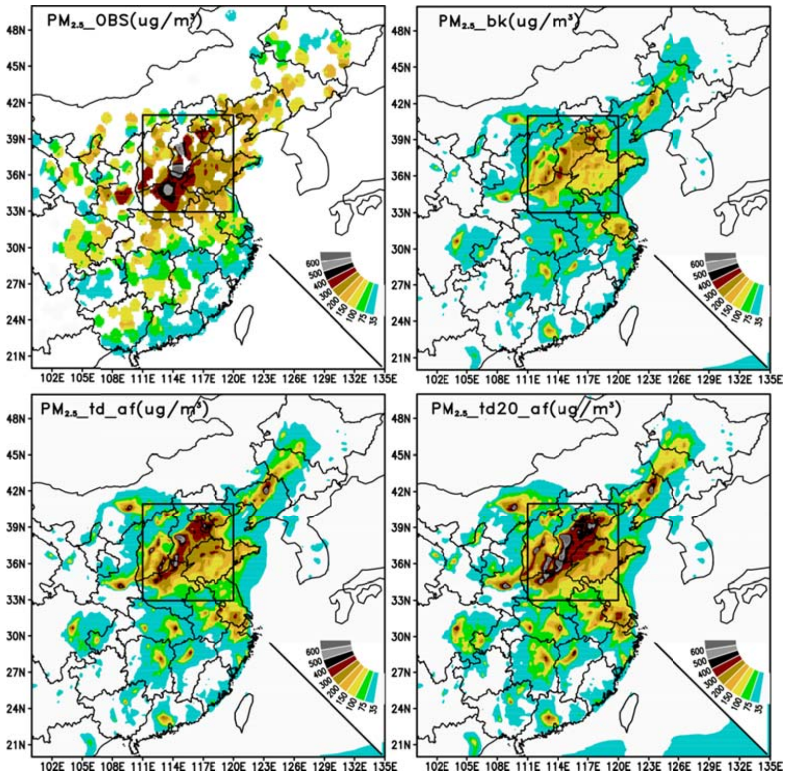

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Discussion started: 12 June 2018

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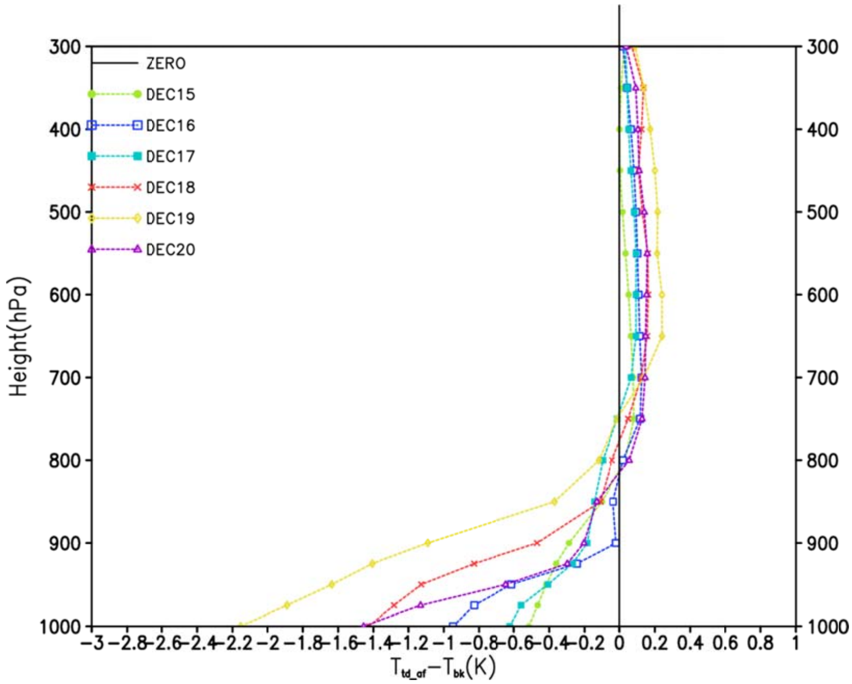

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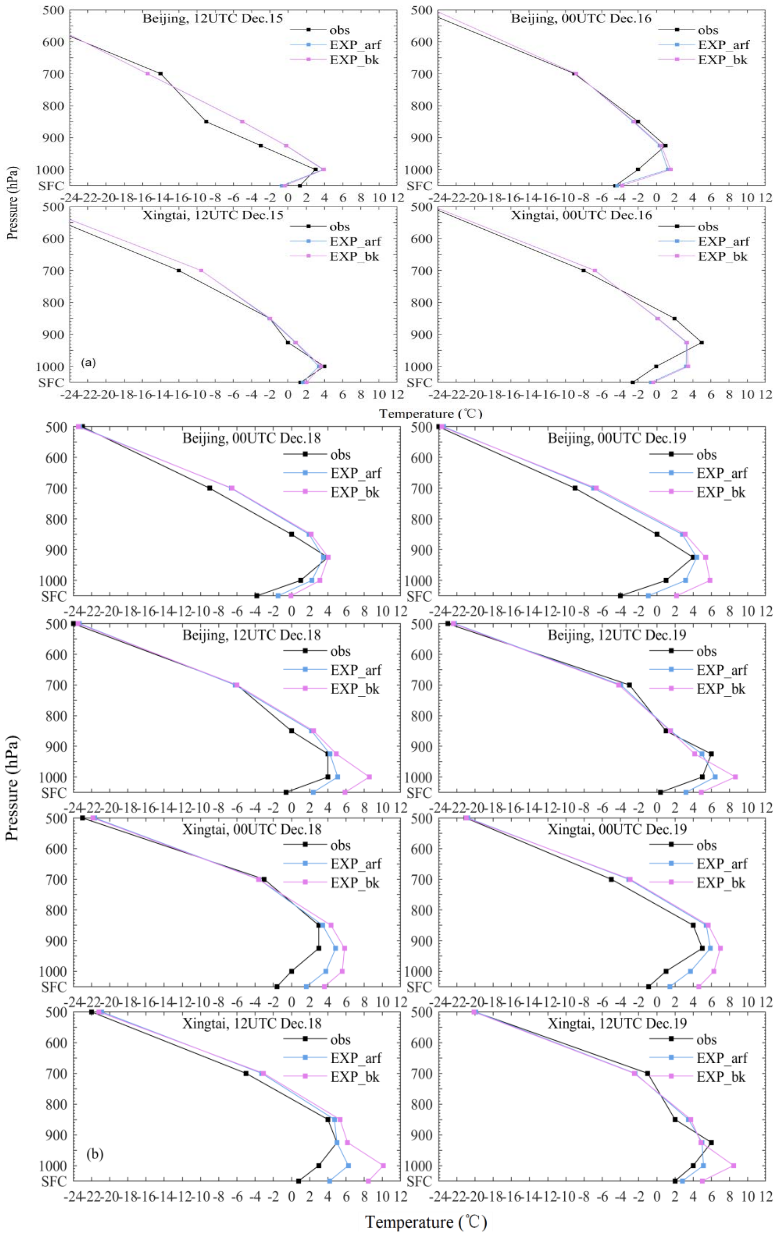
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 12 June 2018



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Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 12 June 2018

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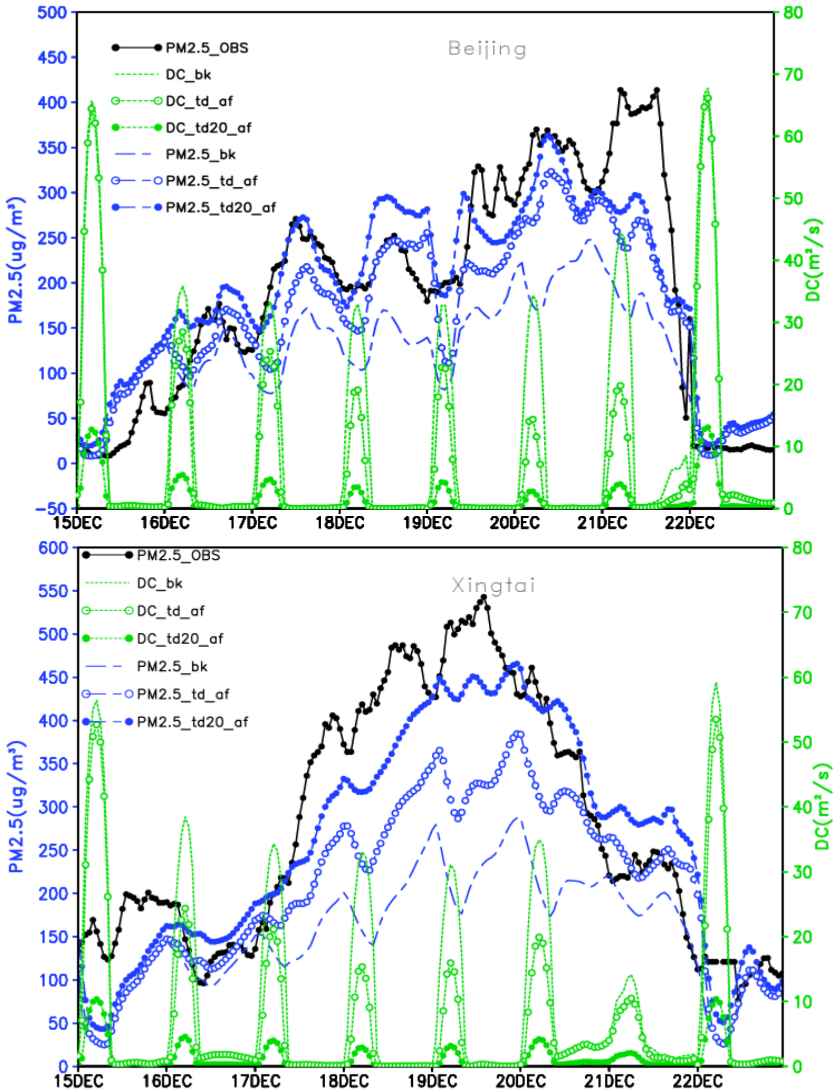

Fig.5 Hourly changing of Observed and modeled $PM_{2.5}$ (μg/m3) of Exp_bk, EXP_td_af, and EXP_td20_tf, together with the turbulent diffusion coefficient (DC) of the three experiments from 15 to 22 December, 2016 in Beijing (a) and Xingtai (b)

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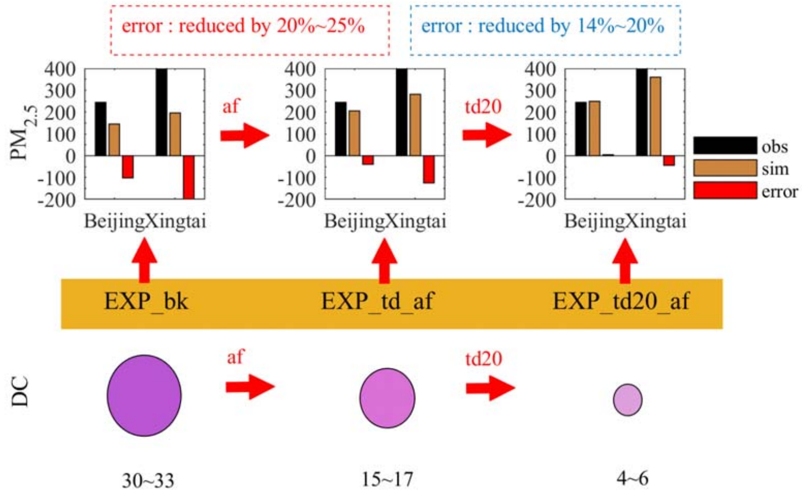

Fig.6 The diagrammatic sketch of the contributions to the $PM_{2.5}$ EG due to AF and DTD