Comments to the Author:

- 2 Although the authors have addressed the major scientific issues raised by the referees, the paper
- 3 is technically not yet ready to be accepted for publication. The language of the papers requires
- 4 major improvements, and should be finally checked out by a native English speaker. My specific
- 5 comments in this regard are the following:

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- First, the use of tense, articles and prepositions needs to be checked out and corrected throughout the paper.
- 9 Response:
- 10 Thank you for pointing out the language problems in this paper and giving us the opportunity to
- 11 correct them. We have asked professional native speaker "LucidPapers" to overall improve the
- 12 English of the paper, and after that we proved their corrections in scientific meaning. Please see
- 13 the edited and clean versions of this paper.

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- 15 Second, there are a number of sentences that are either too long or complicated, or unclear
- 16 parts, which makes them difficult for the readers. Especially, such sentences can be found on
- 17 lines 92-93, 107-110, 127-131, 138-141, 164-166 (one cannot say was observed reduction
- 18 89%), 180-183 (check out how to write dates), 188-192, 205-209, 234-236, 245-251, 263-268,
- 19 285-288, 300-302, 307-308, 320-322, 332-334, 341-343, 348-349, 371-373, 383-384,
- **20** 398-402, 418-420.
- 21 Response:
- 22 The English of the paper has been polished overall including all the sentences on these lines.
- 23 Third, the paper needs to list proper scientific aim(s). What is written on lines 79-82
- 24 does not motivate to read this paper in more detail.
- 25 Response:
- 26 The related context in rewritten in line 92-97 in the edited paper.

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28 Other comments:

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- 30 There should be a space between the number and unit when given values for quantities
- 31 Response:
- 32 This has been corrected in the revised paper.
- 33 Line 121: ... Hong's studies: papers cannot be cited like this
- 34 Response:
- 35 This has been revised as "The turbulent diffusion coefficient (DC) is calculated by the YonSei
- University PBL scheme (Hong et al., 2006)" on line 135 in the edited version.

- 38 I am not sure the authors use correctly the term "trend" on page 10 and later in the text.
- 39 Response:
- 40 All uses of "trend" (line 151,254,257,264,290,440 in the edited version of this paper) have been
- 41 examined and corrected.

- 42 Line 231: should it be ...overprediction of the temperature ... rather than ... temperature positive
- 43 errors
- 44 Response:
- 45 This has been corrected as "overestimation of surface temperature (line 270 in the edited version
- 46 of this paper)
- 47 Line 259: PM2.5 center of... does not sound a right way to express this
- 48 Response:
- 49 This phrase has been revised "The most polluted area with PM2.5 values of 500–700 μg/m3 ···"
- on line "305-306" in the edited paper.
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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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- The Contributions to the Explosive Growth of PM_{2.5} Mass due
- to Aerosols-Radiation Feedback and Further Decrease in
- 54 Turbulent Diffusion during a Red-alert Heavy Haze in
- 55 Jing-Jin-Ji in China
- Hong Wang^{1,2*}, Yue Peng^{1,2}, Xiaoye Zhang^{1,3*}, Hongli Liu¹, Meng Zhang⁴, Huizheng
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Abstract. The explosive growth (EG) of PM_{2.5} mass usually resulted results in PM_{2.5} extreme PM_{2.5} levels and severe haze pollution in eEast China, and they wereis generally underestimated by current atmospheric chemical-chemistry models. Based on the atmospheric chemical one such model, GRPAES_CUACE, three sensitive experiments — of a "background" experiment (EXP1), "online aerosols feedback" online experiment (EXP2), and an "decrease 80% decrease in turbulent diffusion coefficient" (DTD) of chemical tracers" experiment, based on EXP2 (EXP3) — are were designed to study the contributions contributions of to the EG of PM_{2.5} due to aerosols—radiation feedback (AF) and DTD to the explosive growth of PM_{2.5} focusing onduring a "red-alert" heavy haze event in China's Jing—Jin—Ji region in China. The study-results showed that the turbulent diffusion coefficient (DC) calculated by EXP1 is was about 60—70 m²/s on the clear day and 30—35 m²/s on the haze day. This difference of in DC was not enough to discriminate distinguish between the unstable atmosphere on the clear day and extremely stable atmosphere during the EG stage of PM_{2.5} explosive growth stage, and Also, the inversion calculated by EXP1 was obviously weaker than the actual inversion from sounding observations on the haze day. This led to a 40%—51% underestimation of PM_{2.5} by EXP1; AF reduced by about 43%—57% DC during EG stage of the PM_{2.5}

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explosive growth stage, which strengthened the local inversion obviously; and plus, the local inversion indicated by EXP2 was much closer to the sounding observations than that by EXP1. This resulted in a 20%-25% reduction of model negative errors of PM_{2.5} negative errors in the model, and it was reaching as low as -_16% to -_11% in EXP2. However, the inversion produced by EXP2 was still weaker than the actual observation, and AF could not solve all the problems of PM_{2.5} underestimation. Based on EXP2, the 80% DTD of chemical tracers in EXP3 resulted in a-near-zero turbulent diffusion, named-referred to as an "turbulent intermittent intermittence" atmosphere atmospheric state, which resulted in a further 14%-20% reduction of in $PM_{2.5}$ underestimation, and the negative $PM_{2.5}$ errors of waswere reduced to -11% to 2%. The combined effects of AF and DTD solved over 79% of the underestimation of the explosive growth of PM_{2.5} EG in this study. The results show that the online calculation of aerosol-radiation feedback AF is essential for the prediction of PM_{2.5} explosive growthEG and peaks during severe haze in China's Jing--Jin_Ji in Chinaregion. Besides this, an improvement inimproving the arithmetic-planetary boundary layer of PBL scheme calculation focusing on with respect to extremely stable atmosphere atmospheric stratification is also indispensable essential, for a reasonable description of local "turbulent intermittentintermittence" and a more accurate prediction of PM2.5 EG explosive growth during the severe haze in Jing Jin Ji in this region of China.

Keywords: aAerosol_s Rradiation Freedback; Turbulent Deliffusion; PBL planetary boundary layer

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Sscheme; Ttemperature Linversion; PM_{2.5}

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1 Introduction

Since 2013, East eChina has been experienced experiencing unprecedented intrusions of severe hazes accompanied by high levels of particulate matter (PM) of less than 2.5 microns in aerodynamic diameter (PM_{2.5}), caused causing 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., 2016; Yang et al., 2015; Zhong et al., 2017, 2018a, 2018b). The Linstantaneous PM_{2.5} concentration is usually reached in the hundreds of, or even one thousand ug/m³ during severe haze episodes, occasionally exceeding one thousand, in the metropolitans in region of Beijing (JING), Tianjin (JIN), Hebei province (alias JI), referred to here as Jing-Jin-Ji, and their nearits surroundings of East Shanxi, West Shandong, and North Henan in eEast 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 have showed shown, however, that models generally underestimated the explosive growth (EG) and peak values of PM_{2.5} during the severe hazes, especially in Jing-Jin-Ji-region (Wang et al., 2013; Wang et al., 2014; Li et al., 2016).

The causes of PM_{2.5} EG explosive growth and its underestimation by atmosphere atmospheric chemical chemistry models are complex and uncertain at present, which maybut possibly involves in local emissions, reginal transportation, aerosol physicochemical processes, gas—es particles conversion, meteorology—meteorological conditions, and so on. However, the actual atmospheric stability and how accurate it is described by atmospheric models is a fundamental problem that ean't—cannot—be ignored among others. Local or regional meteorology—meteorological conditions dictates whether the haze occurs and what the PM_{2.5} level may be (Zhang et al., 2014; Zheng et al., 2015; Gao et al., 2016) when source emissions are unchanged for a short period of time. The meteorology—meteorological conditions of the planetary boundary layer (PBL) is are the a key and direct trigger for touching offthe emergence of 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. Also, Lit is also a major way—pathway of particles and gascous pollutants exchanging exchange from the surface to upper atmosphere; and when haze occurs, and further cleaned pollutant dispersal by-via the upper-level winds can take place when haze occurs is accompanied by calm surface winds and weak vertical motion of air in surface layers and the PBL. The intensity of turbulent diffusion largely determines the severity of haze pollution. Thus, a Rreasonable

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description of turbulent diffusion by PBL schemes in atmospheric chemical chemistry models is determinant vital for the prediction of severe pollution prediction (Hong et al., 2006; Wang et al., 2015; Hu et al., 2012, 2013a, 2013b; Li et al., 2016). The latest studies in this field of research showed (Wang et al., 2015; Li et al., 2016) that current PBL schemes may be insufficient enough for describing the extremely weak turbulent diffusion conditions when extremely severe hazes occurs in Jing_Jin_Ji, which more broadly may be one important reason for the underestimation of why PM_{2.5} peaks are underestimated by atmospheric chemical chemistry models. There More specifically, there may be two independent reasons resulting in this deficiency why the description of extremely weak turbulent diffusion in atmospheric models is deficient. One is that aerosol—s-radiation feedback (AF) is not calculated online in the model run. AF may restrain turbulence by cooling the surface and PBL while heating the atmosphere above it when aerosols with certain absorption characteristics are concentrated in the PBL (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., 2016; Petaja et al., 2016; Gao et al., 2017; Qiu et al., 2017; Zhong et al., 2018). Ignoring AF is likely to lead to an obvious overestimation of turbulent diffusion when the PM_{2.5} concentration exceeds a certain value, which is worthy of further study. Another The other possible reason is that the extremely weak turbulence resulting in extremely severe hazes is not fully described by the atmospheric ehemistry model (Li et al., 2016).

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In the present work, —Aa "Rred-alert" Hheavy Hhaze event (issued by China's China's Ministry of Environmental Protection issues air quality red alert when the air pollution index is forecasted to exceeding 300 in over the next three days) that occurred on during 15—to 23 December, 2016 in China's Jing—Jin—Ji in Chinaregion was selected to study the contributions—contributing factors to PM_{2.5} EG—explosive growth and peaks—during severe haze due to AF₂ and the possible deficiency of atmospheric models in description describing of the extremely weak turbulent diffusion of atmosphere models in this study.

2 Model, **D**data and **Methodology**methods

2.1 GRAPES_CUACE Model

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Focusing on dust and haze pollutions in China and East Asia, the Chinese Unified Atmospheric Chemistry Environment (CUACE) (Gong and Zhang, 2008) was online—integrated into the mesoscale

version of the Global/Regional Assimilation and PrEdiction System (GRAPES meso), developed by the Chinese Academy of Meteorological Sciences (Chen et. al., 2008; Zhang and Shen, 2008), to build an online chemical weather forecasting model, GRAPES_CUACE (Wang et al., 2009, 2010; 2015a; Zhou et al., 2012). The main components of GRAPES_CAUCE include: a model dynamic core; a modularized physics package (Xu et al., 2008); an atmospheric chemistry module, CUCAE, with online coupling of perosols direct and indirect aerosol feedback; and an emissions inventory. The dynamic framework of GRAPES CUACE is The semi-implicit, semi-Lagrangian, fully compressible, and non-hydrostatical dynamic framework is adopted in GRAPES CUACE (Yang et al., 2007, 2008; Chen et al., 2008). A height-based- terrain-following coordinate system was is used, and there are 33 vertical layers form the surface to 30 kilometerskm. The A longitude_latitude grid is adopted in the spatial discretization of ????? and <u>the several</u> horizontal resolution <u>is optionaloptions are available</u>. The <u>physical physics</u> package is optionalcan also be tailored by the user (Xu et al., 2008), and tage because the specific physics and chemistry schemes used in this study. The Ggas-phase chemistry of RAD II (Stockwell et al., 1990), with 63 gaseous species through 21 photo-chemical reactions and 121 gas-phase reactions, is used in this study. The aerosols includes sea salts (SS), sand/dust (SD), black carbon (BC), organic carbon (OC), sulfates (SFs), nitrates (NIs) and ammonium salts (AMs), and aerosole processes involving in-hygroscopic growth, coagulation, nucleation, condensation, dry and wet depositions, scavenging, aerosol activations, and eteso on. The formation of sulfate-SF aerosols and secondary organic aerosols (SOA) from gases, nitrates NIs and ammonium formed through gaseous oxidation, and ISORROPIA (Fountoukis et al., 2007) calculating the thermodynamic equilibrium between nitrates NIs and ammonium and their gas precursors, are considered in CAUCE, which hads been evaluated and introduced in previous studies (Gong and Zhang et al., 2008; Zhou et al., 2008, 2012).

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Based on the modeled aerosols concentrations, vertical profiles of temperature changing change, including direct aerosols direct impacts (DT/dt due to aerosols), is are calculated by the radiation model and fed back online feedback to the model dynamic core in at each grid point in and every time step, which reforms the model temperature field, dynamic process, regional circulation and meteorology meteorological conditions, in turn finally ultimately impacts impacting the aerosols concentration in turn. The external

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mixing of aerosols species [of-SS, SD, BC, OC, SF, NI, and-AM] and particle size bins is are used in the calculation of aerosols radiation feedback AF, which was introduced and evaluated in detail in previous studies (Wang et al., 2009, 2010, 2015a, 2015b). With this double two-way GRAPES_CUACE model, aerosol_s-radiation_PBL_meteorologyical interactions, as well as aerosol_s-cloud_precipitation interactions; and regional pollution and transportation of PM_{2.5} etc., had have been successfully studied (Wang et al., 2010, 2015a, 2015b; Zhou et al., 2012, 2016; Jiang et al., 2015; Zhang et al., 2018).

The turbulent diffusion coefficient (DC) is calculated by the YonSei University (YSU) PBL scheme (Hong et al., 2006), which is a revised vertical diffusion package based on the nonlocal boundary layer vertical diffusion scheme in a Mmedium-Rrange Fforecast model (MRF) model (Hong et al., 1996). The major ingredient of the revision is the inclusion of an explicit treatment of entrainment processes at the top of the PBL₂ comparing compared with the MRF PBL scheme. The specific DC calculation method of DC wasis shown in Hongest studies et al. (?????). This algorithm of DC and has been selected as a standard option for the Medium Rang Forecastin –(MRF) Model models (Caplan et al. 1997; Farfán and Zehnder, 2001; Basu, et al., 2002; Bright and Mullen, 2002; Mass et al., 2002) and as well as the Weather Research and Forecasting (WRF) model (Hong et al., 2006) in the National Centers for Environmental Predictions (NCEP) since its establishment.—

The model horizontal resolution of the model is adopted as here was 0.15° × 0.15°, to match the resolution of the emission source. Considering the impacts of the interregional transport of pollutants, eEast China (100°—140°E, 20°—60°N) (fFigure 1a) was set as the model domain, but our discussion mainly focuses mainly on the most polluted area, Jing—Jin—Ji-region (the red box—frame in fFigure 1a), and—for which fFigure 1b—showsillustrates the features of geographical location and topography topographical of this region features. There are two balloon sounding stations, Xingtai and Beijing (yellow stars in fFigure 1b) in our study area. Xingtai, located in southern Hebei province, at the eastern foot of the Taihang Mountains, and it is influenced by the sinking descending airflow from Taihang the Mmountains in winter, and in recent years has frequently been ranked is the most polluted city and the PM_{2.5} concentrations usually ranked the first in China-in recently years. The topography of Xingtai and the serious haze pollution it experiences are closely related to its is the typical representative of situation on the southern plain of Jing—Jin—Ji. Beijing,

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located next to Tianjin and surrounded by Hebei, lies in the transitional zone from the Yan Mountains to its southern plain, and next to Tianjin and surrounded by Hebei, representings the most polluted areas in the central part of Jing—Jin—Ji.

2.2 Emissions Linventory

Based on the Multi-resolution Emissions Inventory for ChinaMEIC emission inventory in 2012 (He et al., 2012), the changes of in East China of 5-five kinds of emission sources of industrial, domestic, agricultural, natural, and traffic are were obtained from the national statistics statistical data of China national with respect to industry factories, energy consumption, road networks, and motor vehicles, are and updated to 2015 to and 2016 in east China. Five reactive gases, i.e. (SO₂, NO, NO₂, CO, NH₃), 20 volatile organic compounds [VOCs₇ (i.e. ALD, CH₄, CSL, ETH, HC₃, HC₅, HC₈, HCHO, ISOP, KET, NR, OL₂, OLE, OLI, OLT, ORA₂, PAR, TERPB, TOL, XYL). (VOCs species listed in tTable 2)], and 5-five aerosols species, i.e. (black carbonBC, organic carbonOC, sulfateSF, nitrate NI and fugitive dust), are were obtained by via the above emissions data according to the input requirement of the CUACE model. The horizontal grid resolution is was 0.15° × 0.15° and there is was one emissions data-set for each month with at hourly intervals.

2.3 Data-Used

Hourly observational PM_{2.5} concentration data for more than 1440 surface observational stations (blue dots in Figure 1) from the China National Environmental Monitoring Centre—(CNEMC) (http://www.cnemc.cn) from during 15_to-23 December 2016 were used to evaluate the model results. The hourly observational meteorology meteorological data, including wind speed and temperature, from 500 surface automatic observation stations in of the China Meteorology Meteorological Administration (CMA) in the Jing—Jin—Ji region (red triangle in Figure 1b), were used to for model validation. The mMeteorological balloon sounding data from the CMA at 0000 UTC (early morning) and 1200 UTC (and dusk, in local time) in Beijing and Xingtai (yellow star in Figure 1b) from CMA during the same period were also used to compare with the modeled results. There are is one AERONET station (Holben et al., 1998), Xianghe, and two CARSNET stations (Che et al., 2009; 2014; 2015), Beijing and Shijiazhuang, in the Jing—Jin—Ji region (black crosses in Figure 1b). Observed aerosols optical depth (AOD) and single

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scattering albedo (SSA) date data from the set three stations at during the same time period were also used to for model evaluation. NCEP 0.25° × 0.25° global analysis grids gridded data (https://rda.ucar.edu/datasets/ds083.3) were used as the model's initial and every 6 hours ix-hourly lateral boundary meteorology meteorological input fields. The initial values of chemical tracers were obtained according to their five-year mean climatic values. The results of the first 120 hours of the model start are split outwere discarded to eliminate the effects of the chemical initial fields.

2.4 Experiments Experimental Designdesign

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Both dynamic processes of the regional atmosphere and solar radiation have important impacts on turbulence-turbulent diffusion and PBL processes. When severe haze occurred occurs, it was has been observed showed from the observation study (Zhong et al., 2018) that the surface-level daily direct radiant radiative exposure was observed reductionis reduced by around 89% comparing compared with that on clean days (Zhong et al., 2018), suggesting the possible possibility of a huge difference of in turbulence turbulent diffusion between severe haze and clean days. Ht However, it is difficult to distinguish between the two reasons leading to the for extremely weak turbulence turbulent diffusion in the truth true atmosphere. because of the complicated relationship between atmosphere atmospheric dynamics and solar radiation. However, some meaningful research results could might be expected possible by conducting sensitive sensitivity experiments using an atmosphere atmospheric ehemical chemistry model. Three-Here, three ensitive such experiments (of EXP1, EXP2, and EXP3 - see Table 3 for descriptions) are were designed to discuss the contributions contributing factors to the extremely weak turbulence and corresponding PM_{2.5} EG explosive growthdue to AF, and along with the insufficient description on theof extremely weak turbulent diffusion by PBL schemes in atmospheric chemical chemistry models. (Descriptions of the three experiments listed in Table 3). All other model dynamic processes, physical options, and initial input data of the meteorology and chemical tracers are were same for the three experiments, i.e., except for the differences shown in Table 3. In the sensitive test in EXP3, a further decrease in the turbulence turbulent diffusion coefficient (DTD-) based on EXP2 was only applied to the DC of chemical tracers in CUACE mode; and the DC in other physical packages and the dynamic framework of GRAPES MESO was the same with thatas in EXP1 and EXP2.

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3 Results and Ddiscussions-

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This The studied haze episode began on 15 December; 2016. PM_{2.5} began to gather and climb slowly at this time, but it—was below 150 ug/m³ in most of Jing—Jin—Ji region—from 00:00 UTC on 15 to 00:00 UTC on 17 December; and we name this a period we refer to as the "climbing stage"—(CS) of PM_{2.5.5} From 00:00 UTC on 17 to 00:00 UTC 21 December, PM_{2.5} increased rapidly, and reached reaching the PM_{2.5} peaks of 400—600 ug/m³ in most of the study area. This We refer to this period is named as the "explosive growth (EG)—stage" of PM_{2.5}. This—In this section, we focus mainly focuses—on the contributions contributions of to the PM_{2.5} EG due to AF and further DTD to the PM_{2.5} during this stage.

3.1 The sSynoptic background of the haze episode

The upper atmosphere circulation in the upper atmosphere and the surface-level synoptic system controlling Jing_Jin_Ji region remained relatively stable during the haze maintenance of this haze episode. Figure 2 displays the Ggeopotential height (GPH), temperature (Temp), and Wwinds fields at in the high upper (500 hPa), middle (700 hPa), and lower atmosphere (850 hPa) atmosphere, and as well as PBL levels (900, 950, 1000 hPa), on at 0000 UTC; 19 December, 2016, as the typical representativeto showing the weather meteorological background of this haze event. It is can be seen that GPH the geopotential height in the upper atmosphere (500 hPa) showed zonal circulation in East Asia. There was a horizontal trough north to of Jing_Jin_Ji (black boxframe) in the upper and middle atmosphere (500 and 700_hPa), and Jing Jin Jithe region was controlled by the moderate northwesterly or westerly air flow at the bottom of the trough. The Ttemperature and wind fields at 500 and 700 hPa both showed that cold air in the upper and middle atmosphere was weak. GPH-The 850-hPa geopotential height in 850hPa showed that the subtropical high (SH in figure 2) in the eEast sSea was strong; and also, Jing_Jin_Ji was in the pressure equalization field to the northwest periphery of the subtropical high and the wind was very weak in-at this level due to the blocking of the subtropical high. GPHs at The 900-, 950-, and 100-hPa geopotential heights all showed that Jing_Jin_Ji was located in the pressure equalization field between the "northwest land high" (LH in figure 2) and southeast subtropical high within the whole PBL, and the land high was weaker than the subtropical high. This resulted in a small pressure gradient, weak and thin wind fields, and a stable atmosphere atmospheric situation within the PBL in Jing-Jin-Ji region, which is very helpfulwas conducive

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to the maintenance of the haze episode.

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3.2 The Observation-model Ccomparison study of observation and model results

Not Meteorological factors not only at the surface but also in the PBL meteorology are the key factors in affecting the haze processes haze episode and PM_{2.5} level concentrations (Wang et al., 2014a, 2014b)₅. Unfortunately, however, most numerical models struggle to simulate these aspects but it is well known that surface and PBL meteorology factors are more difficult to be predicted or simulated by most numerical models than those at middle and high atmosphere, which is also the a key point affecting determining the prediction performance of atmospheric chemical chemistry models (Hu et al., 2013a, 2013b; Li et al., 2016).

Using hourly meteorology meteorological data from surface automatic observation stations of the CMA, the surface wind speed and temperature of at Beijing, and Xingtai, and the average in for Jing_Jin_ -Ji, by according to the results of EXP1, EXP2 and EXP3, are were evaluated from for the period 15_to 24 December, 2016 (Figure 3, up). It can be seen that, in Beijing, the modeled surface wind speed by in the three model experiments was in good agreement with the observation, regardless in terms of the changing overall trend, as well as the maximum and the minimum values of wind speed. The observed and modeled wind speed was basically below 2 m/s from during 17_to-21 December (i.e., the EG-explosive growth stage of PM_{2.5}). The Mmodeled wind speed in at Xingtai was slightly worse than those in that at Beijing, but the changing overall trend of wind speedchange was basically consistent with those of observation, and the wind speed was also below 2 m/s during the EG explosive growth stage of PM2.5. The modeled wind speed was to an extent higher than observed to a certain extent at the beginning and ending period in Xingtai. The trend of changing change trend of the modeled average wind speed in-for the Jing_Jin_Ji region showed reasonable agreement with that of observation and was the closest to the observation observed situation at in the EG-explosive growth stage of PM_{2.5}. The In general, the modeled regional wind speed by model was higher than observation observed in general. The eComparison of the wind speed of among the three model experiments showed that the wind speeds by in EXP2 and EXP3 was were basically same, but to a varying degree both were smaller than in EXP1 in various degree in at Beijing, and Xingtai, and as well as average infor Jing_Jin_Ji as a whole, during EG the explosive growth stage, showing that

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AF decreased the surface wind speed. The trend of temperature changing change trend byaccording to the three model experiments was also consisted consistent with that of the observation, on the whole inat Beijing, Xingtai, and Jing_Jin_Ji as a whole. But However, it also can be seenwas found that the modeled temperature was obviously higher than observationobserved, especially during the EG-explosive growth stage. The temperature by in EXP2 and EXP3 was basically same, but lower than that by in EXP1, which is was much closer to the observation, indicating that AF reduced the positive errors overestimation of surface temperature in Beijing, Xingtai, and average in Jing Jin Ji as a whole. However, it can be seen that the temperature by in EXP2 and EXP3 was also higher than observed during the EG-explosive growth stage, suggesting a role played by that some other uncertainties in the PBL scheme led to the temperature positive errors during EG stage besides AF, which is deserves deserving of more detailed further study in detailthe future. Also shown in Figure 3 are the PBL-mean winds of the three experiments in-for Beijing, Xingtai, and regional average in-Jing_Jin_Ji as a whole were calculated and shown in figure 3 (down). Unfortunately, there are not observational data were available to evaluate them. However, Comparison of the PBL's wind and temperature of according to the three model experiments showed that the PBL-mean wind was basically below 4_m/s while the temperature is was high at in the EG explosive stagestage in at Beijing, Xing-tai, and average in Jing-Jin-Ji as a whole. Similar to the ground surface-level results, the PBL-mean wind speed and temperature by in EXP2 and EXP3 were basically the same, but the wind speed by in these two experiments was obviously lower than that by in EXP1. This indicated that the reduction of in wind speed by AF was more obvious in the PBL than that in at ground level -. Meanwhile, comparison of the surface-level and PBL temperature of the three experiments showed that the cooling effect by of AF is was much stronger at the surface than that in the PBL.

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Aerosols optical properties, including AOD, SSA; and asymmetry factor (ASY), largely determines the aerosols direct radiation radiative effects of aerosols. The observed AOD (Table 4) and SSA (Table 5) in at Shijiazhuang, Beijing and Xianghe are were used to evaluate the modeled results from for the period 15—to 22 December. Because the differences of in the modeled AOD and SSA by results of the EXP1, EXP2 and EXP3 are were small, the results those of EXP1 only are used referred to here. It can be seen that the values of modeled AOD and SSA and their temporal changing trends of change from during 15—to 22

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December were basically consistent with the observation in at Beijing, Shijiazhuang and Xianghe, proving thethus demonstrating good model performance in the terms of its description of aerosols optical properties. Both the observed and modeled SSA in at Shijiazhuang, Beijing, and Xianghe (†Table 5) shows showed that the SSA was obviously higher during the EG explosive growth stage of PM_{2.5} thancompared with that at the beginning or ending stage of the haze on 15—to-16 and 22 December, illustrating that the scattering characteristics of composite aerosols increased obviously when high AOD and PM_{2.5} occurred on severe haze days in the Jing—Jin—Ji region. The accurate description in of AOD and SSA, especially the with respect to the change in SSA changing from clean to haze days, is the basic basis in of the following discussion of on the effects of aerosols effects on PM_{2.5}.

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Figure 4 displays the averaged observed PM_{2.5} (PM_{2.5} OBS) and simulated PM_{2.5} of EXP1* (PM_{2.5}_EXP1), EXP2 (PM_{2.5}_EXP2) and EXP3 (PM_{2.5}_EXP3) experiments—during the EG-explosive growth stage. It can be seen from the PM_{2.5} OBS results that the averaged PM_{2.5} values were generally over exceeded 100_µg/m³ in east China, and Jing_-Jin_-Ji eovered comprised the most polluted areas and with PM_{2.5} reached reaching up to 300_to 400_µg/m³ in parts of Beijing, Tianjin, Middlecentral-south Hebei province, western frontier region of Shandong province, and northern Henan province. The most polluted area with PM2.5 values of 500-700 µg/m3 appeared in southern Hebei and northern Henan provinces and the maximum value of PM2.5 even exceeded 700 µg/m3 in part area in southern Hebei. The PM2.5 eventer of 500_700_ug/m³ appeared in south<u>ern</u> Hebei and North <u>northern</u> Henan province, and <u>with</u> the PM_{2.5} maximum of 700 μg/m³ was found in southern Hebei. The eComparison study of PM_{2.5} EXP1 and PM_{2.5} OBS shows that PM_{2.5} EXP1 is was obviously lower than PM_{2.5} OBS on the whole. It is noteworthy that Notably, EXP1 failed to simulate the PM_{2.5} over 300_µg/m³. PM_{2.5} OBS is-was about approximately 200 to 300 µg/m³ over most of Shandong province, while the PM_{2.5} bk is was only 100 to 200 µg/m³ in this region. Compared with PM_{2.5} EXP1, the PM_{2.5} EXP2 values are were significantly improved by AF, and they arewere much closer to the PM_{2.5} OBS. The Hhigh PM_{2.5} OBS centers of 300_ to 400, 400_to 500; and 500_to 600_µg/m³ are were almost simulated by EXP2, indicating the important effects of AF on in the model simulation simulating such high values of $PM_{2.5}$ high values. However, the simulated areas of of these centers simulated PM_{2.5} values of 300 to 400, 400 to 500, and 500 to 600µg/m³

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arewere still smaller than that those of the PM_{2.5}_OBS. EXP2 also fails failed to simulate the maximum PM_{2.5} values over 600 μg/m³ observed in southern Hebei province. PM_{2.5}_EXP3 just about makes made up for this shortageshortcoming; compared with PM_{2.5}_EXP1 and PM_{2.5}_EXP2, PM_{2.5}_EXP3 is was undoubtedly the closest to PM_{2.5}_OBS both in terms of PM_{2.5} extremes and its the area of influence area. This study result These findings illustrates that both AF and DTD in atmospheric ehemical chemistry models are required for the effective prediction of PM_{2.5} EG-explosive growth during the severe haze in China's Jing—Jin—Ji in Chinaregion.

3.3 The Change in downward solar radiation flux change by aerosols and DTD

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PM in the atmosphere will inevitably lead to the changes of in surface and atmosphere atmosphere solar radiation flux. When severe haze occurs, most PM is concentrated in the atmosphere near the surface and within the PBL; solar radiative flux reaching the ground is reduced greatly, which is the a direct trigger factor for the subsequent changes in thermodynamics, dynamics, and then atmospheric stratification. Any factor leading to the a change of in the atmosphere atmospheric PM loading might result in a change of in the surface downward solar radiation flux (SDSRF). We calculated the percentage changes of in SDSRF (W/m²) between EXP2 and EXP1 [((SDSRF EXP1.)/_SDSRF_EXP1)], and EXP3 and EXP1 Figure 5 shows the mean percentage change of in SDSRF (W/m²) by owing to aerosols (a) and aerosols plus DTD, (b) ofduring EG the explosive growth stage. It can be seen that SDSRF was reduced by more than 50% by aerosols in-over most of the study region, (60%-65% in Jing, Jin, most of Ji, and Nnorthern Shandong, and even 65%-70% in Jing, Jin, and part of Ji), indicating the important influence of aerosols on SDSRF. Comparison of Figures 5b and 5a showed shows that this reduction of in SDSRF by owing to aerosols (Figure 5a) in EXP2 was further strengthened by the DTD of chemical tracers in EXP3 (Figure 5b) in certain regions, because DTD made led to the accumulation of more PM_{2.5} gather near the surface (fFigure 3), less transport less and, subsequently, this led to thean increasing increase of in total PM_{2.5} loading. It can also can be seen that the difference of between ffigures 5a and figure 5b was is not too chnegligible. This is because that the major impacts of DTD is was to reform the vertical distribution of the atmosphere atmospheric loading of PM2.54 and its impacts on the total-column of PM2.5 is not so muchwas minor. On the other hand, the reduction of in SDSRF due-owing to aerosols radiation was already very greatconsiderable, and so the change of in SDSRF due-owing to the increased total-column PM_{2.5} by DTD₇ would not be so great on a secondary basis. This value of the SDSRF reduction due-owing to aerosols and DTD is basically consistent with the 56%—89% difference of observational radiant radiative exposure between clear and haze days at during the same period (Zhong et al., 2018).

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3.4 The Influence of aerosols' reform on the reforming of the local atmosphere atmospheric temperature profile

Offline and online studies indicated the a reforming of the atmosphere atmospheric temperature profile by owing to aerosols the direct effect of aerosol radiation (Wang et al., 2010, 2015b; Forkel et al., 2012; Gao et al., 2014, 2015; Wang et al., 2014; Gao et al., 2017; Ding et al., 2016). In our previous works (Wang et al., 2015a, 2015b), Ccomposite aerosols mixing of black carbonBC, organic carbonOC, sulfateSF, nitrate NI, dust, ammonium, and sea salt aerosols had been was online coupled online coupled into the in GRAPES_CAUCE model. On this basis, in the present study, the changes of in the mean temperature profile of Jing_Jin_Ji region ofduring daytime due-owing to aerosols radiation were calculated from for 15_to-20 December, 2016 in this work. It can be seen from Figure 6 that aerosols cooled the atmosphere below 750_to-800 hPa, while but warmed the atmosphereit above this height. Considering the 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 the whole PBL and its near upper atmosphere was were cooled by aerosols to a different varying extent during the different stages of this haze process. The aerosols' warming effects of aerosols above 750-850 hPa height were very weak, and the temperature changes differences among different days were also small. However, the aerosols' cooling effects of aerosols shows varied the most between different days differences from the surface to 975 hPa height on different day. The For instance, surface daytime cooling is was about 2.2 K on 19 December, 1.5 K on 18 and 20 December, 1 K on 17 December, and 0.5-0.6 K on 15-to-16 December. This aerosols' cooling effect of aerosols decreased rapidly with the height. The difference of in the cooling rates between the surface and 850 hPa is was 1.8 K on 19 December, 1.3 K on 18 and 20 December, 1 K on 17 December, and 0.3_0.4 K on 15 and 16 December. The difference of in the cooling rates by owing to acrosols between

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the surface and the upper PBL are was much bigger during the EG explosive growth stage than that of the CSclimbing stage. This may have resulted in the further intensification of the temperature inversion layer that pre already existed during the haze event, which will be discussed in figure 7 in the following section.

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The vertical sounding-meteorologyical data from the vertical soundings in taken at Beijing and Xingtai can bewere used to prove verify if this change of in the temperature profile by owing to acrosols is correct or not. Figure 7 shows the vertical temperature profiles of the sounding observations and the modeled temperature profiles by of EXP1 and EXP2 during the climbing stageCS (Figure 7a) and EG-explosive growth stage (Figure 7b) at the two stations. The temperature profiles (Figure 7a) shows that both the modeled results by of EXP1 and EXP2 both partly simulated in part the observed temperature inversion in at Beijing and Xingtai on 15_to-16 December. The very littlenegligible difference between the temperature profiles by of EXP1 and EXP2 indicated indicates that acrosols radiation had very little impacts on the temperature profiles and local inversion during the climbing stageCS of PM2.5. Nevertheless, Figure 7b shows that the observed temperature inversions were obviously stronger and thicker on 18_to-19 December (EG-explosive growth stage) than those on 15_to-16 (climbing stageCS of PM2.5), both in Xingtai and Beijing. The temperate profiles by of EXP2 were much closer to the observational results than that bythose of EXP1; and especially, the temperature inversions were much stronger and also closer to the observation than that bythose of EXP1. This result proved proves that the effective correction of local inversions by acrosols during the EG stage of PM2.5 explosive growth stage was effective.

However, it also can also be seen, that the inversions by of EXP2, which included online AF, are were still weaker than the truth observed inversion observed in at the two stations. This suggests that there must be other eauses reasons, besides the online calculation of AF, for the underestimation of the observed extremely strong inversion by the model besides the online calculation of AF, which is worthy of studying further study.

3.5 The contributions Contributions of to PM_{2.5} EG due to AF and DTD to PM_{2.5} explosive growth

Turbulent diffusion process—is the main way process of gas and particles exchange from ground the surface to the upper atmosphere, and then removed the high—altitude transport,

which and one of the key tasks of atmospheric chemistry models is to capture this processusually achieved by turbulent diffusion process in the chemical atmospheric models. Firstly, the inversion and weak turbulent diffusion, which generates from atmosphere atmospheric dynamic processes, leads to atmosphere atmospheric stabilization and determines the occurrence of haze and its strength (Zheng et al., 2016). Once the haze occurs, the aerosols radiation may in turn reinforce the inversion in turn when aerosols exceeds a certain critical value, and leading to more PM_{2.5} gathering near the ground. The relative importance of these two aspects on PM_{2.5} EG explosive growth may vary with the PM_{2.5} values concentrations and meteorology meteorological conditions, but they are irreplaceable for the a reasonable prediction and simulation of PM_{2.5} EG explosive growth and peaks by in atmospheric models.

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Figure 8 displays the hourly changing change of in observed PM_{2.5} (PM_{2.5} OBS) and the modeled PM_{2.5} by of EXP1, EXP2₇ and EXP3-experiments, together with the modeled turbulent DC of the three experiments, in Beijing (Figure_8a) and Xingtai (Figure 8b), from for the period_15_to_23 December. Comparison of the PM_{2.5} modeled by EXP1, EXP2₇ and EXP3 with observation in Beijing (Figure 8a) shows that the modeled PM_{2.5} modeled by EXP3 was the closest to observation during the whole haze episode, which agreed with the results of the regional distribution of the EG explosive growth stage illustrated in Figure 4. EXP1 underestimated the PM_{2.5} obviously from during 17_to_22 December, and this underestimation was even more obvious with the increasing of PM_{2.5}. This difference between the modeled and observed PM_{2.5} was the largest during the EG explosive growth stage of PM_{2.5}. AF shortened reduced this difference to a great considerable extent, and the PM_{2.5} by of EXP2 was much closer to the observation than that by of EXP1 during the EG explosive growth stage of PM_{2.5}. However, it can be seen that there was were certain differences between the observed and modeled PM_{2.5} and that modeled by EXP2, illustrating that AF can't cannot completely fill the big sizeable gap between observed and modeled PM_{2.5}. The PM_{2.5} by of EXP3 shortened reduced this gap further, and showshowing the best agreement with observation, especially during the PM_{2.5} EG explosive growth stage.

It <u>can</u> also <u>ean</u> be seen from <u>fF</u>igure 8a that the DC <u>by of</u> EXP1 was about 30_40 m²/s during the <u>EG</u> explosive growth stage <u>of PM_{2.5}</u>, which was about 50% of the 60_70 m²/s on <u>the</u>-clear days (15 or 22 December). Obviously, <u>the this</u> 50% DC differences between <u>the</u>-clear and severe haze days may <u>not</u>-be

enough-insufficient to discriminate-separate the difference of in turbulent diffusion intensity between the extremely stable atmosphere on haze days and the unstable atmosphere on clear days, which is the an important reason for the underestimation underestimated of PM_{2.5} EG-explosive growth by in EXP1.

Compared with EXP1, the AF in EXP2 led to a notable enhancement of the temperature inversion (Figure 7b), a significant decrease in the turbulent diffusion on of PM_{2.5} during the EG-explosive growth stage, and a low maximum DC at noon by EXP2 (was as low as 14 m²/s on 20 December, which decreased about—a reduction of 50% comparing compared with that by EXP1). The Mmaximum DC at noon by EXP2 on haze days in EXP2 was only about 20% of that on clear days. The maximum DC at noon by in EXP3 was lower than 5 m²/s on 20 December and, at the same time, the PM_{2.5} modeled by EXP3 was further increased and it was also much further closer to the observed PM_{2.5} observation than the PM_{2.5} by of EXP2.

It can be seen from the comparative studyThrough comparison of the temporal changing change between of DC and PM_{2.5} by in EXP1, EXP2₅ and EXP3 in Beijing, that it is clear that the an overestimation of turbulent DC owning to lack the absence of online ealculation calculated of AF, and as well as a deficient description of the extremely stable stratification by in the PBL schemes in of the atmospheric model, can lead to a distinct underestimation of PM_{2.5} EG explosive growth and peaks when severe haze occurs in China's Jing—Jin—Ji in Chinaregion.

The trends of changing change trends of in DC and PM_{2.5} at Xingtai by in the three sensitive experiments in Xingtai (Figure 8b) shows theare similar results withto those in at Beijing. The PM_{2.5} by of EXP3 was also the closest to observation, followed by EXP2, and then EXP1 was the worst, during the whole haze episode. However, during the EG explosive growth stage of PM_{2.5}, the relative contributions on the PM_{2.5} peak values due to of AF and DTD to the PM_{2.5} peak values showed some differences with to those in at Beijing. The contributions to PM_{2.5} peaks due to of DTD to PM_{2.5} peaks were more important than that those by of AF in at Xingtai. Located at in the eastern foothills of the Taihang Mountains, Xingtai is usually affected by the downhill airflow, and tTemperature inversions in this area is form and strengthen easy casily to form and strengthened, leading to stronger inversion, weaker turbulent diffusion, and more stable atmospheric stratification, but However, this kind of inversion and weak turbulent diffusion, derived from the local terrain, is more difficult harder to describe by for PBL schemes in atmospheric chemical

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chemistry models to describe, and likely underestimated ...

Figure 9 shows their a diagrammatic sketch of the contributions to the PM2.5 of EG stage due to of AF and DTD to the PM2.5 of the explosive growth stage according to summarized by the results of at Beijing and Xingtai. It can be seen that the DC by of EXP1 was 30_35 m²/s, DC bywhile that of EXP2 was 15_17 m²/s, means meaning that AF reduces reduced the DC by about 43%_57% DC of EXP1, which led to the rise in simulated PM2.5 from 144 ug/m³ by in EXP1 to 205 ug/m³ by in EXP2 in at Beijing, and from 280 ug/m³ by in EXP1 to 360 ug/m³ in EXP2 in at Xingtai. This means that AF reduced the underestimation of PM2.5 at Beijing and Xingtai by 20% in Beijing and 25% in Xingtai of simulated PM2.5 negative errors, respectively. The DC by of EXP3 was as low as 4_6 m²/s during the EG explosive growth stage of PM2.5, showing demonstrating the joint effects of AF and DTD reduced the DC to less than 4_6 m²/s, near-zero, which we name-refer to it as "turbulent intermittentintermittence" is was the a further increasing increase of in the simulated surface PM2.5 based on EXP2. DTD decreases reduced 14% to 20% the underestimation of simulated PM2.5 by 14% to 20%, and the errors of PM2.5 errors inby EXP3 were reduced to as low as -11% to 2%.

4. Conclusions

Using an_atmospheric ehemical_chemistry_model, GRAPES_CUACE, three experiments (EXP1, EXP2 and EXP3) were designed to study the reason for the explosive growth of PM_{2.5} mass during a "red-alert" heavy haze event that occurred on during 15_to-23 December, 2016 in China's Jing_Jin_Ji in Chinaregion. The contributions to the PM_{2.5} by of AF and DTD to the PM_{2.5} aerosols feedback and a further decrease in turbulent diffusion coefficient of chemical tracers, representing a compensation for the deficient description of extremely weak turbulent diffusion by in the PBL scheme in of the atmospheric models, are were studied by analysing analyzing the changes of in PM_{2.5}, surface downward solar radiation fluxSDSRF, wind speed and temperature, diffusion coefficientDC, and the relationships between among them, of in the three experiments.

The studyResults shows that the diffusion coefficientDC by in EXP1 is was about 60_-70_m²/s on clear days and 30_35_m²/s on haze days. The 50% difference of between the two was not considered enough insufficient to discriminate separate the unstable atmosphere on clear days and the

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atmosphere on severe haze days, comparing compared with the differences of in direct downward solar radiation between clear and haze days, which is was also proved proven indirectly by the weaker inversion calculated byof EXP1 than that of from the actual sounding observations. This led to a 40%-51% underestimation of the PM_{2.5} peaks by in EXP1 during the explosive growth stage of PM_{2.5} explosive growth stage. Online calculation of aerosols radiation feedback AF reduced the surface and PBL wind speed and cooled the surface and PBL atmosphere. The surface daytime cooling due to aerosols radiation was 1.5_2.2 K during the explosive growth stage of PM_{2.5} and 0.5_0.6 K during the climbing stage of PM_{2.5}. The aerosols' cooling effect of aerosols decreased rapidly with the height, and this is was the a major reason for the strengthening of the temperature inversion during the explosive growth stage of PM2.5. The reduced DC by-owing to AF was up to reached 43%-57% during the PM_{2.5} EG-explosive growth stage-of PM_{2.5}. The impacts on PM2.5 due to AF was distinct during the explosive growth stage of PM2.5 while very little during climbing stage of PM2.5 in the model run, indicating a critical value of 150 ug/m3 of PM2.5 leading to an effective AF in online atmospheric chemical model. The local inversion simulated by <u>in</u> EXP2 was strengthened and closer to the actual sounding observation than that by of EXP1. This resulted in a 20% -25% reduction of in the PM_{2.5} underestimation of PM_{2.5} and with PM_{2.5} errors by in EXP2 was being as low as -16 to -11% during the explosive growth stage of PM_{2.5}. The impact on PM_{2.5} owing to AF in the model run was distinct during the explosive growth stage, but minor during the climbing stage, indicating a critical value of 150 ug/m³ of PM_{2.5} leading to an effective AF in online atmospheric chemistry models. 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/m³ of PM_{2.5} leading to an effective AF in online atmospheric chemical model. However, the local inversion simulated by EXP2 was still weaker than the actual observationobserved, and the PM_{2.5} by of EXP2 was still smaller than observationobserved, illustrating that AF could not solve all the PM_{2.5} underestimation problems. In EXP3, the DC Further DTD of particles and gas based on EXP2 resulted in a 14%-20% lessening of the PM_{2.5} underestimation based on EXP2, and the PM_{2.5} errors of EXP3 was were reduced to -11% to 2%.

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chemical chemistry models is are probably insufficient for describing the extremely stable atmosphere

This The present study result illustratedillustrates that the PBL schemes in current atmospheric

resulting in explosive growth of PM_{2.5} and severe haze in <u>China's Jing_Jin_Ji in Chinaregion</u>, which <u>This</u> may involve in—two important reasons: <u>One is</u>—the absence of <u>an</u> online calculation of AF, another is the and/or a deficient description of <u>the</u>—extremely weak turbulent diffusion by <u>the PBL</u> scheme in the atmospheric <u>chemical_chemistry model</u>. Our study suggests that <u>an online calculation of AF</u> and an improvement in <u>the arithmetic_representation</u> of turbulent diffusion in PBL schemes, <u>with a focusing on extremely stable atmosphere atmospheric stratification</u>, in atmospheric <u>chemical_chemistry models</u>, are indispensable for <u>a reasonable description of local "turbulent intermittentintermittence</u>" and <u>an accurate prediction of</u> the explosive growth and peaks of PM_{2.5} of severe haze in <u>China's Jing_Jin_Ji in Chinaregion</u>.—

569	Author Contributions:
570	Hong Wang and Xiaoye Zhang designed the idea and experiments; Hong Wang and Yue Peng carried them
571	out; Hongli Liu prepared the emissions data and introduction; Meng Zhang did certainperformed some of
572	the model rung: Huizheng Che and Yu Zheng processed the AOD and SSA observational data; Yanli
573	Cheng completed *Table 3 and the related introduction.—
574	Acknowledgements
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Table 1. Physics Physical and Cchemistrycal processes in GRAPES_CUACE.

Physics and Chemistry Process	e <u>O</u> ption s	Reference s
Explicit precipitation	WDM6	Lim and Hong , (2010)
Cumulus clouds	KFETA Scheme	Kain, (2004)
Longwave radiation	Goddard	Chou et al., (2001)
Shortwave radiation	Goddard	Chou et al. , (1998)
Surface layer	SFCLAY Scheme	Pleim , (2007)
Planatory Boundary layerPBL	MRF <mark>S</mark> scheme	Hong et al., (1996, 2006)
Land surface	SLAB <u>S</u> cheme	Kusaka et al. , (2001)
Gas-phase chemistry	RADM II	Stockwell et al., (1990)
Aerosol-Scheme	CUACE	Zhou et al. , (2012)
Aerosol <u>Dd</u> irect effect	External Mmixing	Wang et al., (2015)
Aerosol Lindirect effect	CAUCE+WDM6	Zhou et al., (2016)

Table 2. Sensitive Experiments-Design of sensitivity experiments.

Experiments	Description-of model Experiments
EXP1	Background experiment: ignoring aerosols radiation and -conventional DC of
	chemical tracers by PBL scheme in GRAPES_CUACE
EXP2	Sensitive experiment with Online aerosols radiation feedback AF online and
	conventional DC of chemical tracers by PBL scheme in GRAPES_CUACE
EXP3	Sensitive experiment with Online aerosols radiation feedback AF-online, only and
	DC of chemical tracers is set as to 20% of the conventional DC- calculated by
	PBL scheme, representing a supposed compensation for the deficient description
	of extremely weak turbulent diffusion by the PBL scheme; DC in physical and
	dynamic processes was the same with as EXP1

808	Table 3. VOCs in the emissions data.		
809	VOC ₅	Full name	
810	ALD	Acetaldehyde and higher aldehydes	
811	CH4	Methane	
812	CSL	Cresol and other hydroxy substituted aromatics	
813	ETH	Ethane	
814	HC3	Alkanes w/ $2.\frac{7 \times 10^{7} \times 10^{-13}}{10^{2}} > \text{kOH} < 3.\frac{4 \times 104}{10^{2}} \times 10^{-13}$	
815		<u>10</u> ⁼¹²	
816	HC5	Alkanes w/ $3.4 \times 10^{-12} > \text{kOH} < 6.8 \times 108 \times 10^{-12}$	
817		<u>10</u> =12	
818	HC7	10^{-12} w/kOH > 6.8×10^{-12}	
819	НСНО	Formaldehyde	
820	ISOP	Isoprene	
821	KET	Ketones	
822	OL2	Ethene	
823	OLI	Internal olefins	
824	OLT	Terminal olefins	
825	ORA2	Acetic and higher acids	
826	PAR	Paraffin carbon bond	
827	TERPB	Monoterpenes	
828	TOL	Toluene and less reactive aromatics	
829	XYL	Xylene and more reactive aromatics	

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Table 4. Observed and Mmodeled daily AOD (* stands for shortage of observation).

Date	Shi	ijiazhuang	Beijing		Xianghe	
	OBS	MODEL	OBS	MO <u>D</u> EL	OBS	MODEL
15	0.46	0.55	0.07	0.12	0.10	0.15
16	0.62	0.60	0.14	0.18	0.60	0.40
17	1.30	1.10	0.50	0.56	1.33	1.05
18	1.42	1.20	0.69	0.75	0.87	0.97
19	1.26	1.30	0.50	0.86	0.96	0.90
20	*	1.20	1.90	1.70	*	1.50
21	*	0.65	1.76	1.50	1.78	1.60
22	0.18	0.30	0.10	0.20	0.18	0.22

Table 5. Observed and $\underline{\mathsf{Mm}}$ odeled daily SSA (* stands for shortage of observation).

Date	Shi	Shijiazhuang		ng	Xianghe	
	OBS	MODEL	OBS	MO <u>D</u> EL	OBS	MODEL
15	0.83	0.85	0.81	0.83	0.86	0.84
16	0.83	0.85	0.88	0.86	0.92	0.86
17	0.88	0.89	0.88	0.90	0.93	0.90
18	0.87	0.89	0.91	0.92	0.90	0.90
19	0.86	0.91	0.90	0.93	0.92	0.91
20	*	0.90	*	0.93	*	0.92
21	*	0.88	0.93	0.93	*	0.90
22	0.82	0.83	0.84	0.86	0.88	0.84

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Figure captions

批注 [LP32]: See captions below figures for individual edits.

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Fig. 1. (a) Model domain and location of Jing-Jin-Ji. (b) Geographic location and topography of Jing-Jin-
Ji. Blue dots are the locations of PM _{2.5} observations; red triangles are the locations of automatic weather
stations; yellow stars are the two sounding stations; black crosses are the CARSNET and AEROSNET
stations.Fig.1 Model domain and location of Jing Jin Ji (a), Features of geographical location and
topography of Jing-Jin-Ji (b) (blue dots are the locations of PM _{2,5} observation, red triangles stands for the
locations of automatic weather stations, and yellow stars are the two sounding station, black crosses are the
CARSNET and AEROSNET stations)
Fig. 2. Geopotential height (color-shaded; gp10m), temperature (dashed black contours; K) and wind (wind
bars; m/s) in the (a) upper (500 hPa) and (b) middle (700 hPa) atmosphere, and geopotential height and
wind in the (c) lower atmosphere (850 hPa) and (d-f) PBL (900, 950, 1000 hPa), at 0000 UTC 19
December 2016. Fig. 2 GPH (shaded, gp10m), Temp (broken black line, K) and Wind (wind bar, m/s) at
high (500hPa) and middle (700hPa), and GPH and Wind at low atmosphere (850hPa) and PBL levels (900,
950, 1000hPa) on 00 UTC, 19 December, 2016
Fig. 3. Observed and modeled wind speed and temperature at the surface (upper panels), and the PBL-mean
wind speed and temperature (lower panels), from the results of EXP1, EXP2 and EXP3 for Beijing, Xingtai,
and the average for Jing-Jin-Ji as a whole, during 15-24 December 2016. Fig. 3 Observed and modeled
wind speed and temperature at surface (up) and PBL mean wind speed and temperature (down) by EXP1,
EXP2, and EXP3 in Beijing, Xingtai, and average in Jing Jin Ji from 15 to 24 December
Fig. 4. Mean observed (OBS_PM _{2.5}) and modeled PM _{2.5} concentration (μg/m³) of the PM _{2.5} explosive
growth stage, from the results of EXP1, EXP2 and EXP3 (PM25_EXP1, PM25_EXP2 and PM25_EXP3,
respectively). Fig. 4 Mean Observed (OBS_PM _{2.5}) and Modeled PM2.5 concentration (µg/m3) of EG stage
of PM _{2.5} -by EXP1, EXP2, EXP3 (PM _{2.5} -EXP1, PM _{2.5} -EXP2, and PM _{2.5} -EXP3)
Fig. 5. Mean percentage change in SDSRF (W/m²) owing to (a) aerosols and (b) aerosols+DTD during the
explosive growth stage. Fig. 5 The mean percentage change of SDSRF (W/m²) due to aerosol (a) and
aerosol and DTD (b) of EG stage
Fig. 6. Profiles of average temperature changes in Jing-Jin-Ji owing to AF (K) during 15-20 December

2016. Fig. 6 Profiles of the average temperature changes in Jing Jin Ji due to AF (K) from 15 to 20 December, 2016.

Fig. 7. Sounding-observed and modeled temperature profiles in EXP1 and EXP2 during the (a) climbing stage and (b) explosive growth stage in Beijing and Xingtai. Fig. 7 The Sounding observed and modeled temperature profiles by EXP1 and EXP2 during CS (a) and EG stage (b) in Beijing and Xingtai.

Fig. 8. Hourly change of PM_{2.5} OBS, PM_{2.5} EXP1, PM_{2.5} EXP2, and PM_{2.5} EXP3 (μg/m³), together with the DC at 950 hPa of the three experiments (DC EXP1, DC EXP2, DC EXP3) during 15–22 December 2016 in (a) Beijing and (b) Xingtai. Fig.8 Hourly changing of PM_{2.5} OBS, PM_{2.5} EXP1, PM_{2.5} EXP2, and PM_{2.5} EXP3 (μg/m³), together with the turbulent diffusion coefficient at 950hPa of the three experiments (DC EXP1, DC EXP2, DC EXP2) from 15 to 22 December, 2016 in Beijing (a) and Xingtai (b)

Fig. 9. Diagrammatic sketch of the contributions to the PM_{2.5} EG due to AF and DTD

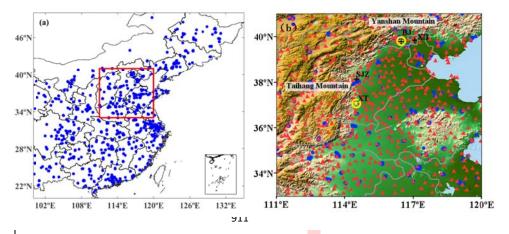


Fig. 1. (a) Model domain and location of Jing Jin Ji (b) Features of gGeographical location and topography of Jing Jin Ji (b). (bBlue dots are the locations of PM_{2.5} observations: red triangles stands

批注 [LP33]: In (b), change the labelling to Taihang Mountains and Yanshan Mountains (i.e., plural).

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for are the locations of automatic weather stations, and yellow stars are the two sounding stations, black crosses are the CARSNET and AEROSNET stations.

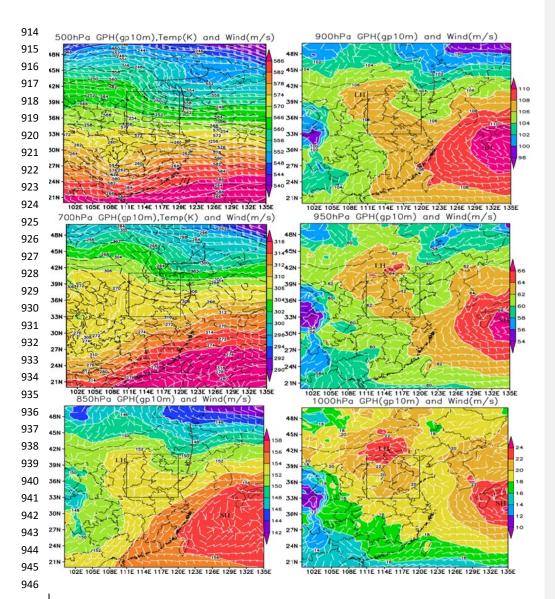


Fig. 2. GPH—Geopotential height (color-shaded; gp10m), Temp—temperature (broken—dashed_black linecontours; K) and Wwind (wind bars; m/s) at in the (a) high-upper (500 hPa) and (b) middle (700 hPa) atmosphere, and GPH-geopotential height and Wwind at in the (c) lower atmosphere (850 hPa) and (d-f) PBL levels (900, 950, 1000 hPa), on-at 0000 UTC; 19 December; 2016.

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批注 [LP34]: Please label the panels (a) to (f).

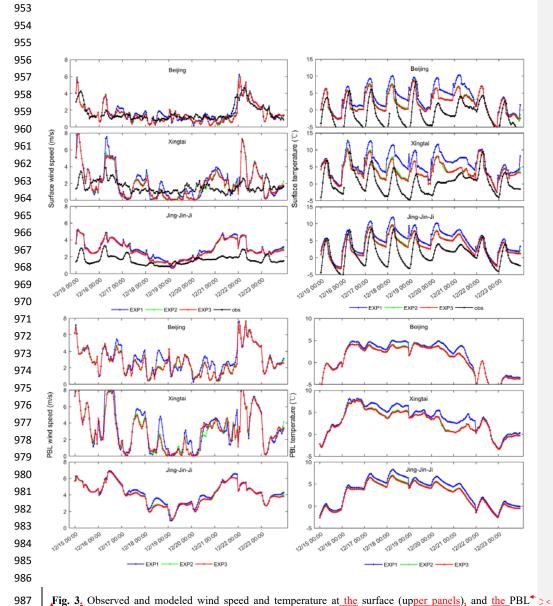


Fig. 3. Observed and modeled wind speed and temperature at the surface (upper panels), and the PBL mean wind speed and temperature (downlower panels), by from the results of EXP1, EXP2, and EXP3 in for Beijing, Xingtai, and the average in for Jing Jin Ji as a whole, from during 15 to 24 December 2016.

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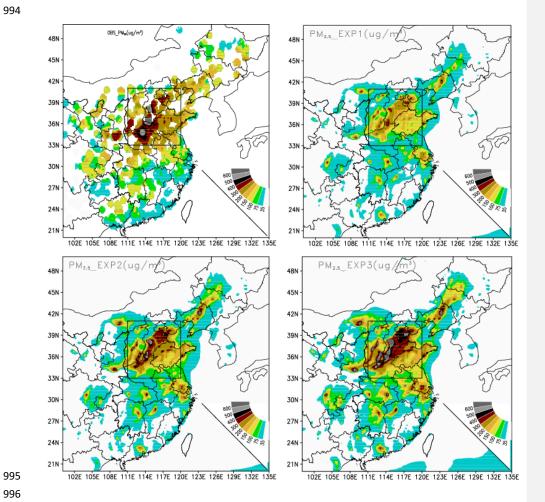


Fig. 4. Mean Oobserved (OBS_PM_{2.5}) and Mmodeled PM_{2.5} concentration (μg/m³) of EG stage of the PM_{2.5} explosive growth stage, from the results of EXP1, EXP2, and EXP3 (PM_{2.5}_EXP1, PM_{2.5}_EXP2, and PM_{2.5}_EXP3, respectively).

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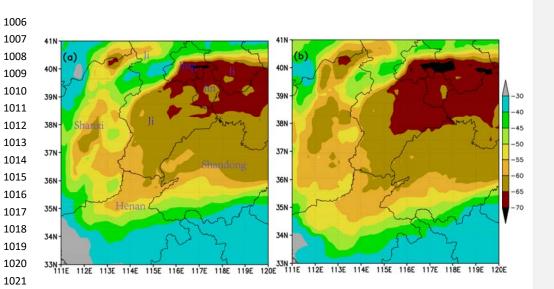


Fig. 5. The mMean percentage change of in SDSRF (W/m²) due owing to (a) aerosols (a) and (b) aerosols and +DTD (b) of during the EG explosive growth stage.

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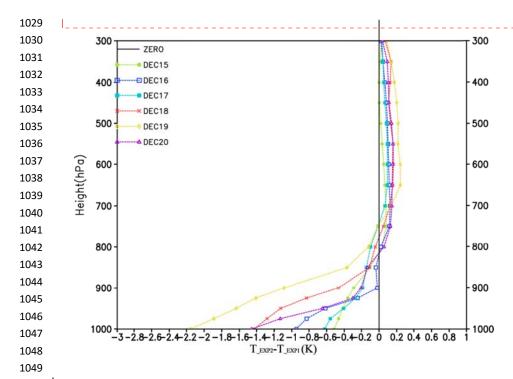


Fig. 6. Profiles of the average temperature changes in Jing_-Jin_-Ji due-owing to AF (K) from during 15_to 20 December, 2016.

批注 [LP35]: Insert a space before the bracketed units in the *y*-axis label.

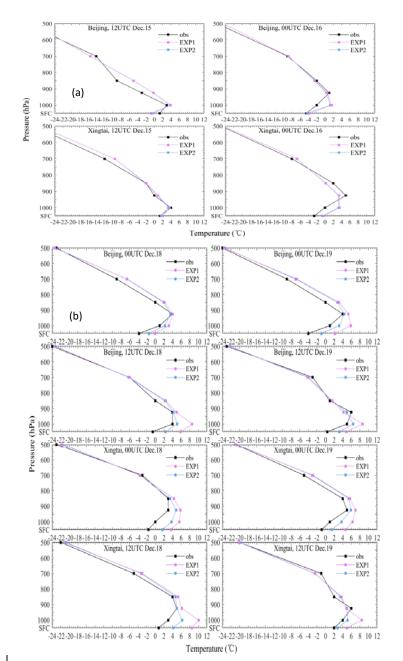
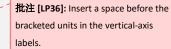


Fig._7. Sounding—observed and modeled temperature profiles by in EXP1 and EXP2 during the (a) climbing stage (b) and (b) EG explosive growth stage (b) in Beijing and Xingtai.



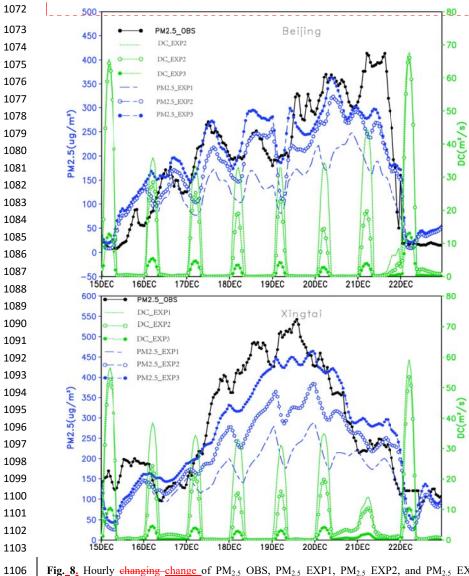


Fig._8. Hourly changing change of PM_{2.5}_OBS, PM_{2.5}_EXP1, PM_{2.5}_EXP2, and PM_{2.5}_EXP3 (μg/m³), together with the turbulent diffusion coefficientDC at 950_hPa of the three experiments (DC_EXP1, DC_EXP2, DC_EXP3) from during 15_to-22 December 2016, 2016 in (a) Beijing (a) and (b) Xingtai (b).

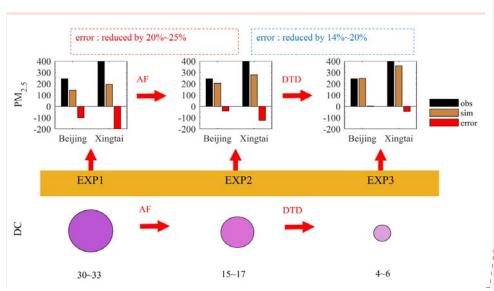


Fig. 9. The dDiagrammatic sketch of the contributions to the PM_{2.5} EG due to of AF and DTD to the PM_{2.5} explosive growth.

批注 [LP37]: Replace the tildes (~) with en dashes (–) in the annotation to indicate a range.