Response to Referee#1

Dear Referee,

Thanks for giving us an opportunity to revise our manuscript (acp-2022-752). We appreciate your constructive comments and suggestions. We have studied them carefully and made revisions on the manuscript. These comments, suggestions and the corresponding replies are listed below.

Note that the title has been changed to "Impact of aerosol optics on vertical distribution of ozone in autumn over YRD" to clarify the study region and applicability.

The referee's comments are highlighted by gray. Followed by the comments are our responses. The texts led by **line number** are the current texts in manuscript, with some important revisions colored by red. The underlined blue texts, e.g., See the Response to Comment S6, means that the detailed information are provided in our response to the comment numbered with S6.

With regards,

Shuqi Yan, Bin Zhu*, and all co-authors.

General comments:

G1. A wider literature review needs to be incorporated to better provide background information on modeling features of aerosol mixing states and on prior studies on the topic, particularly in different parts of the world. This will provide a more complete statement of the problem, current uncertainties and gaps and further contextualize the presented results.

Thanks for this suggestion. We have addressed this comment by the following aspects.

1)Basic concepts of aerosol mixing and roles of physical and chemical properties

The influences of aerosol morphology, hygroscopicity, coating process and chemical composition on aerosol optics have been discussed in the Introduction (See the Response to Comment G2).

2)Definitions of the three mixing states and model limitations

The brief definitions of aerosol mixing states, model treatments on aerosol optics and the model limitations are included in Section 2.3 (See the Response to Comment G2).

3)Model evaluations compared with previous works

We add more sites and calculate the statistic metrics on meteorology, $PM_{2.5}$ and ozone. The statistical metrics are compared with previous studies (See the Response to Comment S6).

G2. A clear definition of the mixing states is missing and particularly how WRF-Chem treats them and what existing modeling limitations are. A discussion on the role of aerosol composition vs physical properties should be included. Please refer to the relevant literature including but not limited to the following:
Riemer, N., Ault, A. P., West, M., Craig, R. L., & Curtis, J. H. (2019). Aerosol mixing state: Measurements, modeling, and impacts. Reviews of Geophysics, 57, 187– 249. https://doi.org/10.1029/2018RG000615

Thanks for this suggestion. We have addressed this comment by the following aspects.

1)The definition of mixing states, WRF-Chem treatments on mixing states and model limitations:

We have added brief definitions of the mixing states and the model treatments on aerosol optics in Section 2.3. The treatment of mixing states, including aerosol species, aerosol number distribution, and formula of optical parameters of MOSAIC sectional approach are reasonably documented by previous papers (e.g., Fast et al., 2006; Grell et al., 2005), which have been adopted in this study and cited in the manuscript.

One major limitation of WRF-Chem model is basically discussed in Section 2.3. The real-world aerosol mixing state varies with emission, meteorology, chemical composition and other factors. The WRF-Chem, as well as other popular 3D models, is hard to trace and resolve the dynamic evolution and impact factors of aerosol mixing state at present state. The three mixing states in this study are idealized cases, which will inevitably cause the simulated aerosol optics deviating from observation.

In Section 2.3

In this work, the effect of aerosol optics on ozone profiles is addressed by its mixing states. We study three ideal types of mixing states: internal mixing, core-shell mixing and external mixing, which depend on the mixing behavior hypothesis of scattering and absorbing components. In internal mixing state, the relative fractions of chemical species in one particle are the same as that of the bulk aerosols. The complex refractive index (RI) of bulk aerosols is calculated by the volume-averaged RI of all aerosol species, and then it is passed to Mie optical module to calculate the required optical parameters (e.g., scattering coefficient, absorbing coefficient and single scattering albedo). The detailed formulas of aerosol optical parameters for MOSAIC sectional scheme are documented by previous works (e.g., Fast et al., 2006; Grell et al., 2005). In core-shell mixing, aerosol particles are hypothesized to be concentric spheres with BC as the core and non-BC aerosols as the coating shell (Riemer et al., 2019). The RI of the shell is the volume-averaged RI of non-BC aerosols, and the optics of core-shell mixed particles can also be treated by the Mie optical module (Ackerman & Toon, 1981). In external mixing state, each particle contains only one species with fixed optical characteristics. It is not included in the current WRF-Chem model, and the approximate treatment has been proposed by Gao et al. (2021b). In general, the Mie optical module separates BC aerosols from the bulk aerosols, and treats the optics of nonBC and BC aerosols individually.

To study the aerosol effect on ozone, four experiments are conducted (Table 2). The case "int" is the base experiment (the default option in WRF-Chem), in which the aerosols are internally mixed. The cases "csm" and "ext" are core-shell mixing and external mixing, respectively. The case “noARI” turns off aerosol-radiation feedback by setting aerosol optical depth as zero in radiation and photolysis modules. Therefore, the difference between noARI and three other experiments indicates the effect of aerosols in the corresponding mixing state.

One should note that the real-world aerosol mixing state varies with emission, meteorology, composition, and other factors. The dynamic evolution of aerosol mixing state and its influencing factors have not been addressed in most current 3D models (Matsui et al., 2013). This work addresses aerosol optics by the three ideal mixing states, which will inevitably cause the simulated aerosol optics deviating from observation.

2)The role of aerosol composition vs physical properties:

At monthly or annual scales, the aerosol chemical compositions in East China are dominated by SNA (sulfate, nitrate and ammonium) (commonly larger than 50%), followed by OM (organic matter) and BC (Tao et al., 2017; Yang et al., 2011). The scattering components, SNA and some OM, account for the majority of total aerosol concentration, and the absorbing components (mainly related to BC-contained aerosols) commonly account for less than 10% of total aerosol concentration (Tan et al., 2020, 2022).

We agree that aerosol physical properties (e.g., morphology, hygroscopicity) and chemical composition notably influence aerosol optical properties. The scattering components (e.g., SNA) generally contribute dominantly to aerosol extinctions. The contribution of SNA to total aerosol scattering coefficient can reach up to 60% (Tian et al., 2015). Under high humidity conditions, the hygroscopic growth of SNA can further enhance its extinction by 2~3 times (Zeng et al., 2019). Although BC takes a small proportion in aerosol mass concentration, the light absorption of BC contributes more than 70% to aerosol absorption coefficient (Yang et al., 2008), and the mass

absorbing efficiency of BC is comparable to the mass scattering efficiency of $PM_{2.5}$ (Tao et al., 2017). Additionally, the morphology of BC and BC-related coating process can change aerosol mixing state and optical properties (Bond et al., 2006; Liu et al., 2019). The BC light absorption can be amplified by a factor of 50~200% due to coatings (Cappa et al., 2012; Jacobson, 2001; Liu et al., 2017).

Aerosol mixing states also have significant effects on optical properties. The relative importance of aerosol mixing state and chemical composition on aerosol optics can be inferred from Zeng et al. (2019). Supposing aerosol is composed of BC and sulfate, where BC mass fraction is 5%. Aerosol exerts negative radiative forcing (RF) at the near surface in all mixing states. When the mixing state is changed from external to core-shell mixing (sulfate coating on BC), the decrease in RF (ΔRF) is 7.5W/m^2. The ΔRF becomes 7.9W/m^2 if sulfate is completely replaced with organic matter. The variation in ΔRF induced by coating material change is slighter than that by mixing state under various RH conditions. Curci et al. (2015) quantified the sensitivity of aerosol optical properties to aerosol mixing state, chemical composition and other parameters. Aerosol mixing state is found to be the dominant factor introducing uncertainties, explaining 30~35% of the uncertainty in AOD and single scattering albedo. Therefore, we infer that the effect of mixing state on aerosol properties could be more important than the effect of chemical composition.

In Introduction

The effect of aerosols on BL is related to aerosol optics, which are determined by aerosol morphology (Liu et al., 2019), hygroscopicity (Zeng et al., 2019), coating process (Bond et al., 2006) and chemical composition. The aerosol chemical composition in East China is dominated by SNA (sulfate, nitrate and ammonium) (larger than 50%), followed by organic matter and BC (3~8%) (Yang et al., 2011; Tan et al., 2020, 2022). The contribution of SNA to total aerosol scattering coefficient can reach up to 60% (Tian et al., 2015), and BC accounts for more than 70% of total aerosol absorbing coefficient (Yang et al., 2009). Furthermore, aerosol optics are strongly affected by aerosol mixing states. Since the real-world mixing state is highly variable and hard to be explicitly resolved (Riemer & West, 2013), three typical mixing states are generally hypothesized by previous works: internal mixing, core-shell mixing and external mixing. The mixing state is largely affected by the mixing behavior of BC with other aerosol species. The freshly emitted BC is commonly externally mixed with other species, but it will become more internally mixed due to coating process (Riemer et al., 2019). The BC light absorption can be amplified by a factor of 50~200% after being coated with scattering aerosols (Cappa et al., 2012; Jacobson, 2001; Liu et al., 2017). ~~The mixing behaviour hypothesis of aerosol scattering and absorbing components yields three major mixing states: internal mixing, core-shell mixing and external mixing. In internal and core-shell mixing, BC absorption can be enhanced by 50~100% (Bond et al., 2006; Jacobson, 2001). In external mixing, the absorption ability is weaker but scattering ability is stronger (Zeng et al., 2019).~~ Accordingly, aerosol mixing state alters aerosol optical properties and affects its interactions with BL and photolysis. Gao et al. (2021b) found that aerosols result in smaller boundary layer height (PBLH) reduction in external mixing (11.6 m) than in core-shell mixing (24 m), consequently leading to different changes in photolysis rates and ozone concentration.

***Summary*:** The discussions of mixing state definition, roles of aerosol composition vs physical properties are included now. In this work, the hypothesis of three ideal mixing states will inevitably cause the simulated optics deviating from observation. However, the major chemical compositions in the atmospheric aerosols (e.g., SNA, OM, BC) have been included in the model, and the effects of coating and hygroscopicity are also considered by optical modules. The effect of chemical composition on aerosol optics has been reasonably addressed in the three ideal mixing states. In future, the role of aerosol composition on aerosol optics should be further addressed.

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G3. Generalizability of the results. The WRF-Chem simulations are performed over a period of 4 days, while most of the presented analyses focus on a single day. Given the authors apply WRF-Chem in a configuration with a very small one-nested domain, the computational cost is very limited, so a justification on why the analysis is limited to such a few days should be provided, particularly in the context of the generalizability of the results. Robust statistics should be presented to make claims on physical and chemical mechanisms occurring in the atmosphere. Further, the authors should consider how different emission and meteorological conditions may play a role in impacting ozone processes. For example, it would be relevant to analyze different ozone regimes by looking at a full year of simulations. At the least, simulations of a representative month for each season (or the season with most ozone formation) should be included.

Thanks for this suggestion. In general, we have done the additional works: 1) Extending the simulation for one month (15 Oct to 15 Nov). 2) Finding that domain size and domain resolution have limited influence on ozone simulations. 3) Adding more sites and providing statistic metrics on meteorology, $PM_{2.5}$ and ozone. It can reflect the effect of different aerosol levels and aerosol-meteorology feedback under different emissions. 4) Finding that aerosols consistently decrease ozone concentration in polluted and clean days, and the effect is stronger in polluted days as expected. The details can be referred to the Response to Comment S5, S6.

We agree that ozone regime and ozone chemistry are highly variable among different seasons and meteorological conditions. In this study, we focus on the ozone characteristic in autumn season, considering the seasonal synoptic situation over the Yangtze River Delta Region. In spring and summer, the weather conditions vary significantly and precipitation events frequently occur. In winter, the solar radiation is relatively weak, so the ozone concentration is commonly not high. In autumn, the Yangtze River Delta Region is dominated by calm weather conditions. The weak winds and less cloudy weather conditions are better for studying the effect of aerosols on ozone. Therefore, we conduct our field observations and model simulations in a representative month of autumn season (actually from 15 Oct to 15 Nov). The aerosol-ozone relationships are applicable to only autumn season.

Specific Comments:

S1. Line 51: as mentioned in the general comments, the three mixing states need to be defined and explained also in the context of the modeling tool adopted. Also, an explicit discussion on the role of aerosol composition should be included. Are measurements of aerosol composition available at that or nearby sites?

Thanks for this suggestion. We provide the definition of three aerosol mixing states and discuss the importance of aerosol chemical composition in the Response to Comment G2.

The aerosol chemical composition measurements (e.g., SNA) are not available at nearby sites in the autumn of 2020. We have investigated some previous observations conducted in Nanjing in recent years. Generally, SNA are the dominant component in $PM_{2.5}$, with the proportion of about 58~81% (Guo et al., 2019; Liu et al., 2019; Liu et al., 2021; Sun et al., 2020; Wang et al., 2016; Wang et al., 2019; Yu et al., 2019). The BC commonly takes a minor part in $PM_{2.5}$. Tan et al. (2020, 2022) and Shen et al. (2021) reveal that the ratio of $BC/PM_{2.5}$ is approximately in the range of 3~8% in autumn season. Our simulations indicate that the ratio of BC to $PM_{2.5}$ is 1.9~5.6%, and the ratio of $SNA/PM_{2.5}$ is 55~75%, which are generally consistent with previous observations. Therefore, the simulation in this study can reasonably reproduce the observed aerosol composition and address the effect of composition on optics. Additionally, in the Response to Comment G2, we have suggested that the effect of aerosol composition on aerosol optics is reasonably addressed in the three ideal mixing states.

References

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Liu, Y., Li, H., Cui, S., Nie, D., Chen, Y., & Ge, X. (2021). Chemical characteristics and sources of water-soluble organic nitrogen species in PM2. 5 in Nanjing, China. Atmosphere, 12(5), 574.

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S2. Line 72: how much representative of the overall physics and chemistry of the atmosphere are the chosen days of November 2020 whose anthropogenic emissions may be still strongly impacted by COVID lockdowns?

Thanks for this suggestion. We have collected the MEIC emissions in 2019 and 2020 during the manuscript revision. Table X1 presents the reductions of anthropogenic emissions in 2020 (during the pandemic) with respect to 2019 (before the pandemic). In March 2020 when China is undergoing strict lockdowns, the $PM_{2.5}$ and NO_X emissions are reduced by 10~15% compared with 2019. In November, the pandemic is effectively controlled in China. $PM_{2.5}$ and NO_X emissions are reduced by at most 2.1%. It indicates that during the study period, COVID lockdowns have limited impacts on the emissions over China. We believe that the physics and chemistry of the atmosphere are still representative. **The MEIC emission in current manuscript has been changed from 2016 based to 2020 based now**.

Table X1. Relative reductions of $PM_{2.5}$ and NO_X in 2020 compared with 2019. The comparisons are performed in March (strict lockdowns in China) and November (lockdowns in only a few cities). The units are 10^4Mg for $PM_{2.5}$ and 10^4 Mmol for NO_X.

	East China (110~125°E, 25~45°N)		Whole China	
	$PM_{2.5}$	NO_X	$PM_{2.5}$	NO_X
201903	31.9	114.5	54.1	178.9
202003	28.1	98.8	48.7	155.7
reduction	11.9%	13.7%	10.0%	13.0%
201911	33.0	122.2	56.9	194.2
202011	32.3	122.0	56.1	191.5
reduction	2.1%	0.1%	1.4%	1.4%

S3. Line 73: How were the vertical profiles of meteorological and chemical species measured? At which heights?

The meteorological elements are measured by XLS-II tethered balloon system with a sounding balloon. The data are sampled every second until it loses signal. The air pollutants sensors are mounted on UAV platform. The UAV climbs vertically from the ground to about 1 km with a speed of 2m/s, and it descends along the same path with the same speed. Meteorology and air pollutants vertical data are averaged to 50 m intervals. The introduction of observation instruments of $PM_{2.5}$, BC and ozone can be referred to Shi et al. (2020, 2021). These texts are added into Section 2.1.

References

Shi, S., Zhu, B., Lu, W., Yan, S., Fang, C., Liu, H., Liu, D., Liu, C.: Estimation of radiative forcing and heating rate based on vertical observation of black carbon in Nanjing, China, Sci. Tot. Environ., https://doi.org/10.1016/j.scitotenv.2020.144135, 2020.

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S4. Line 82: a reference for WRF-Chem should be included

Thanks for this suggestion. We have added two references.

In Section 2.2:

The model used in this study is the WRF-Chem (V3.9.1.1) model (Fast et al., 2006; Grell et al., 2005).

Fast, J. D., Gustafson, W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model, J. Geophys. Res., 111, https://doi.org/10.1029/2005jd006721, 2006.

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S5. Section 2.2 (Model configuration): More details and clarifications are needed. For example, the defined domains are unusually small (less than 100 x 100 grid cells) which raise concerns about the model's ability to develop proper meteorology and also chemical processes. Also, is 9 km resolution small enough to capture the spatial variability in ozone? Are there other ground-based sites in the region that would enable a more complete model evaluation (so at more than one point)? The authors mention the MEIC emissions are used. What is their temporal and spatial resolution? From the listed website it appears that the latest emission inventory available is for the year 2017. However, WRF-Chem was run over November 2020, a year that experienced significant changes in most anthropogenic emissions due to COVID lockdowns. How was this mismatch in emissions accounted for? Also, how often were the boundary conditions from ERA-5 and chemical species updated?

Thanks for this suggestion. It includes the following sub-comments:

1) The domain size (less than 100x100 grids) and resolution (9km) may be inadequate to develop proper meteorology, chemical processes and capture the spatial variability in ozone.

We agree that domain size and resolution are important to develop proper meteorology, chemical processes and capture the spatial variability in ozone. We have evaluated the performance of meteorology and chemistry simulations at additional sites. The model reasonably captures the temporal variation of temperature, wind speed,

wind direction, $PM_{2.5}$ and ozone (See the Response to Comment S6). We additionally conduct a simulation with the resolution of 3×3km and the domain size of 120×120, finding that the ozone and aerosol-induced ozone change have indiscernible differences with those of original 9km resolution. Some previous works also study ozone by resolutions coarser than 9km or domain size less than 100×100 grids, and get reasonable simulations (Gao et al., 2018; Li et al., 2021; Wang et al., 2020; Yang et al., 2022; Zhang et al., 2021).

References:

Gao, J., Li, Y., Zhu, B., Hu, B., Wang, L., and Bao, F.: What have we missed when studying the impact of aerosols on surface ozone via changing photolysis rates?, Atmos. Chem. Phys., 20, 10831–10844, https://doi.org/10.5194/acp-20-10831-2020, 2020. (12km)

Li, L., Hu, J., Li, J., Gong, K., Wang, X., Ying, Q., & Zhang, Y. (2021). Modelling air quality during the EXPLORE-YRD campaign–Part II. Regional source apportionment of ozone and PM2. 5. Atmospheric Environment, 247, 118063. (12km)

Wang, P., Qiao, X., & Zhang, H. (2020). Modeling PM2.5 and O3 with aerosol feedbacks using WRF/Chem over the Sichuan Basin, southwestern China. Chemosphere, 254, 126735. (12km)

Yang, H., Chen, L., Liao, H., Zhu, J., Wang, W., & Li, X. (2022). Impacts of aerosol–photolysis interaction and aerosol–radiation feedback on surface-layer ozone in North China during multi-pollutant air pollution episodes. Atmospheric Chemistry and Physics, 22(6), 4101-4116. (9km, less than 50*50 grids)

Zhang, M., Zhao, C., Yang, Y., Du, Q., Shen, Y., Lin, S., & Liu, C. (2021). Modeling sensitivities of BVOCs to different versions of MEGAN emission schemes in WRF-Chem (V3.6) and its impacts over eastern China. Geoscientific Model Development, 14(10), 6155-6175. (12km)

2) Are there other ground-based sites in the region that would enable a more complete model evaluation?

We have extended the simulation for one month (15 Oct to 15 Nov 2020) and select four additional sites around the Nanjing (the main study site) to evaluate the model performance (See the Response to Comment S6).

3) What is their temporal and spatial resolution of MEIC? The latest inventory is in year 2017 from the website, but the simulation year (2020) has experienced significant changes in most anthropogenic emissions due to COVID lockdowns.

The MEIC dataset is published by month and the spatial resolution is approximately 25km. Although the emissions have experienced significant changes in 2020, we have revealed that the emissions in our study period are less affected by COVID lockdowns (See the Response to Comment S6). **We have replaced The MEIC emission inventory in 2016 based to 2020 based**.

4) How often were the boundary conditions from ERA-5 and chemical species updated?

The ERA-5 boundary conditions are updated 6 hourly. It is the same for chemical species.

In Section 2.2:

The initial and boundary fields of meteorology are provided by ERA5 0.25°×0.25° reanalysis data. The chemical initial and boundary fields are provided by WACCM. They are all updated every 6 hours.

S6. Line 114: the quantified WRF-Chem skills should be put into the context of other literature studies (not necessarily in the same region) to verify if the model performance with the proposed setup is aligned with prior published work. Adding more sites to the evaluation and extending the simulation to more days/months will make the assessment of WRF- Chem performance more robust.

Thanks for this suggestion. We have extended the simulation to one month and evaluated model performance at four additional sites around Nanjing (Figure 2). The statistical metrics of temperature, wind speed, wind direction, $PM_{2.5}$ and ozone temporal variations are provided in Table 3 in the main text. Although a few metrics slightly exceeds its benchmark values, most of them are in acceptable ranges. The statistical metrics of $PM_{2.5}$ and ozone are consistent with previous works (e.g., Chen et al., 2022; Hu et al., 2016; Singh et al., 2012; Zhang et al., 2014). It increases the robustness of our simulation results.

Additionally, we compare the aerosol effect on ozone during some representative polluted and clean days. It is found that BL ozone is generally reduced by aerosol effect, and the reduction is more significant in polluted conditions.

In Section 3.1

Four Additional sites around Nanjing, i.e., Changzhou (CZ), Huainan (HN), Maanshan (MS), and Huaian (HA) (Figure 1) are chosen to evaluate the performance on the time variation of meteorological parameters (tempera-

ture, wind speed and wind direction), $PM_{2.5}$ and ozone in the base experiment (internal mixing). The statistical metrics include index of agreement (IOA), mean bias (MB), root mean square error (RMSE), mean normalized bias (MNB) and mean fractional bias (MFB). The calculations are from Lu et al. (1997), especially, the IOA of wind direction is from Kwok et al. (2010). Benchmark values of meteorology and air pollutants are derived from Emery et al. (2011) and EPA (2005; 2007). The temporal variations of simulated meteorology and air pollutants are generally in good agreement with observations (Figure 2). From Table 3, temperature presents the highest IOA, with a slightly large MB at HA site. The simulated wind direction is similar to observation, and MB exceeds benchmark value at only one site. The simulated wind speed is a bit higher, which is because the WRF model tends to overestimate wind speed due to the description of surface roughness (Jia and Zhang, 2020, 2021; Jiménez and Dudhia, 2012). $PM_{2.5}$ is moderately overestimated, but all the metrics are within the benchmarks. The IOA of ozone exceeds 0.8 at all sites, and only one site shows a MNB out of benchmark. The model statistical metrics of $PM_{2.5}$ and ozone are consistent with previous works (Chen et al., 2022; Hu et al., 2016; Singh et al., 2012; Zhang et al., 2014a). Generally, the base experiment simulations on the temporal variation of meteorology and air pollutants are acceptable, which reflects a reasonable atmosphere reproduced by the model.

Table 3. The statistic metrics of the model performance on time series of temperature (Tem), wind speed (WS), wind direction (WD), $PM_{2.5}$ and ozone. The benchmark values are from Emery et al. (2011) and EPA (2005; 2007). Metrics that out of bechmarks are marked with red.

Variable	Metric	NJ	CZ	HN	MS	HA	benchmark
Tem	IOA	0.97	0.97	0.96	0.97	0.96	>0.8
	MB	0.18	0.18	0.42	0.31	0.50	<±0.5
	RMSE	1.07	1.07	1.43	1.10	1.52	
WS	IOA	0.64	0.63	0.66	0.71	0.64	>0.6
	MB	0.47	0.68	0.52	-0.05	0.71	<±0.5
	RMSE	1.13	1.06	1.09	0.88	1.09	<2
WD	IOA	0.94	0.93	0.93	0.95	0.88	
	MB	-3.32	10.47	9.91	-4.65	6.16	<±10
	RMSE	35.91	38.53	46.31	36.56	52.92	
$PM_{2.5}$	IOA	0.74	0.84	0.83	0.64	0.86	
	MNB	0.26	0.01	0.12	0.36	0.34	
	MFB	0.17	-0.04	0.06	0.23	0.22	<±0.6
Ozone	IOA	0.87	0.88	0.91	0.83	0.88	
	MNB	-0.07	-0.03	0.03	0.03	0.20	<±0.15
	MFB	-0.15	-0.07	0.02	0.03	0.17	

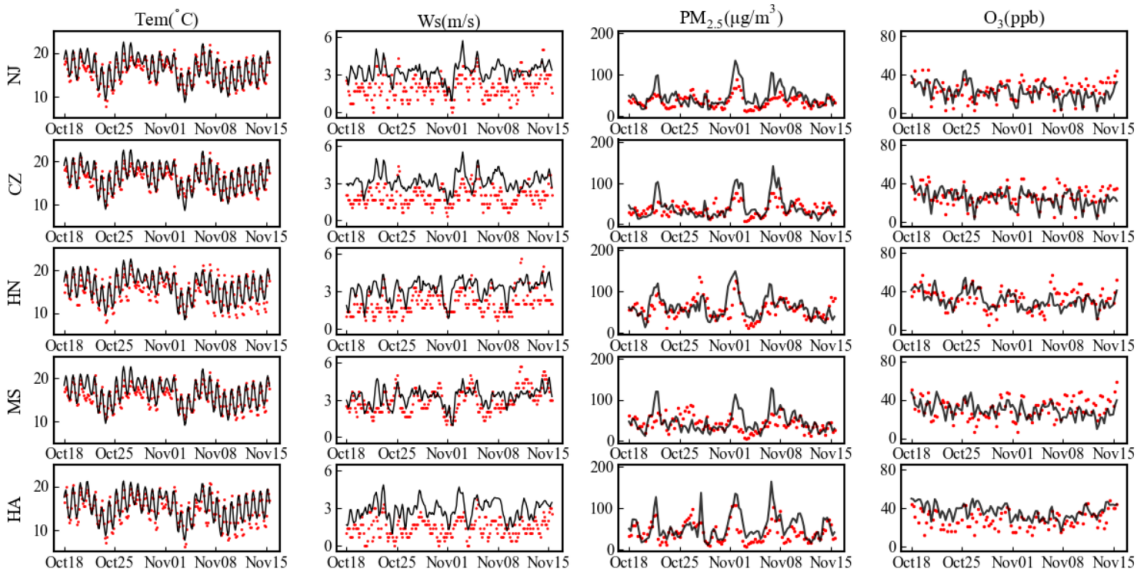

Figure 2. Model evaluations on the time series on temperature (Tem), wind speed (Ws), $PM_{2.5}$ and Ozone at five sites. The Changzhou (CZ), Huainan (HN), Maanshan (MS) and Huaian (HA) sites are located to the east, west, south and north of Nanjing, respectively. The red dots are observations and black lines are simulations (after 3-point running average). The time range is from 08:00 on 15 October to 20:00 on 15 November.

In Section 3.4

The ozone variations during representative clean and polluted episodes are shown in Table 5. The ozone concentrations within BL in the internal mixing experiment are consistently reduced during all episodes. The core-shell mixing state shows slightly lower reductions than internal mixing, and the ozone reductions are the least in external mixing state. The differences in ozone relative changes between clean and polluted episodes are distinct. For example, in the internal mixing state, the relative reductions are about 0~5% in clean episodes and 6~11% in polluted episodes, indicating that the aerosol effect is more profound under high aerosol contents. On 2 November which is the highest pollution episode during the study period, the relative changes of ozone are approximately -11~-2%. It can be inferred that aerosol effect on photolysis rates, ozone precursors and ozone concentration might be consistent under different underlying surface and pollution conditions, and it is more significant in polluted conditions.

Table 5. The diurnal averaged (08:00~17:00) quantities within BL during some representative clean and polluted episodes. The PM2.5 (μg/m3) and ozone (ppb) are the values in the base experiment (internal mixing). The last three columns are the changes and relative changes of ozone under different mixing states.

Date	$PM_{2.5}$	Ozone	Δint	Δcsm	Δext
Clean episode					
10-19	32	53	-1.9 (-3.5%)	-1.7 (-3.1%)	+0.0 (+0.0%)
10-20	18	49	-0.8 (-1.5%)	-0.7 (-1.4%)	+0.1 (+0.1%)
10-25	28	53	-2.0 (-3.6%)	-1.9 (-3.5%)	-0.2 (-0.3%)
11-03	33	39	-0.7 (-1.8%)	-0.7 (-1.8%)	-0.4 (-1.1%)
11-05	17	44	-1.5 (-3.3%)	-1.5 (-3.3%)	-0.9 (-1.9%)
11-12	23	36	-1.9 (-4.9%)	-1.9 (-4.9%)	-0.9 (-2.5%)
Polluted episode					
10-22	91	46	-3.0 (-6.1%)	-2.8 (-5.6%)	-0.3 (-0.7%)
11-02	111	56	-7.7 (-10.5%)	-6.4 (-8.6%)	-1.5 (-2.0%)
11-07	87	39	-4.6 (-10.7%)	-4.6 (-10.6%)	-1.6 (-3.7%)
11-08	82	39	-3.0 (-7.0%)	-2.8 (-6.6%)	-0.6 (-1.4%)

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(with relevant refs listed)

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S7. Line 200: how is ADVC defined and computed?

The dynamic modules of WRF-Chem model can diagnose all the physical and chemical processes contributing to ozone variation. The ozone variation is affected by the following processes:

$$\frac{\partial \mathrm{O3}}{\partial t} = \underbrace{-\left(u\frac{\partial}{\partial x}+v\frac{\partial}{\partial \mathrm{y}}+w\frac{\partial}{\partial z}\right)\mathrm{O3}}_{\mathrm{Advc}} + \left(\frac{\partial \mathrm{O3}}{\partial t}\right)_{\mathrm{Vmix}} + \left(\frac{\partial \mathrm{O3}}{\partial t}\right)_{\mathrm{Chem}}$$

The first term is ADVC, i.e., the ozone tendency (ppb/h) caused by horizontal and vertical advections.