We would like to express our sincere appreciation to the reviewer for the valuable and constructive suggestions, which have helped us improve the quality of this manuscript. We have addressed all these comments carefully and revised the manuscript accordingly. Following the Reviewer' comments in black, please find our point-to-point responses in blue. Hereafter, all new added or modified sentences are marked in blue and italic in this response.

Anonymous Referee #1

1. Introduction. “The feedbacks of aerosols to meteorology have been widely investigated by two-way coupled meteorology and air quality models in the past two decades.” Two-way coupled meteorological and air quality models have been developed and applied for almost three decades (Jacobson, 1994; 1997; 1998, 2001).

Response: According to this suggestion, the sentence in Introduction has been revised as “*The feedbacks of aerosols to meteorology have been widely investigated by two-way coupled meteorology and air quality models in the past three decades (Jacobson, 1994, 1997, 1998, 2001, 2002; Grell et al., 2005; Wong et al., 2012; Wang et al., 2014; Zhou et al., 2016; Briant et al., 2017; Feng et al., 2021).*” in the revised manuscript.

2. Table 1. what is the vertical resolution of the boundary layer in each model (how many layers in the bottom 1 km and what is the bottom-layer thickness?

Response: All the three coupled models used in this study have 30 levels (i.e., 29 layers) from the surface to 100 hPa. There are 11 layers in the bottom 1 km and the bottom-layer thickness is 23.2 m. The sentence “The vertical resolution for all simulations consisted of 30 levels from the surface (~20 m) to 100 hPa.” was revised as “*All the three coupled models used in this study have 30 levels* (i.e., 29 layers) *from the surface to 100 hPa with 11 layers in the bottom 1 km and the bottom-layer thickness being 23.2 m.*”. We also revised Table 1 accordingly.

Table 1. Model setups and inputs for the two-way coupled models (WRF-CMAQ, WRF-Chem and WRF-CHIMERE).

		WRF-CMAQ	WRF-Chem	WRF-CHIMERE
Domain configuration	Horizontal grid spacing	27 km (110 × 150)	27 km (120 × 160)	27 km (120 × 170)
	Vertical resolution	*30 levels*	*30 levels*	*30 levels*
Physics parameterization	Shortwave radiation	RRTMG	RRTMG	RRTMG
	Longwave radiation	RRTMG	RRTMG	RRTMG

	Cloud microphysics	Morrison	Morrison	Thompson
	PBL	ACM2	YSU	YSU
	Cumulus	Kain-Fritsch	Grell-Freitas	Grell-Freitas
	Surface	Pleim-Xiu	Monin-Obukhov	Monin-Obukhov
	Land surface	Pleim-Xiu LSM	Noah LSM	Noah LSM
	Icloud	*Xu-Randall method*	*Xu-Randall method*	*Xu-Randall method*
Chemistry scheme	*Aerosol mechanism*	*AERO6*	*MOSAIC*	*SAM*
	Aerosol size distribution	*Modal (3 modes)*	*Sectional (4 bins)*	*Sectional (10 bins)*
	Aerosol mixing state	Core-Shell	Core-Shell	Core-Shell
	Gas-phase chemistry	CB6	CBMZ	MELCHIOR2
	Photolysis	*Fast-JX with cloud effects*	*Fast-JX with cloud effects*	*Fast-JX with cloud effects*
Emission	Anthropogenic emission	MEIC 2017	MEIC 2017	MEIC 2017
	Biogenic emission	MEGAN v3.0	MEGAN v3.0	MEGAN v3.0
	Biomass burning emission	FINN v1.5	FINN v1.5	FINN v1.5
	Dust emission	Foroutan	GOCART	Menut
	Sea-salt emission	Gong	Gong	Monahan
Input data	Meteorological ICs and BCs	FNL	FNL	FNL
	Chemical ICs and BCs	MOZART	MOZART	LMDZ-INCA

3. Table 1. How many aerosol size bins and components per bin? Do you use a modal or discrete bin approach?

Response: For aerosol size distribution, the modal approach was used in the WRF-CMAQ model (Binkowski and Roselle, 2003) and included Aitken, accumulation and coarse modes with 9 (black carbon (BC), organic carbon (OC), sulfate, nitrate, ammonium, remaining unspeciated particulate matter (PMOTHR), primary non-carbon organic matter (PNCOM), water, metals), 11 (BC, OC, sulfate, nitrate, ammonium, PMOTHR, PNCOM, water, metals, sea salt, dust) and 3 (coarse primary particulate matter (PMC), sea salt, dust) aerosol components, respectively. WRF-Chem and WRF-CHIMERE applied the sectional approach with 4 and 10 size bins covering dry diameters ranging from 0.039 to 10 µm and 0.039 to 40 µm, respectively (Zaveri et al., 2008; Nicholls et al., 2014; Menut et al., 2013, 2016). In WRF-Chem, BC, OC, sulfate, nitrate and sea salt are put in Bins 1–3 and dust, sea salt, and other inorganic matter (OIN) in Bin 4. For WRF-CHIMERE, BC, OC, sulfate and primary particulate matter (PPM) are assigned in Bins 1–5, BC, OC, sulfate, dust and sea salt in Bin 6, dust and sea salt in Bins 7 & 9, BC, OC, PPM, dust and sea salt in Bin 8 and dust in Bin 10. The approaches for aerosol size distributions used in the three coupled models are listed in the revised Table 1, as shown in the reply of Question 2. We also compiled all the components in each mode or bin in Table S2 and added it into the

Supplement of the revised manuscript. In addition, we added the sentence "*As illustrated in Table 1 and Table S2 for aerosol size distribution, we used modal approach with Aitken, accumulation and coarse modes in WRF-CMAQ, and the 4-bin and 10-bin sectional approaches in WRF-Chem and WRF-CHIMERE models, respectively (Binkowski and Roselle, 2003; Zaveri et al., 2008; Nicholls et al., 2014; Menut et al., 2013).*" in the Section 2.1 of the revised manuscript. We revised the sentence "These biases were produced by the configurations of different aerosol and gas phase mechanisms, online dust emission schemes, and chemical ICs and BCs in the two-way coupled models.". In lines 536-538 of the revised manuscript, the sentence "These biases were produced by the configurations of different aerosol and gas phase mechanisms, online dust emission schemes, and chemical ICs and BCs in the two-way coupled models." is revised as "*These biases could be related to different aerosol and gas phase mechanisms, dust and sea salt emission schemes, chemical ICs and BCs, and aerosol size distribution treatments applied in the three two-way coupled models.*".

Table S2. Summary of the aerosol size distribution treatments and components in each mode or bin for the coupled WRF-CMAQ, WRF-Chem and WRF-CHIMERE models.

Model	*Aerosol mechanism*	*Modal approach*									
		Aitken				*Accumulation*		*Coarse*			
WRF-CMAQ	*AERO6*	*BC, OC, sulfate, nitrate, ammonium, PMOTHR[d], PNCOM[e] water, metals*				*BC, OC, sulfate, nitrate, ammonium, PMOTHR, PNCOM, water, metals, sea salt, dust*		*PMC[f], sea salt, dust*			
		Sectional approach									
WRF-Chem	*MOSAIC[a]*	*Bin 1* *0.039–0.156 μm* *BC, OC, sulfate, nitrate, sea salt[d]*		*Bin 2* *0.156–0.625 μm* *BC, OC, sulfate, nitrate, sea salt*		*Bin 3* *0.625–2.5 μm* *BC, OC, sulfate, nitrate, sea salt*		*Bin 4* *2.5–10.0 μm* *Duste, sea salt, OIN[g]*			
WRF-CHIMERE	*SAM[b]*	*Bin 1* *0.039–0.078 μm* *BC, OC, sulfate, PPM[c]*	*Bin 2* *0.078–0.156 μm* *BC, OC, sulfate, PPM*	*Bin 3* *0.156–0.312 μm* *BC, OC, sulfate, PPM*	*Bin 4* *0.312–0.625 μm* *BC, OC, sulfate, PPM*	*Bin 5* *0.625–1.25 μm* *BC, OC, sulfate, PPM*	*Bin 6* *1.25–2.5 μm* *BC, OC, sulfate, dust, sea salt*	*Bin 7* *2.5–5.0 μm* *Dust, sea salt*	*Bin 8* *5.0–10.0 μm* *BC, OC, PPM, dust, sea salt*	*Bin 9* *10.0–20.0 μm* *Dust, sea salt*	*Bin 10* *20.0–40.0 μm* *Dust*

[a] *MOSAIC is the Model for Simulating Aerosol Interactions and Chemistry, and the cbmz-mosaic emissions in "PNNL" format (emiss_inpt_opt==101) was used in WRF-Chem simulations.*

[b] *SAM is the sectional aerosol mechanism.*

[c] *PPM is the primary particulate matter.*

[d] *PMOTHR is the remaining unspeciated particulate matter in fine mode and more detailed information is at https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list.*

[e] *PNCOM is the primary non-carbon organic matter in fine mode and more detailed information is at https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list.*

[f] *PMC is the primary particulate matter in coarse mode and more detailed information is at https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list.*

[g] *OIN is the other inorganic matter.*

4. Table 1. Does photolysis account for clouds? How are clouds treated for radiative transfer calculations?

Response: Yes, all the three coupled models considered the effects of cloud on photolysis in the photolysis calculation. Even though the Fast-JX photolysis scheme was applied in the three coupled models, how the cloud effects were treated was different. For WRF-

CMAQ, the impacts of cloud cover and cloud optical properties on the radiative transfer and actinic flux are taken into account. Both cloud fraction (CF) from WRF and CF calculated using relative humidity (RH) and RH thresholds (set to 0.85 over ocean and 0.75 over land (Mocko and Cotton, 1995)) are utilized in the CMAQ version of Fast-JX (Sundqvist et al., 1989). The total column CF is determined by exponential-random overlapping. The optical properties of hydrometeors (cloud liquid water, rain, snow, graupel and ice) output from WRF are included in the computation of cloud optical properties in the CMAQ version of Fast-JX (Hu and Stamnes, 1993; Fu, 1996; Binkowski et al., 2007). In the WRF-Chem version of Fast-JX, CF is set to 1 when cloud liquid water content (CLWC) is greater than 0 and CF is set to 0 when CLWC = 0, and the calculation of cloud optical depth only considers CLWC from WRF. In WRF-CHIMERE, CF = 1 when CLWC or cloud ice content (CIC) is greater than 0.00001 g m^{-3} and CF = 0 if CLWC or CIC is 0. To compute cloud optical depth in the CHIMERE version of Fast-JX, both cloud liquid water and ice output from WRF are taken into account (Mailler et al., 2017).

These information is reflected in the revised Table 1, and we also added this sentence *"In the Fast-JX photolysis scheme used by the three coupled models, the impacts of clouds are included by considering cloud cover and cloud optical properties. However, the calculations of cloud cover and cloud optical properties are different in these models and all the relevant information is listed in Table S1."* in Lines 166-170 of the revised manuscript. Table S5 is in Supplement of the revised manuscript.

Table S1. Summary of representations of cloud cover and cloud optical properties in the Fast-JX scheme for WRF-CMAQ, WRF-Chem and WRF-CHIMERE.

Model	*Cloud clover*	*Cloud optical properties*			
		Optical properties	*Effective Wavelength*	*Hydrometeor types*	*Method*
WRF-CMAQ	*1. CF[a] from WRF and CF calculated using RH and RH thresholds* *2. Exponential-random overlapping*	*Extinction, single scattering albedo and asymmetry factor*	*294.6, 303.2, 310.0, 316.4, 333.1, 382.0 and 607.7 nm*	*Cloud liquid water, rain, snow, graupel and ice*	*The parameterizations proposed by Hu and Stamnes (1993) and Fu (1996)*
WRF-Chem	*1. CF=0 if CLWC[b]=0* *2. CF=1 if CIC[c]>0*	*Cloud optical depth*	*300, 400, 600 and 999 nm*	*Cloud liquid water*	*Based on the empirical functions of relative humidity and cloud liquid water content*
WRF-CHIMERE	*1. CF=0 if CLWC or CIWC=0* *2. CF=1 if CLWC or CIC>0*	*Cloud optical depth*	*200, 300, 400, 600, and 999 nm*	*Cloud liquid water and ice*	*Based on the functions of cloud effective radiuses and cloud liquid water/ice contents*

[a]CF is cloud fraction. [b]CLWC is cloud liquid water content. [c]CIC is cloud ice content.

In this study, the RRTMG shortwave radiation (SWR) and longwave radiation (LWR) schemes were chosen for the three two-way coupled models. The considerations of cloud effects on SWR and LWR in RRTMG are twofold, as listed below:

(1) Regarding the effects of cloud cover on radiation: the cloud fraction (CF) at grid scale is calculated using relative humidity and mixing ratio of all hydrometeors (Xu and Randall, 1996) and then the total column CF is determined by maximum-random overlapping (Iacono et al., 2008). The cumulus CF is only considered when the Kain-Fritsch cumulus scheme is chosen and computed as a function of the updraft mass flux in cloud (Alapaty et al., 2012). Therefore, the coupled WRF-CMAQ model with the Kain-Fritsch cumulus scheme included the cumulus CF impacts on RRTMG radiation but not the WRF-Chem and WRF-CHIMERE models with the Grell-Freitas cumulus scheme.

(2) Regarding the impacts of cloud optical properties on radiation: the treatments of cloud liquid water and ice optical properties as proposed by Hu and Stamnes (1993) and Fu (1996) are deployed in both RRTMG SWR and LWR schemes in all three coupled models.

Thus, we added these descriptions in Lines 160-163 of the revised manuscript as follows.

"*To consider the effects of clouds on radiative transfer calculations, the fractional cloud cover and cloud optical properties were included in the RRTMG shortwave/longwave radiation schemes used by all three coupled models (Xu and Randall, 1996; Iacono et al., 2008). The coupled WRF-CMAQ model with the Kain-Fritsch cumulus scheme included the cumulus cloud fraction impacts on RRTMG radiation (Alapaty et al., 2012), but not the WRF-Chem and WRF-CHIMERE models with the Grell-Freitas cumulus scheme.*"

5. Table 1. What height is the model top and how are model-top boundary conditions treated?

Response: The height of model top is about 16 km (100 hPa). For the meteorological model-top boundary conditions, WRF assumes zero flux at the model top. Regarding the chemical model-top boundary conditions, WRF-CMAQ and WRF-Chem models both take into account the impacts of stratosphere-troposphere O_3 exchange using the parameterization of O_3-potential vorticity (Safieddine et al., 2014; Xing et al., 2016). For WRF-CHIMERE, climatological data from the Laboratoire de Météorologie Dynamique (LMDz) coupling a global chemistry and aerosol model INteractions between Chemistry and Aerosols (INCA) were used for model-top boundary conditions (Mailler et al., 2017).

To distinguish lateral and model-top BCs used in this study, these sentences are edited in the revised manuscript as follows:

"The meteorological ICs and BCs were derived from the National Center for Environmental Prediction Final Analysis (NCEP-FNL) datasets (http://rda.ucar.edu/datasets/ds083.2), with a horizontal resolution of 1° × 1° at 6-hour intervals for each of the three coupled models." was revised as "*The meteorological ICs and lateral BCs were derived from the National Center for Environmental Prediction Final Analysis (NCEP-FNL) datasets (http://rda.ucar.edu/datasets/ds083.2), with a horizontal resolution of 1° × 1° at 6-hour intervals for each of the three coupled models, and the flux at the model-top boundary is set to zero.*".

"The chemical ICs/BCs were downscaled from the Whole Atmosphere Community Climate Model (WACCM) for WRF-CMAQ and WRF-Chem via the mozart2camx and mozbc tools, respectively." was revised as "*The chemical ICs/lateral BCs were downscaled from the Whole Atmosphere Community Climate Model (WACCM) for WRF-CMAQ and WRF-Chem via the mozart2camx and mozbc tools, respectively. WRF-CHIMERE used the climatology data from a general circulation model developed at the Laboratoire de Météorologie Dynamique (LMDz) coupling a global chemistry and aerosol model INteractions between Chemistry and Aerosols (INCA) (Mailler et al., 2017). For chemical model-top BCs, WRF-CMAQ and WRF-Chem models both take into account the impacts of stratosphere-troposphere O_3 exchange using the parameterization of O_3-potential vorticity (Safieddine et al., 2014; Xing et al., 2016), and WRF-CHIMERE utilized the LMDz-INCA climatology data (Mailler et al., 2017).*"

6. The authors evaluate with RMSE, which is an absolute quantity for each variable. However, normalized gross error (absolute value of differences between model and data, divided by data, summed over all locations and normalized by the number of locations, is a more useful metric since it gives error relative to the data values rather than an absolute amount. It is similar to NMB, but with absolute values taken, since NMB cancels out large errors of the opposite sign. Also, it would be useful to see some time-series plots of model results versus data.

Response: We agree that it would be useful to add the normalized gross error (NGE) in our simulation assessment. We added NGE in Table 3, Table 4, Table 5 and Table S3 as well as descriptions in the revised manuscript.

Lines 226-227: "*normalized gross error (NGE)*"

Lines 565-568: "*All models with ARI feedbacks enabled resulted in slight decreases in annual and seasonal* O_3 *NMBs and NGEs, ranging from −3.02% to +0.85% (the only positive value of +0.85% was produced by WRF-CMAQ in summer) and from −1.42% to −0.75%, respectively.*"

Lines 568-570: "*Meanwhile, for ACI effects, WRF-Chem and WRF-CHIMERE had increased annual* O_3 *NMBs and NGEs of 0.12%−0.65% and 0.40%−0.55%, respectively.*"

We presented the time-series plots of simulated and observed hourly meteorology and air qualit over Eastern China during the year of 2017 in Figs. 3 and 6, respectively. The metrological variables involved surface shortwave radiation (SSR), temperature (T2), specific humidity (Q2), relative humidity (RH2) and wind speed (WS10). The air quality variables included $PM_{2.5}$ and O_3 concentrations. These two figures are put into the revised manuscript.

The related descriptions are added in the revised manuscripts as follows:

Lines 278-280: "*Looking at annual and seasonal T2, models tended to have a negative bias, and T2 underestimations in spring and winter were greater than those in summer and autumn (Figs. 3 and 4).*"

Lines 317-319: "*The R values for all three models ranged from 0.47 to 0.60; WRF-CMAQ and WRF-Chem overestimated wind speed by approximately 0.5 m* s^{-1}*, while WRF-CHIMERE overestimated it by approximately 1.0 m* s^{-1} *(Table S3 and Figs. 3–4)).*"

Lines 531-535: "*As shown in Table 4 and Figs. 6–7, WRF-CMAQ underestimated annual and seasonal (except for autumn)* $PM_{2.5}$ *concentrations with NMBs ranging from −9.78% to −6.39% and −17.68% to +5.17%, respectively. WRF-Chem generated both overestimations and underestimations of* $PM_{2.5}$ *at the annual and seasonal scales, with related NMBs varying from −39.11% to +24.72%, respectively.*"

Table 3. Statistical metrics (R, MB, NMB, *NGE*, and RMSE) between annual simulations and satellite retrievals of surface shortwave and longwave radiation, TOA shortwave and longwave radiation, precipitation, cloud fraction, and liquid water path in eastern China. The best results are in bold, while mean simulations and observations are in italics.

Variables	Statistics	WRF-CMAQ_NO	WRF-CMAQ_ARI	WRF-Chem_NO	WRF-Chem_ARI	WRF-Chem_BOTH	WRF-CHIMERE_NO	WRF-CHIMERE_ARI	WRF-CHIMERE_BOTH
	Mean_sim	*197.15*	*180.94*	*203.48*	*194.52*	*201.45*	*197.39*	*191.34*	*195.58*
	R	0.76	0.75	0.73	**0.78**	0.75	0.61	0.64	0.66

Surface shortwave radiation (*172.74* W m^{-2})	MB	24.41	8.21	30.74	21.78	28.71	24.75	18.71	22.94
	NMB (%)	14.13	**4.75**	17.79	12.61	16.62	14.34	10.84	13.29
	NGE (%)	*15.13*	***8.66***	*18.61*	*13.53*	*17.38*	*17.44*	*14.42*	*15.83*
	RMSE	30.25	**20.37**	35.34	26.88	32.80	34.70	29.60	31.45
Surface longwave radiation (*322.3* W m^{-2})	Mean_sim	*316.25*	*315.83*	*312.96*	*312.60*	*312.32*	*313.33*	*314.60*	*314.47*
	R	0.98	0.98	0.98	0.98	0.98	**0.99**	**0.99**	**0.99**
	MB	-6.05	-6.46	-9.34	-9.70	-9.97	-9.66	-8.39	-8.53
	NMB (%)	**-1.88**	-2.00	-2.90	-3.01	-3.09	-2.99	-2.60	-2.64
	NGE (%)	***3.22***	*3.46*	*3.70*	*3.77*	*3.84*	*3.96*	*3.60*	*3.66*
	RMSE	**13.65**	14.13	14.81	14.97	15.17	15.47	14.52	14.72
TOA shortwave radiation (*111.56* W m^{-2})	Mean_sim	*107.76*	*112.68*	*110.38*	*110.95*	*107.16*	*114.33*	*116.62*	*113.09*
	R	**0.81**	0.79	0.69	0.68	0.62	0.65	0.65	0.65
	MB	-3.80	1.13	-1.18	-0.61	-4.40	3.12	5.42	1.89
	NMB (%)	-3.40	**1.01**	-1.05	-0.55	-3.94	2.81	4.87	1.70
	NGE (%)	***10.19***	*10.45*	*11.52*	*10.96*	*11.69*	*14.43*	*14.36*	*12.93*
	RMSE	**15.75**	16.04	17.07	16.10	17.21	20.85	20.67	18.96
TOA longwave radiation (*233.68* W m^{-2})	Mean_sim	*231.54*	*232.26*	*234.34*	*233.96*	*234.39*	*232.52*	*232.17*	*233.18*
	R	0.88	0.90	0.91	0.91	**0.92**	0.74	0.74	0.76
	MB	-2.14	-1.42	0.66	0.28	0.71	-0.61	-0.96	0.05
	NMB (%)	-0.92	-0.61	0.28	**0.12**	0.30	-0.26	-0.41	0.02
	NGE (%)	*2.28*	*2.04*	*1.79*	*1.79*	***1.74***	*3.02*	*2.98*	*2.92*
	RMSE	6.94	6.20	6.00	5.94	**5.86**	10.10	10.07	9.70
Precipitation (*948.91* mm y^{-1})	Mean_sim	*872.42*	*896.98*	*1069.06*	*1056.95*	*1081.84*	*1165.06*	*1160.35*	*1163.77*
	R	**0.71**	**0.71**	**0.71**	**0.71**	0.70	0.69	0.69	0.69
	MB	-76.49	-51.93	120.15	108.04	132.94	207.05	202.35	205.76
	NMB (%)	-9.23	**-8.40**	12.66	11.39	14.01	21.61	21.12	21.48
	NGE (%)	***32.46***	*34.36*	*44.54*	*43.38*	*45.13*	*42.54*	*42.52*	*42.58*
	RMSE	**573.14**	595.76	675.91	668.92	693.74	776.60	786.36	790.73
Cloud cover (*64.09* %)	Mean_sim	*52.51*	*53.32*	*48.18*	*47.80*	*47.46*	*58.12*	*57.98*	*58.55*
	R	0.68	0.68	**0.69**	**0.69**	0.68	0.66	0.66	0.64
	MB	-11.58	-10.77	-16.12	-16.50	-16.83	-6.60	-6.74	-6.18
	NMB (%)	-18.07	-16.80	-25.07	-25.66	-26.18	-10.20	-10.41	**-9.54**
	NGE (%)	*19.48*	*18.87*	*26.01*	*26.56*	*26.97*	*16.74*	*16.92*	***16.72***
	RMSE	16.47	16.28	20.17	20.48	20.73	15.28	15.33	15.34
liquid water path (*88.44* g m^{-2})	Mean_sim	*53.50*	*57.15*	*32.29*	*31.87*	*31.08*	*56.23*	*56.21*	*54.00*
	R	**0.61**	0.58	0.47	0.46	0.28	0.55	0.55	0.51
	MB	-34.94	-31.29	-56.16	-56.58	-57.36	-32.37	-32.40	-34.61
	NMB (%)	-39.51	**-35.38**	-63.49	-63.97	-64.86	-36.54	-36.56	-39.06
	NGE (%)	*57.05*	*57.99*	*66.88*	*67.25*	*67.91*	***53.15***	*53.33*	*56.88*
	RMSE	54.35	54.31	63.54	63.92	67.21	**53.39**	53.42	55.86

Table 4. Statistical metrics (R, MB, NMB, *NGE*, and RMSE) between annual simulations and observations of surface $PM_{2.5}$, O_3, NO_2, SO_2, and CO in eastern China. The best results are in bold, while mean simulations and observations are in italics.

Variables	Statistics	WRF-CMAQ_NO	WRF-CMAQ_ARI	WRF-Chem_NO	WRF-Chem_ARI	WRF-Chem_BOTH	WRF-CHIMERE_NO	WRF-CHIMERE_ARI	WRF-CHIMERE_BOTH
$PM_{2..5}$ (*44.99* μg/m^3)	Mean_sim	*40.59*	*42.12*	*44.45*	*46.65*	*38.33*	*62.17*	*65.36*	*65.13*
	R	**0.68**	**0.68**	0.65	0.65	0.69	0.52	0.53	0.53
	MB	-4.40	-2.87	-0.54	1.66	-6.66	17.18	20.37	20.14
	NMB (%)	-9.78	-6.39	**-1.21**	3.69	-14.81	38.19	45.27	44.76
	NGE (%)	***46.41***	*47.08*	*57.82*	*59.91*	*52.10*	*89.85*	*94.10*	*94.01*
	RMSE	**27.62**	27.69	32.58	34.64	32.48	55.13	60.25	59.41
O_3 (*62.23* μg/m^3)	Mean_sim	*55.06*	*54.41*	*88.53*	*87.81*	*87.89*	*76.92*	*76.48*	*76.89*
	R	0.54	0.55	0.46	0.45	0.45	**0.62**	**0.62**	**0.62**
	MB	-7.17	-7.83	26.30	25.58	25.65	14.69	14.25	14.66
	NMB (%)	**-11.52**	-12.57	42.26	41.10	41.22	23.60	22.90	23.55
	NGE (%)	***41.02***	*41.40*	*87.02*	*86.17*	*86.57*	*58.17*	*57.63*	*58.18*
	RMSE	**28.32**	28.68	48.10	47.99	47.82	29.65	29.46	29.75
NO_2 (*31.2* μg/m^3)	Mean_sim	*33.94*	*34.46*	*21.17*	*21.98*	*21.40*	*21.85*	*22.20*	*22.24*
	R	0.59	**0.60**	0.50	0.50	0.50	0.55	0.56	0.56
	MB	2.74	3.26	-10.03	-9.22	-9.80	-9.35	-9.00	-8.96
	NMB (%)	**8.77**	10.44	-32.14	-29.55	-31.40	-29.96	-28.84	-28.73
	NGE (%)	*55.04*	*55.74*	*54.57*	*54.37*	*54.43*	*50.56*	***50.82***	*50.89*
	RMSE	**19.14**	19.48	21.23	21.21	21.21	18.72	18.68	18.70
SO_2 (*18.51* μg/m^3)	Mean_sim	*14.02*	*14.39*	*8.22*	*8.56*	*7.85*	*8.88*	*9.18*	*9.19*
	R	0.40	0.40	0.44	0.44	**0.46**	0.40	0.41	0.41
	MB	-4.49	-4.12	-10.29	-9.95	-10.66	-9.63	-9.33	-9.32
	NMB (%)	-24.25	**-22.24**	-55.61	-53.76	-57.57	-52.02	-50.39	-50.34
	NGE (%)	*75.44*	*76.26*	*64.18*	*64.20*	***66.09***	*75.54*	*75.86*	*75.87*
	RMSE	21.11	21.30	20.13	**20.02**	20.20	22.07	22.17	22.18
CO (*0.96* mg/m^3)	Mean_sim	*0.44*	*0.45*	*0.53*	*0.54*	*0.53*	*0.56*	*0.58*	*0.57*
	R	0.23	0.24	0.21	0.22	0.22	0.47	**0.48**	0.47
	MB	-0.52	-0.51	-0.43	-0.42	-0.43	-0.40	-0.39	-0.39
	NMB (%)	-53.97	-52.99	-45.10	-43.94	-44.68	-41.82	**-40.11**	-40.28
	NGE (%)	*65.44*	*65.11*	*53.63*	*53.38*	*53.80*	*47.27*	***47.08***	*47.09*
	RMSE	0.90	0.90	0.82	0.83	0.83	**0.62**	**0.62**	**0.62**

Table 5. Statistical metrics (R, MB, NMB, *NGE*, and RMSE) of simulated and satellite-retrieved AOD, total column ozone, tropospheric column NO_2, PBL column SO_2, total column CO, and total column density of NH_3 in eastern China. The best results are in bold,

while annual mean simulations and observations are in italics.

Variables	Statistics	WRF-CMAQ_NO	WRF-CMAQ_ARI	WRF-Chem_NO	WRF-Chem_ARI	WRF-Chem_BOTH	WRF-CHIMERE_NO	WRF-CHIMERE_ARI	WRF-CHIMERE_BOTH
AOD (*0.27*)	Mean_sim	*0.26*	*0.27*	*0.35*	*0.36*	*0.25*	*0.21*	*0.22*	*0.22*
	R	0.80	0.80	0.80	0.80	0.75	**0.87**	**0.87**	0.86
	MB	-0.01	-0.01	0.09	0.09	-0.01	-0.05	-0.05	-0.04
	NMB (%)	-3.99	**-2.93**	34.14	35.03	-4.92	-18.72	-17.37	-16.22
	NGE (%)	*34.90*	*34.82*	*58.21*	*58.89*	*41.46*	*32.15*	***32.11***	*32.06*
	RMSE	**0.09**	**0.09**	0.15	0.15	0.10	**0.09**	**0.09**	0.10
O_3 VCDs (*312.07* DU)	Mean_sim	*306.15*	*306.15*	*300.77*	*300.73*	*300.46*	*307.69*	*307.47*	*307.75*
	R	**0.98**	**0.98**	0.97	0.97	0.97	0.65	0.65	0.65
	MB	-5.92	-5.92	-10.68	-10.72	-10.99	-3.69	-3.91	-3.63
	NMB (%)	-1.90	-1.90	-3.43	-3.44	-3.53	-1.19	-1.26	**-1.17**
	NGE (%)	***2.46***	***2.46***	*25.02*	*25.02*	*25.08*	*10.95*	*10.89*	*10.93*
	RMSE	**8.91**	**8.91**	83.72	83.73	83.94	39.88	39.71	39.73
Tropospheric NO_2 VCDs (*2.71×10^{15}* molecules cm^{-2})	Mean_sim	*3.80*	*3.91*	*3.07*	*3.08*	*3.06*	*2.62*	*2.63*	*2.63*
	R	0.85	0.85	**0.87**	**0.87**	**0.87**	**0.87**	**0.87**	**0.87**
	MB	1.09	1.21	0.62	0.63	0.61	0.28	0.29	0.29
	NMB (%)	40.35	44.64	25.27	25.52	24.89	**12.03**	12.47	12.42
	NGE (%)	*52.80*	*55.08*	*46.01*	*46.05*	***45.17***	*46.06*	*46.31*	*46.24*
	RMSE	3.18	3.33	2.27	2.27	2.27	**1.65**	1.67	1.68
PBL SO_2 VCDs (*0.09* DU)	Mean_sim	*0.07*	*0.07*	*0.09*	*0.09*	*0.06*	*0.06*	*0.06*	*0.06*
	R	0.53	0.53	**0.56**	**0.56**	0.54	0.50	0.50	0.50
	MB	-0.03	-0.02	-0.03	-0.02	-0.03	-0.03	-0.02	-0.02
	NMB (%)	-27.32	-25.48	-32.50	**-21.50**	-35.08	-28.64	-27.31	-27.51
	NGE (%)	***57.45***	*58.26*	*67.55*	*68.07*	*64.83*	*68.31*	*68.61*	*68.80*
	RMSE	**0.07**	**0.07**	0.08	0.08	**0.07**	**0.07**	**0.07**	**0.07**
Total CO VCDs (*21.60×10^{17}* molecules cm^{-2})	Mean_sim	*20.34*	*20.35*	*22.20*	*22.20*	*22.21*	*22.34*	*22.36*	*22.35*
	R	0.83	0.83	**0.87**	**0.87**	**0.87**	0.86	0.86	0.86
	MB	-1.26	-1.24	0.93	0.93	0.94	1.19	1.21	1.19
	NMB (%)	-5.83	-5.75	**4.35**	4.37	4.44	5.64	5.70	5.65
	NGE (%)	*9.33*	***9.31***	*10.30*	*10.28*	*10.32*	*11.02*	*11.06*	*11.10*
	RMSE	**2.54**	**2.54**	2.69	2.68	2.69	2.57	2.58	2.58
Total NH_3 VCDs (*16.05×10^{15}* molecules cm^{-2})	Mean_sim	*13.06*	*13.15*	*12.31*	*12.27*	*8.63*	NA	NA	NA
	R	**0.76**	**0.76**	0.73	0.73	0.76	NA	NA	NA
	MB	-3.00	-2.90	-3.27	-3.32	-3.34	NA	NA	NA
	NMB (%)	-18.66	**-18.08**	-21.01	-21.28	-21.41	NA	NA	NA
	NGE (%)	***47.69***	*48.09*	*50.84*	*50.80*	*50.99*	*NA*	*NA*	*NA*
	RMSE	**9.26**	9.47	9.48	9.46	9.61	NA	NA	NA

NA indicates that outputs of NH_3 column concentrations were not extracted from WRF-CHIMERE with/without aerosol feedback simulations.

Table S3. Statistic metrics (R, MB, NMB, *NGE*, and RMSE) between simulated and observed annual SSR, T2, RH2, Q2, WS10, WD10, precipitation, and PBLH at LT 08:00 and 20:00) in eastern China. The best results are in bold, while mean simulations and observations are in italics.

Variables	Statistics	WRF-CMAQ_NO	WRF-CMAQ_ARI	WRF-Chem_NO	WRF-Chem_ARI	WRF-Chem_BOTH	WRF-CHIMERE_NO	WRF-CHIMERE_ARI	WRF-CHIMERE_BOTH
SSR	Mean_sim	*191.12*	*171.14*	*194.52*	*180.04*	*191.71*	*197.88*	*188.63*	*189.54*
(*155.22* W m^{-2})	R	0.88	**0.89**	0.88	**0.89**	0.88	0.85	0.85	0.85
	MB	35.89	15.91	39.30	24.82	36.48	42.65	33.41	34.32
	NMB (%)	23.12	**10.25**	25.32	15.99	23.50	27.48	21.52	22.11
	NGE (%)	*206.62*	*170.85*	*202.41*	***170.70***	*208.05*	*242.53*	*221.67*	*226.29*
	RMSE	133.05	**120.60**	134.16	123.94	134.45	154.71	147.73	148.57
T2	Mean_sim	*12.81*	*12.61*	*12.99*	*12.84*	*12.96*	*11.84*	*11.68*	*11.69*
(*13.68* ℃)	R	**0.97**	**0.97**	**0.97**	**0.97**	**0.97**	0.96	0.96	0.96
	MB	-0.86	-1.06	-0.68	-0.83	-0.71	-1.83	-2.00	-1.98
	NMB (%)	-6.33	-7.76	**-4.97**	-6.09	-5.21	-13.39	-14.60	-14.50
	NGE (%)	***10.58***	*10.76*	*10.79*	*10.95*	*10.86*	*17.00*	*17.65*	*17.60*
	RMSE	**2.88**	2.94	3.05	3.07	3.05	3.87	3.94	3.97
Q2	Mean_sim	*8.69*	*8.51*	*8.57*	*8.54*	*8.58*	*8.35*	*8.30*	*8.30*
(*8.87* g kg^{-1})	R	**0.90**	**0.90**	0.89	0.89	0.89	0.88	0.88	0.88
	MB	-0.18	-0.35	-0.30	-0.32	-0.28	-0.52	-0.57	-0.56
	NMB (%)	**-2.00**	-3.98	-3.36	-3.66	-3.19	-5.84	-6.37	-6.35
	NGE (%)	***16.80***	*16.85*	*19.70*	*19.66*	*19.77*	*20.55*	*20.65*	*20.62*
	RMSE	**2.93**	2.95	3.09	3.09	3.10	3.17	3.18	3.18
RH2	Mean_sim	*71.03*	*70.51*	*70.01*	*70.33*	*70.13*	*70.41*	*70.58*	*70.46*
(*67.48* %)	R	**0.73**	**0.73**	0.68	0.68	0.68	0.65	0.65	0.65
	MB	3.55	3.03	2.53	2.85	2.64	2.93	3.10	2.97
	NMB (%)	5.26	4.49	**3.74**	4.22	3.92	4.34	4.59	4.41
	NGE (%)	***19.90***	*19.91*	*23.45*	*23.71*	*23.71*	*24.77*	*24.88*	*24.90*
	RMSE	**18.92**	18.98	19.78	19.79	19.84	20.81	20.82	20.84
WS10	Mean_sim	*3.27*	*3.23*	*3.30*	*3.29*	*3.30*	*3.85*	*3.83*	*3.83*
(*2.81* m s^{-1})	R	**0.62**	0.61	0.60	0.59	0.59	0.47	0.47	0.47
	MB	0.45	0.42	0.49	0.48	0.49	1.04	1.02	1.02
	NMB (%)	16.16	**14.98**	17.45	17.11	17.53	36.98	36.27	36.34
	NGE (%)	*96.20*	***95.00***	*100.16*	*100.09*	*100.55*	*136.55*	*135.59*	*135.75*
	RMSE	1.89	1.88	1.92	1.92	1.93	2.46	2.45	2.45
WD10	Mean_sim	177.13	176.62	177.87	177.82	178.11	171.97	171.53	171.68
(*175.27* °)	R	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.02
	MB	1.85	1.35	2.60	2.55	2.83	-3.31	-3.74	-3.60
	NMB (%)	1.06	0.77	1.48	1.45	1.62	-1.89	-2.14	-2.05
	NGE (%)	*94.30*	***94.00***	*101.16*	*101.09*	*101.55*	*126.75*	*125.79*	*125.85*
	RMSE	149.57	149.45	149.45	149.38	149.57	148.70	148.47	148.71

Precipitation	*Mean_sim*	*2.46*	*2.31*	*3.24*	*3.19*	*3.26*	*3.31*	*3.24*	*3.21*
(PREC)	R	**0.59**	**0.59**	0.50	0.50	0.50	0.35	0.34	0.34
(*2.72* mm d^{-1})	MB	-0.27	-0.42	0.51	0.46	0.53	0.59	0.52	0.48
	NMB (%)	**-9.80**	-15.35	18.86	16.83	19.43	21.46	18.96	17.63
	NGE (%)	*310.71*	***283.10***	*442.60*	*428.11*	*445.89*	*573.24*	*565.36*	*557.56*
	RMSE	8.03	**7.96**	10.32	10.26	10.33	10.87	10.85	10.93
PBLH00	*Mean_sim*	*253.54*	*251.61*	*288.41*	*263.16*	*282.81*	*276.45*	*270.28*	*269.63*
(*432.13* m)	R	**0.21**	**0.21**	0.17	0.17	0.17	0.17	0.17	0.17
	MB	-178.59	-180.52	-143.72	-168.97	-149.32	-155.68	-161.85	-162.50
	NMB (%)	-41.33	-41.77	**-33.26**	-39.10	-34.55	-36.03	-37.45	-37.61
	NGE (%)	*58.89*	*58.75*	***54.37***	*56.96*	*54.51*	*57.20*	*57.63*	*57.28*
	RMSE	380.23	378.79	**371.27**	379.72	372.14	373.78	375.85	374.52
PBLH12	*Mean_sim*	*230.14*	*236.80*	*358.05*	*332.45*	*346.54*	*363.47*	*360.13*	*359.03*
(*547.02* m)	R	**0.40**	**0.40**	0.39	**0.40**	0.39	0.34	0.35	0.35
	MB	-316.88	-310.22	-188.97	-214.57	-200.48	-183.55	-186.89	-188.00
	NMB (%)	-57.93	-56.71	-34.55	-39.22	-36.65	**-33.56**	-34.16	-34.37
	NGE (%)	*65.84*	*65.23*	*59.55*	*59.05*	*59.49*	*59.65*	***59.32***	*59.66*
	RMSE	505.64	502.24	459.64	460.51	**459.50**	470.39	467.90	469.19

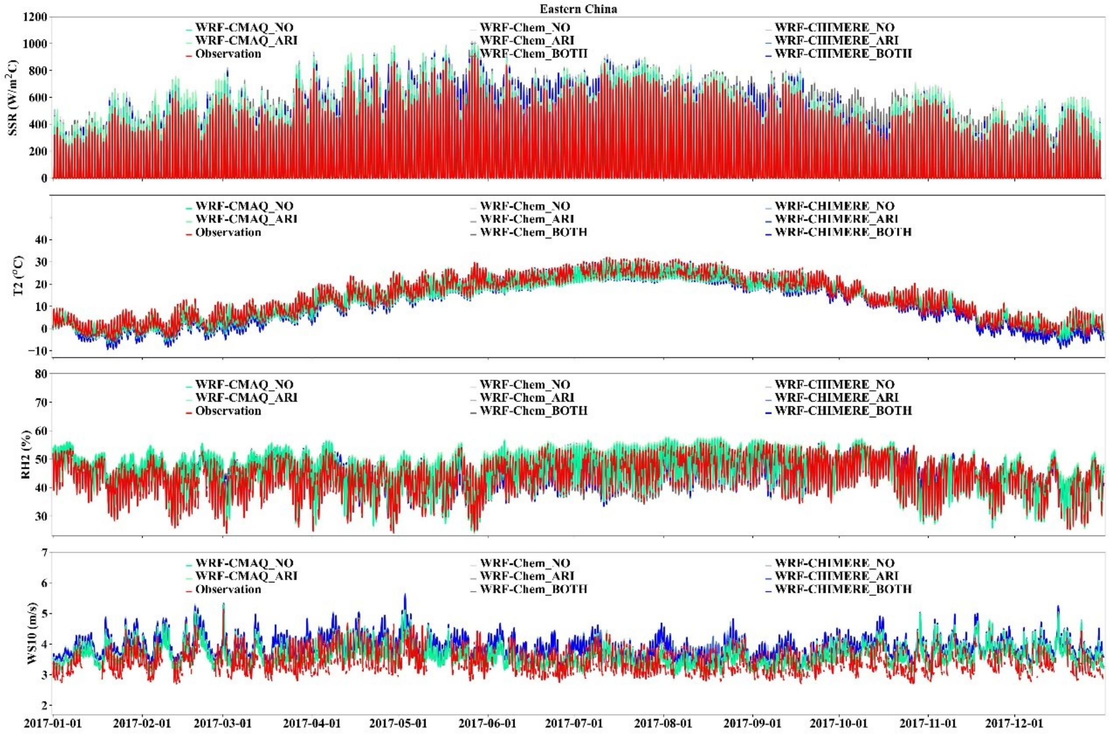

Figure 3. Time series of observed and simulated hourly SSR, T2, RH2 and WS10 by coupled WRF-CMAQ, WRF-Chem and WRF-CHIMERE with/without aerosol feedbacks over Eastern China during the year of 2017.

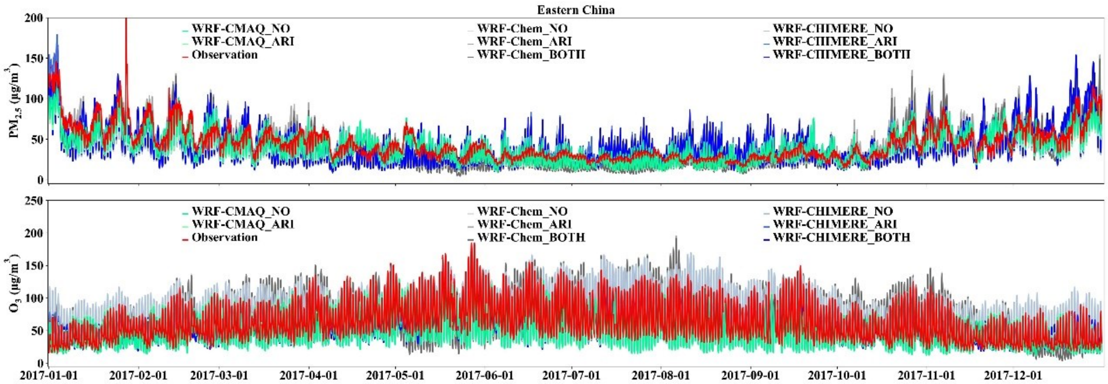

Figure 6. Time series of observed and simulated hourly $PM_{2.5}$ and O_3 concentrations by WRF-CMAQ, WRF-Chem and WRF-CHIMERE with/without aerosol feedbacks over Eastern China during the year of 2017.

7. A lot of comparisons are performed, but what are the most relevant comparisons with data? Ozone and $PM_{2.5}$ calculations? Please focus the discussion of results more. Right now the results section is crammed with lots of information that is not easy to determine from what is important and not important.

Response: We agreed that we need to be more focused while evaluating the simulation results from the three coupled models. At the same time, we believe the most relevant comparisons in this paper should look into the surface meteorological variables (SSR, T2, RH2, WS10) and air quality variables ($PM_{2.5}$ and O_3). The comparisons against satellite data should focus on SSR, SLR, PREC, cloud fraction, and cloud liquid water path. To improve the paper's readability, we rearranged some paragraphs and figures and added sentences in the revised manuscript, as listed below:

(1) The results and discussion about the comparisons of simulated Q2, PREC, PBLH00, PBLH12 against ground-based observations are moved to Section 1.1 of Supplement. In Lines 250-252 of the revised manuscript, we added "Here, we mainly focused on the comparisons of SSR, T2, RH2, and WS10, and the analysis of PREC, PBLH00, and PBLH12 are presented in Section 1.1 of Supplement."

(2) The comparisons of simulated TSR and TLR against satellite observations are moved to Section 1.2 of Supplement. We modified the sentences in Lines 373-380 of the revised manuscript as "To further evaluate the performance of WRF-CMAQ, WRF-Chem, and WRF-CHIMERE against satellite observations, we analyzed the annual and seasonal statistical metrics of short- and long-wave radiation at the surface, precipitation, cloud

cover, and liquid water path simulated by the three coupled models with and without aerosol feedbacks, via comparisons between simulations and satellite-borne observations (Table 3; Figures 5, S9, S12–S14). In addition, the evaluations of short- and long-wave radiation at top of the atmosphere (TOA) are presented in Section 1.2 of Supplement."

(3) The evaluation of simulated NO_2, SO_2 and CO against surface measurements is moved to Section 2 of Supplement. In Lines 525-527 of the revised manuscript, we added "The evaluations between surface measurements and simulations of $PM_{2.5}$ and O_3 are presented below, and the performance assessments of other gaseous pollutants are in Section 2 of Supplement."

(4) The original Figure 4 and Figure 7 are moved to Supplement as Figure S8 and Figure S20, respectively.

We added more discussions of in-depth analysis in the result part of revised manuscript as follows:

Lines 401-402: "*the representation differences for aerosol components, size distributions and mechanisms contributed to the diversity of seasonal MBs (Tables 1 and S2).*"

Lines 407-421: "*When ARI effects are enabled, the diversities of refractive indices of aerosol species groups lead to the discrepancies of online calculated aerosol optical properties in different shortwave and longwave (SW and LW) bands in the RRTMG SW/LW radiation schemes of WRF-CMAQ, WRF-Chem, and WRF-CHIMERE (Tables S5–S6). The online calculated cloud optical properties induced by aerosol absorption in the RRTMG radiation schemes are different in treatments of aerosol species groups in the three coupled models. With enabling ACI effects, the activation of cloud droplets from aerosols based on the Köhler theory is taken into account in WRF-Chem and WRF-CHIMERE, in comparison to simulations without aerosol feedbacks (Table S7). The treatments of prognostic ice nucleating particles (INP) formed via heterogeneous nucleation of dust particles (diameters > 0.5 μm) and homogeneous freezing of hygroscopic aerosols (diameters > 0.1 μm) are only considered in WRF-CHIMERE, but the prognostic ice nucleating particles are not included in WRF-CMAQ and WRF-Chem. These discrepancies eventually contribute to the differences of simulated radiation changes caused by aerosols.*"

Lines 485-495: "*This may be explained as the different parameterization treatments of cloud droplet number concentration (CDNC) simulated by the three coupled models with/without enabling ACI effects. The cloud condensation nuclei activated from aerosol* *particles can increase CDNC and impact on LWP and CF. Without enabling any aerosol feedbacks or only enabling ARI, the CDNC is default prescribed as a constant value of 250 cm^{-3} in the Morrison scheme of WRF-CMAQ and WRF-Chem and 300 cm^{-3} in the Thompson scheme of WRF-CHIMERE. When only ACI or both ARI and ACI are enabled, the online calculating of prognostic CDNC is performed in WRF-Chem and WRF-CHIMERE by using the method of maximum supersaturation (Abdul-Razzak and Ghan, 2002; Chapman et al., 2009; Tuccella et al., 2019).*"

Lines 537-539: "*These biases could be related to different aerosol and gas phase mechanisms, dust and sea salt emission schemes, chemical ICs and BCs, and aerosol size distribution treatments applied in the three two-way coupled models.*"

Lines 573-578: "*Such diversity in NMB and NGE variations can be explained by two aspect differences. For model-top boundary conditions, the WRF-CMAQ and WRF-Chem models employed the parameterization scheme of* O_3*-potential vorticity and WRF-CHIMERE used the climatological data from LMDz-INCA. For gas-phase chemistry mechanisms, three coupled models incorporate a variety of photolytic reactions, with a more comprehensive explanation provided in Section 4.2.*"

Lines 675-685: "*More detailed interpretations were grouped into four aspects: (1) AODs are calculated via Mie theory using refractive indices of different numbers (5, 6 and 10) of aerosol species group in different coupled models (WRF-CMAQ, WRF-Chem and WRF-CHIMERE) (Tables S5–S6); (2) 7 (294.6, 303.2, 310.0, 316.4, 333.1, 382.0 and 607.7 nm), 4 (300, 400, 600 and 999 nm), and 5 (200, 300, 400, 600, and 999 nm) effective wavelengths are used in calculating actinic fluxes and photolysis rates in Fast-JX photolysis modules of WRF-CMAQ, WRF-Chem and WRF-CHIMERE, respectively;(3) Different calculating methods of aerosol and cloud optical properties exist in the Fast-JX schemes of three coupled models (Tables S1 and S5–S6); (4) 77, 52 and 40 gas-phase species involve 218, 132, 120 gas-phase reactions in CB6, CBMZ and MELCHIOR2 mechanisms, respectively.*"

The added references were listed as follows.

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