

1 Inter-comparison of multiple two-way coupled meteorology and air quality models (WRF
2 v4.1.1-CMAQ v5.3.1, WRF-Chem v4.1.1, and WRF v3.7.1-CHIMERE v2020r1) in
3 eastern China

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16
17 **Abstract**

18 Two-way coupled meteorology and air quality models, which account for aerosol–
19 radiation–cloud interactions, have been employed to simulate meteorology and air quality
20 more realistically. Although numerous related studies have been conducted, none
21 compared the performances of multiple two-way coupled models in simulating
22 meteorology and air quality over eastern China. Thus, we systematically evaluated annual
23 and seasonal meteorological and air quality variables simulated by three open-sourced,
24 widely utilized two-way coupled models (Weather Research and Forecasting (WRF)–
25 Community Multiscale Air Quality (WRF–CMAQ), WRF coupled with chemistry
26 (WRF–Chem), and WRF coupled with a regional chemistry-transport model named
27 CHIMERE (WRF–CHIMERE)) by validating their results with surface and satellite
28 observations for eastern China in 2017. Although we have made every effort to evaluate
29 these three coupled models under configurations as consistent as possible, there are still
30 unavoidable differences in the treatments of physical and chemical processes in them.
31 Our thorough evaluations revealed that all three two-way coupled models reasonably
32 captured the annual and seasonal spatiotemporal characteristics of meteorology and air
33 quality. Notably, the roles of aerosol–cloud interaction (ACI) in improving the models’
34 performances were limited compared to those of aerosol–radiation interaction (ARI). The
35 sources of uncertainties and bias among the different ACI schemes in the two-way
36 coupled models were identified. With sufficient computational resources, these models
37 can provide more accurate air quality forecasting to support atmospheric environment
38 management and deliver timely warning of heavy air pollution events. Finally, we
39 proposed potential improvements of two-way coupled models for future research.

41 1 Introduction

42 Aerosols in the atmosphere due to anthropogenic and nature emissions not only
43 cause air pollution but also induce climate and meteorological impacts through
44 aerosol-radiation interaction and aerosol-cloud interaction (Carslaw et al., 2010;
45 Rosenfeld et al., 2014; Fan et al., 2016; IPCC, 2021). The feedbacks of aerosols to
46 meteorology have been widely investigated by two-way coupled meteorology and air
47 quality models in the past two decades (Jacobson, 1994, 1997, 1998, 2001, 2002; Grell et
48 al., 2005; Wong et al., 2012; Wang et al., 2014; Zhou et al., 2016; Briant et al., 2017;
49 Feng et al., 2021). In these models, two-way interactions between meteorology and
50 aerosols are enabled by including all the processes involving ARI or/and ACI (Grell and
51 Baklanov, 2011; Wang et al., 2014; Briant et al., 2017; Wang et al., 2021). The
52 fundamental theories, modeling technics, developments, and applications of two-way
53 coupled meteorology and air quality models in North America, Europe and Asia have
54 been systemically reviewed (Zhang, 2008; Baklanov et al., 2014; Gao et al., 2022).

55 As pointed out by these review papers, the treatments and parameterization schemes
56 of all the physiochemical processes involving ARI and ACI can be very different in
57 two-way coupled models, so that the simulation results from these models could vary in
58 many aspects. At the same time, the configurations of coupled models, such as
59 meteorological and chemical initial and boundary conditions (ICs and BCs), horizontal
60 and vertical resolutions, and emission inventories and processing tools, etc., play
61 important roles in models' simulations. In the past, model inter-comparison projects have
62 been carried out targeting various two-way coupled meteorology and air quality models.
63 For example, the Air Quality Model Evaluation International Initiative Phase II focused
64 on the performance of multiple two-way coupled models and the effects of aerosol
65 feedbacks in Europe and the United States (Brunner et al., 2015; Im et al., 2015a, b;
66 Makar et al., 2015a, b). In Asia, the Model Inter-Comparison Study for Asia Phase III
67 was conducted to evaluate ozone (O_3) and other gaseous pollutants, fine particular matter
68 ($PM_{2.5}$), and acid and reactive nitrogen deposition with various models with/without ARI
69 or/and ACI (Li et al., 2019; Chen et al., 2019; Itahashi et al., 2020; Ge et al. al., 2020;
70 Kong et al., 2020). With respect to this project, Gao et al. (2018, 2020) have reviewed in
71 detail the model performance of seven two-way coupled models from different research
72 groups in simulating a heavy air pollution episode during January 2010 in North China
73 Plain and how aerosol feedbacks affected simulations of meteorological variables and
74 $PM_{2.5}$ concentrations. Targeting the heavy polluted India region, Govardhan et al. (2016)
75 compared aerosol optical depth (AOD) and various aerosol species (black carbon,
76 mineral dust, and sea salt) modeled by WRF-Chem (with ARI) and Spectral
77 Radiation-Transport Model for Aerosol Species (with both ARI and ACI), but under
78 different model configurations.

79 So far, there is no comprehensive comparisons of multiple coupled models under the
80 same model configuration with respect to the high aerosol loading region over eastern
81 China, where has experienced rapid growth of economy, urbanization, population, as well

82 as severe air quality problems in the past decades (He et al., 2002; Wang and Hao, 2012;
83 Gao et al., 2017; Geng et al., 2021). In the eastern China region (ECR), several
84 open-source and proprietary two-way coupled models have been applied to investigate
85 the ARI and/or ACI effects, yet most studies have focused on certain short-term episodes
86 of heavy air pollution without any year-long simulations (Xing et al., 2017; Ding et al.,
87 2019; Ma et al., 2021). The commonly used open-source models in ECR are WRF-Chem
88 and WRF-CMAQ (Grell et al., 2005; Wong et al., 2012), but there is no any application
89 of the two-way coupled WRF-CHIMERE model that has been applied to examine
90 aerosol-radiation-cloud interactions in Europe and Africa (Briant et al., 2017; Tuccella et
91 al., 2019). At the same time, model simulations should be compared not only against
92 surface measurement data but also satellite data (Zhao et al., 2017; Hong et al., 2017;
93 Campbell et al., 2017; Wang et al., 2018). Even though the running time of an individual
94 modeling system (e.g., WRF-CMAQ and WRF-CHIMERE) was evaluated by
95 considering its online and offline versions and under various computing configurations
96 (Wong et al., 2012; Briant et al., 2017), the computational efficiencies of multiple
97 two-way coupled models need to be accessed under the same computing conditions as
98 well.

99 In this paper, a comparative evaluation of three open-sourced two-way coupled
100 meteorology and air quality models (WRF-CMAQ, WRF-Chem and WRF-CHIMERE) in
101 ECR is conducted. The remainder of the paper is organized as follows: Section 2
102 describes the study methods including model configurations and evaluation protocols.
103 Sections 3 and 4 presents the analyses and intercomparisons of simulations from these
104 three two-way coupled models with regard to meteorology and air quality, respectively.
105 The major findings of this work are summarized in Section 5.

106

107 2 Data and methods

108 2.1 Model configurations and data sources

109 One-year simulations of meteorology and air quality in eastern China were
110 examined using the two-way coupled WRF-CMAQ, WRF-Chem, and WRF-CHIMERE
111 models with and without enabling ARI and/or ACI, as well as with a 27 km horizontal
112 grid resolution (the east-west direction comprised 110, 120, and 120 grid cells, and the
113 north-south direction 150, 160, and 170 grid cells for the WRF-CMAQ, WRF-Chem,
114 and WRF-CHIMERE models, respectively). All the three coupled models used in this
115 study have 30 levels (i.e., 29 layers) from the surface to 100 hPa with 11 layers in the
116 bottom 1 km and the bottom-layer thickness being 23.2 m. The anthropogenic emissions
117 of Multiresolution Emission Inventory for China (MEIC) (Li et al., 2017) and the Fire
118 INventory from NCAR, version 1.5 (FINN v1.5), biomass burning emissions
119 (Wiedinmyer et al., 2011) were considered in our simulations, and their spatial, temporal,
120 and species allocations were performed using Python (Wang et al., 2023). Biogenic
121 emissions were calculated using the Model of Emissions of Gases and Aerosols from

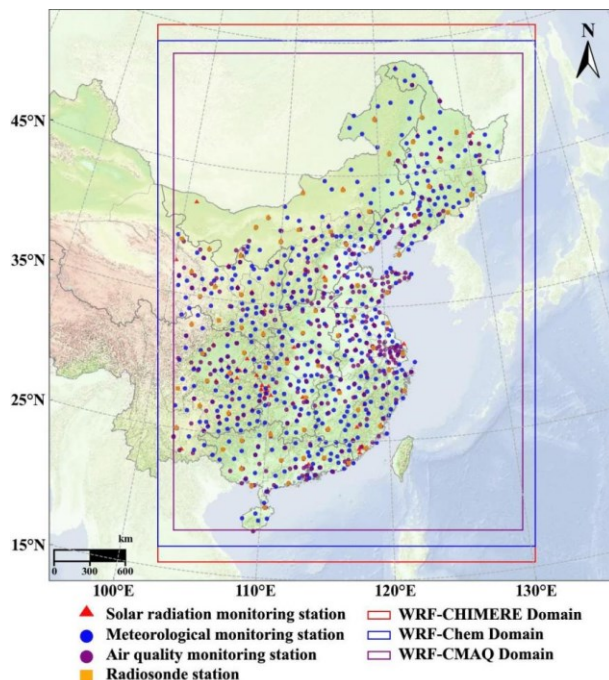
122 Nature, version 3.0 (MEGAN v3.0; Gao et al., 2019). Dust and sea salt emissions were
123 used in the calculations of the inline modules (Table 1). The meteorological ICs and
124 lateral BCs were derived from the National Center for Environmental Prediction Final
125 Analysis (NCEP-FNL) datasets (<http://rda.ucar.edu/datasets/ds083.2>), with a horizontal
126 resolution of $1^\circ \times 1^\circ$ at 6-hour intervals for each of the three coupled models, and the flux
127 in the model-top boundary was set to zero. To improve the long-term accuracy of the
128 meteorological variables when utilizing the WRF model, we turned on the options of
129 observational and grid four-dimensional data assimilation (FDDA), and pressure, station
130 height, relative humidity, wind speed (WS), and wind direction were observed four times
131 per day at 00:00, 06:00, 12:00, and 18:00 UTC from 2,168 stations
132 (<https://doi.org/10.5281/zenodo.6975602>, Gao et al., 2022). Notably, turning on FDDA in
133 two-way coupled models could dampen the simulated aerosol feedback (Wong et al.,
134 2012; Forkel et al., 2012; Hogrefe et al., 2015; Zhang et al., 2016). To mitigate the effects
135 of turning on FDDA on aerosol feedback in long-term simulations, we set the nudging
136 coefficients of the u/v wind, temperature, and water vapor mixing ratio above the
137 planetary boundary layer to 0.0001, 0.0001, and 0.00001 s^{-1} , respectively. The chemical
138 ICs/lateral BCs were downscaled from the whole atmosphere community climate model
139 (WACCM) for WRF-CMAQ and WRF-Chem using the `mozart2camx` and `mozbc` tools,
140 respectively. WRF-CHIMERE employed the climatology from a general circulation
141 model developed at the Laboratoire de Météorologie Dynamique (LMDz) coupling a
142 global chemistry and aerosol model INteractions between Chemistry and Aerosols (INCA;
143 Mailler et al., 2017). For chemical model-top BCs, the WRF-CMAQ and WRF-Chem
144 models consider the impacts of stratosphere-troposphere O_3 exchange using O_3 -potential
145 vorticity parameterization (Safieddine et al., 2014; Xing et al., 2016). The related options
146 of both models were used in this study. WRF-CHIMERE employs the climatology from
147 the LMDz-INCA data (Mailler et al., 2017).

148 Table 1 lists the options of parameterization schemes of aerosol-radiation-cloud
149 interactions. To maintain the consistency of physical schemes, the same Rapid Radiative
150 Transfer Model for General Circulation Models Applications (RRTMG) short-wave (SW)
151 and long-wave (LW) radiation schemes and the Morrison microphysics scheme were
152 adopted in the WRF-Chem and WRF-CMAQ models. WRF-CHIMERE applies the
153 same radiation schemes, as well as the Thompson microphysics scheme. The other
154 different schemes (cumulus, surface, and land surface) in the WRF-CMAQ and WRF-
155 Chem models were selected, following Gao et al.'s (2022) widely utilized options
156 outlined in Table S1. The other schemes employed in WRF-CHIMERE are the same as
157 those in WRF-Chem. To consider the effects of clouds on radiative transfer calculations,
158 the fractional cloud cover and cloud optical properties were included in the RRTMG
159 SW/LW radiation schemes employed in the three coupled models (Xu and Randall, 1996;
160 Iacono et al., 2008). The coupled WRF-CMAQ model with the Kain-Fritsch cumulus
161 scheme included the impacts of cumulus cloud fraction (CF) on RRTMG radiation
162 (Alapaty et al., 2012), whereas the WRF-Chem and WRF-CHIMERE models with the

163 Grell–Freitas cumulus scheme did not. In the Fast-JX photolysis scheme employed by the
164 three coupled models, the impacts of clouds were included by considering the cloud
165 cover and cloud optical properties. However, the calculations of the cloud cover and
166 cloud optical properties differed in these models, and Table S1 presents the relevant
167 information. Regarding the aerosol-size distribution, we used the modal approach with
168 Aitken, accumulation, and coarse modes in WRF–CMAQ, as well as the 4 and 10 bin
169 sectional approaches in the WRF–Chem and WRF–CHIMERE models, respectively
170 (Binkowski and Roselle, 2003; Zaveri et al., 2008; Nicholls et al., 2014; Menut et al.,
171 2013, 2016).

172 To demonstrate the capabilities of the three two-way coupled models with/without
173 aerosol feedbacks in simulating meteorology and air quality, we comprehensively
174 evaluated the strengths and weaknesses of each coupled model and validated them
175 against extensive ground-based and satellite measurements. The ground-based data
176 included 572 hourly ground-based meteorological observations (air temperature (T2) and
177 relative humidity (RH2) at 2 m above the surface, WS at 10 m above the surface (WS10),
178 and precipitation (PREC)) (<http://data.cma.cn>); 327 hourly national environmental
179 observations [fine particulate matter (PM_{2.5}), ozone (O₃), nitrogen dioxide (NO₂), sulfur
180 dioxide (SO₂), and carbon monoxide (CO)] (<http://106.37.208.233:20035>); 109 hourly
181 surface SW radiation (SSR) measurements (Tang et al., 2019); and 74 radiosonde data
182 retrieved two times per day, which used to calculate planetary boundary layer height at
183 08:00 and 20:00 local time (PBLH08 and PBLH12) (Guo et al., 2019). Figure 1 shows
184 the locations of these data. Because there were no observed water vapor mixing ratio (w)
185 data, this parameter was calculated by $w = \frac{rh}{w_s}$, where rh is the relative humidity and w_s
186 is the saturation mixing ratio (Wallace and Hobbs, 2006).

187 The satellite data included the following: monthly average downwelling SW/LW
188 flux at the surface and SW/LW flux at the top of the atmosphere (TOA) obtained from the
189 clouds and the Earth’s radiant energy system (CERES) (<https://ceres.larc.nasa.gov>);
190 PREC from the Tropical Rainfall Measuring Mission (TRMM); CF, liquid-water path
191 (LWP), and AOD from the Moderate Resolution Imaging Spectroradiometer (MODIS);
192 tropospheric NO₂ and SO₂ columns in the planetary boundary layer (PBL) from the
193 Ozone Monitoring Instrument; total CO column from the Measurements of Pollution in
194 the Troposphere (<https://giovanni.gsfc.nasa.gov/giovanni>); total column ozone (TCO)
195 from the Infrared Atmospheric Sounding Interferometer-Meteorological Operational
196 Satellite-A (IASI-METOP-A) (<https://cds.climate.copernicus.eu/cdsapp#!/dataset/satellite-ozone?tab=form>); and total
197 ammonia (NH₃) column from IASI-METOP-B
198 (https://cds-espri.ipsl.fr/iasib13/iasi_nh3/V3.1.0). These data were downloaded and
199 interpolated to the same horizontal resolution as the model results using the Rasterio
200 library (Gillies et al., 2013). Thereafter, the model and observed values at each grid point
201 were extracted.
202
203



204

205 Figure 1. Modeling domains (WRF-CMAQ, WRF-Chem, and WRF-CHIMERE) and solar radiation,
 206 meteorology, air quality, and radiosonde stations.

207

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

		WRF-CMAQ	WRF-Chem	WRF-CHIMERE
Domain	Horizontal grid spacing	27 km (110 × 150)	27 km (120 × 160)	27 km (120 × 170)
configuration	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

Input data	Biomass burning emission	FINN v1.5	FINN v1.5	FINN v1.5
	Dust emission	Foroutan	GOCART	Menut
	Sea-salt emission	Gong	Gong	Monahan
	Meteorological ICs and BCs	FNL	FNL	FNL
	Chemical ICs and BCs	MOZART	MOZART	LMDZ-INCA

210

211 2.2 Scenario setup

212 To comprehensively assess the performances of WRF v4.1.1–CMAQ v5.3.1, WRF–
213 Chem v4.1.1, and WRF v3.7.1–CHIMERE v2020r1 and performances affected by
214 aerosol feedbacks over eastern China in 2017, eight sets of annual hindcast simulations
215 with/without ARI and/or ACI were performed (Table 2). Compared with WRF v4.1.1–
216 CMAQ v5.3.1 and WRF–Chem v4.1.1, this WRF v3.7.1–CHIMERE v2020r1 version can
217 be officially obtained, and a higher version of WRF–CHIMERE has not been developed.
218 Notably, the official WRF–Chem and WRF–CHIMERE can execute the simulation of
219 ARI and ACI, whereas WRF–CMAQ cannot. In all of the simulations performed in this
220 study, a spin-up time of one month was set up to reduce the influence of the initial
221 conditions. Multiple statistical metrics, including the correlation coefficient (R), mean
222 bias (MB), normalized mean bias (NMB), normalized gross error (NGE), and root mean
223 square error (RMSE), were used between each scenario simulation and
224 ground-based/satellite-borne observations. The mathematical definitions of these metrics
225 are provided in Supplementary Information (SI) S1. To compare the simulations by the
226 three coupled models, the respective model configurations of the physics and chemistry
227 routines were set as consistent as possible. We systemically analyzed the annual and
228 seasonal statistical metrics of the meteorological and air quality variables, including
229 simulations by the three two-way coupled models with/without the ARI and/or ACI
230 effects. Thereafter, we quantified the respective contributions of the ARI and ACI effects
231 to model performance.

232 Table 2. Summary of scenario settings in the three coupled models.

Model	Scenario	Configuration option	Description
WRF-CMAQ	(1) WRF-CMAQ_NO	DO_SW_CAL=F	Without aerosol feedbacks
	(2) WRF-CMAQ_ARI	DO_SW_CAL=T	ARI
WRF-Chem	(3) WRF-Chem_NO	aer_ra_feedback=0 wetscav_onoff=0 cldchem_onoff=0	Without aerosol feedbacks
	(4) WRF-Chem_ARI	aer_ra_feedback=1 wetscav_onoff=0 cldchem_onoff=0	ARI
	(5) WRF-Chem_BOTH	aer_ra_feedback=1 wetscav_onoff=1	ARI and ACI

		cldchem_onoff=1	
WRF-CHIMERE	(6) WRF-CHIMERE_NO	direct_feed_chimere=0	Without aerosol feedbacks
		indirect_feed_chimere=0	
	(7) WRF-CHIMERE_ARI	direct_feed_chimere=1	ARI
		indirect_feed_chimere=0	
	(8) WRF-CHIMERE_BOTH	direct_feed_chimere=1	ARI and ACI
		indirect_feed_chimere=1	

233

234 3 Multimodel meteorological evaluations

235 This section presents the annual and seasonal (March–May, Spring; June–August,
 236 Summer; September–November, Autumn; and December–February, Winter) statistical
 237 metrics of the simulated meteorological variables and air quality, as well as their
 238 comparison with the ground-based and satellite observations. The running times of the
 239 eight simulation scenarios are also discussed.

240 3.1 Ground-based observations

241 Figures 2 and S1–S7 show the spatial distributions of R, MB, and RMSE for hourly
 242 SSR, T2, Q2, RH2, WS10, PREC, PBLH08, and PBLH120 from WRF–CMAQ, WRF–
 243 Chem, and WRF–CHIMERE with/without turning on aerosol feedback against
 244 ground-based observations from each site throughout 2017. The calculated annual model
 245 evaluation metrics for all sites in eastern China are summarized in Table S1, and the
 246 related seasonal R and MB values are presented in Fig. 3. Here, we mainly focused on the
 247 comparisons of SSR, T2, RH2, and WS10. Further, Section 1.1 of SI presents the
 248 analyses of PREC, PBLH08, and PBLH20.

249 The accuracy of radiation prediction is of great significance in ARI evaluation. The
 250 annual and seasonal average simulated SSR data were compared with the ground-based
 251 observations (Figs. 3–4 and Table S3), and SSR over eastern China was simulated very
 252 reasonably by the models, with R-values of 0.61–0.78. The simulated results were
 253 overestimated on the annual and seasonal scales (MBs in spring and summer were larger
 254 than those in autumn and winter). The overestimated annual SSRs were 19.98, 14.48, and
 255 9.24 W m⁻² for WRF–CMAQ, WRF–Chem, and WRF–CHIMERE, respectively. Brunner
 256 et al.’s (2015) comparative study also reported that most two-way coupled models
 257 overestimated SSR for Europe and North America. Such overestimations could be caused
 258 by multiple factors, namely, the uncertainties in cloud development owing to PBL and
 259 convection parameterizations (Alapaty et al., 2012) and the diversity in the treatment of
 260 land-surface processes (Brunner et al., 2015), which tend to play more important roles
 261 than the enabling of the two-way aerosol feedbacks on SSR through the effects of ARI
 262 and ACI. When the three models incorporated the ARI effects, the simulation accuracies
 263 for SSR over the whole year and four seasons improved, although the enabling of the ACI
 264 effects resulted in relatively limited improvement. Additionally, the MB variations of
 265 WRF–CMAQ and WRF–Chem simulations were higher in spring and winter than in

266 summer and autumn, whereas the maximum and minimum MBs of WRF–CHIMERE
267 simulations were obtained in summer (-10.33 W m^{-2}) and autumn (-7.64 W m^{-2}),
268 respectively. The annual and seasonal decrease in SSR simulated by WRF–Chem and
269 WRF–CHIMERE with enabled ACI effects were significantly smaller than those with
270 enabled ARI effects.

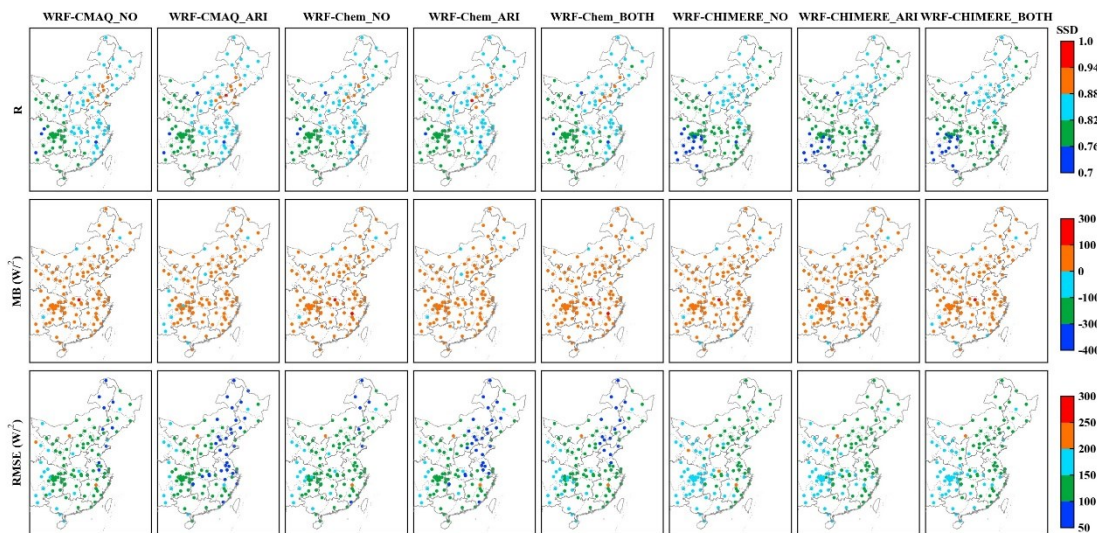
271 Generally, the simulated magnitudes and temporal variations in the air temperature
272 at 2 m above the ground exhibited high consistency with the observations ($R = 0.88$ –
273 0.97). Considering the annual and seasonal T2, the models tended to display a negative
274 bias, and the T2 underestimations in spring and winter were greater than those in summer
275 and autumn (Figs. 3 and 4). Following Makar et al. (2015a), WRF–Chem and GEM–
276 MACH produced negative MBs in summer and positive MBs in winter with enabled ACI
277 and ARI effects; additionally, WRF–CMAQ with only the ARI effects enabled produced
278 negative MBs in summer over North America in 2010. Notably, Makar et al.’s. (2015a)
279 study lacked winter meteorology evaluations using WRF–CMAQ. The comparison
280 results of MBs revealed the following order: WRF–CHIMERE > WRF–CMAQ > WRF–
281 Chem. The annual and seasonal MBs of WRF–CMAQ and WRF–Chem were
282 approximately -1°C , whereas that of WRF–CHIMERE ranged from -2 to -1°C . The
283 RMSE values of WRF–CMAQ (2.71 – 3.05°C) and WRF–Chem (2.82 – 3.27°C) were
284 almost equal. Those of WRF–CHIMERE (3.39 – 4.53°C) were larger on the annual and
285 seasonal scales. Notably, reduced underestimations of the annual and seasonal T2 by the
286 three coupled models were observed in eastern China when the ARI effects were enabled.
287 With the enabled ACI effects, the MBs for T2 simulated by WRF–Chem_BOTH did not
288 change significantly compared with those of WRF–Chem_NO; additionally, compared
289 with WRF–CHIMERE_NO, WRF–CHIMERE_BOTH further enhanced the
290 underestimations of T2 in the full year (-1.30°C), spring (-0.12°C), and winter
291 (-0.40°C).

292 Regarding RH2, the annual and seasonal simulations using WRF–CMAQ exhibited
293 the highest correlation with the observed values, followed by WRF–Chem and WRF–
294 CHIMERE, and the smallest correlation coefficients of the three models were observed in
295 autumn (~ 0.5). The spatial MBs between the simulations using the three models and
296 observations displayed a general converse trend compared with T2 (i.e., RH2 was
297 overestimated where T2 was underestimated, and vice versa). This can be explained by
298 calculating RH2 based on T2 in the models (Wang et al., 2021). The annual and seasonal
299 MBs were 0.65% – 71.03% and -21.30% to 60.00% , respectively (Fig. 4 and Table S3);
300 only WRF–Chem produced negative MBs in the summer. The magnitude of RMSE
301 exhibited an inverse pattern compared with R for the three models, with maximum
302 (28.48% – 29.52%) and minimum (12.57% – 16.07%) values observed in autumn and
303 summer, respectively. Figs. 3–4 and Table S3 show that WRF–CMAQ_ARI further
304 reduced the overestimations of the annual and seasonal RH2 in ECR, whereas WRF–
305 Chem_ARI (except for summer) and WRF–CHIMERE_ARI displayed the opposite trend.
306 Moreover, the variations in the annual and seasonal RH2 MBs simulated by WRF–

307 Chem_BOTH and WRF-CHIMERE_BOTH were further reduced compared with those
308 simulated by WRF-Chem_ARI (except for summer) and WRF-CHIMERE_ARI,
309 respectively.

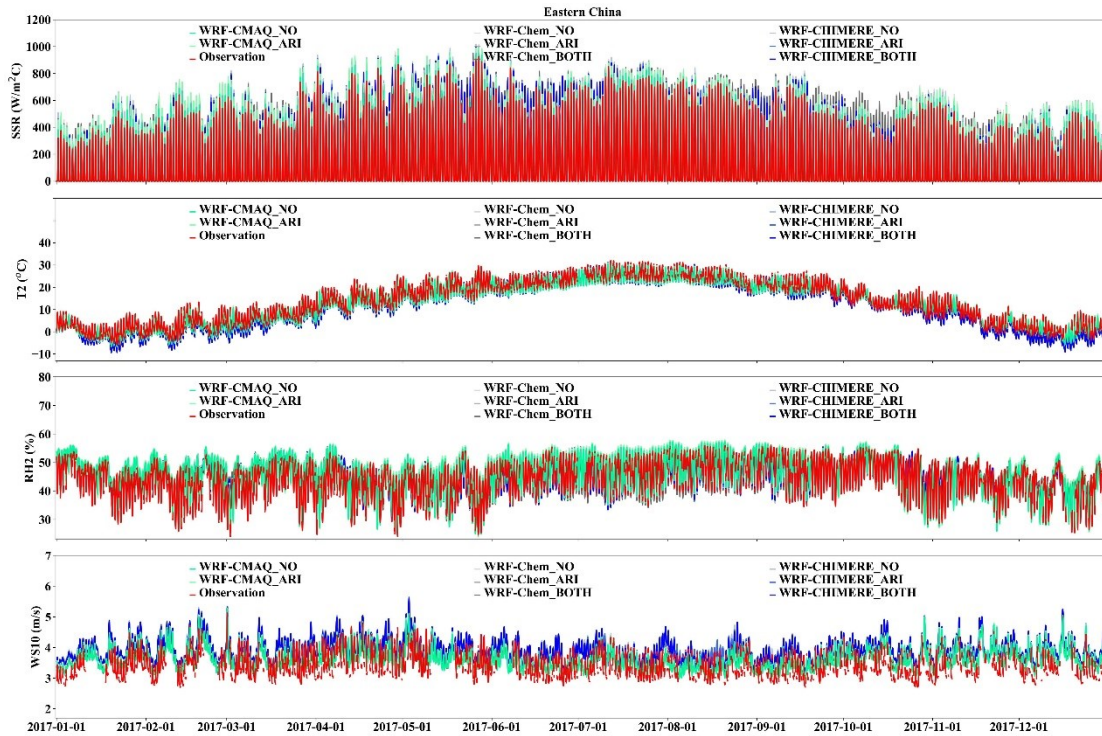
310 Furthermore, similar analyses were performed for WS10, and the results revealed
311 that WRF-CMAQ performed better in capturing the WS10 patterns than WRF-Chem and
312 WRF-CHIMERE. The R-values for all three models ranged from 0.47 to 0.60; WRF-
313 CMAQ and WRF-Chem overestimated WS by $\sim 0.5 \text{ m s}^{-1}$, whereas WRF-CHIMERE
314 overestimated it by $\sim 1.0 \text{ m s}^{-1}$ (Table S3 and Figs. 3-4). The overestimation of WS10
315 under real-world low-wind conditions is a common phenomenon of existing weather
316 models, and it is mainly caused by outdated geographic data, coarse model resolution,
317 and a lack of good physical representation of the urban canopy (Gao et al., 2015, 2018).
318 The three models exhibited lower correlations (0.31-0.54) and MBs (0.20-0.86 m s^{-1}) in
319 summer compared with the other seasons, and the RMSEs were $\sim 2.0 \text{ m s}^{-1}$. Enabling the
320 ARI effects mitigated the overestimations of the three models, particularly WRF-
321 CMAQ_ARI.

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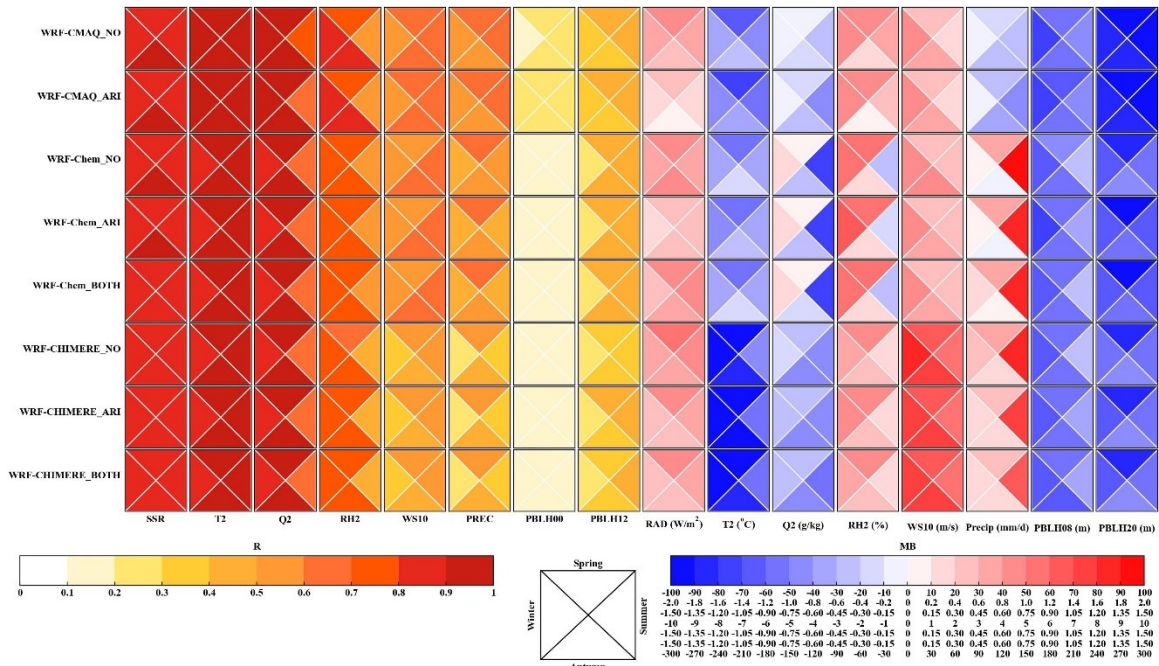
324 Figure 2. Statistical metrics (R, MB, and RMSE) for annual simulations and observations of SSR in
325 eastern China.



326

327 Figure 3. Time series of the observed and simulated hourly SSR, T2, RH2, and WS10 by coupled
 328 WRF-CMAQ, WRF-Chem, and WRF-CHIMERE with/without aerosol feedbacks over ECR in 2017.

329



330

331 Figure 4. Portrait plots of the statistical indices (R and MB) between the seasonal simulations and
 332 surface observations of the meteorological variables (SSR, T2, Q2, RH2, WS10, PREC, and PBLH at

333 08:00 and 20:00 LT) in ECR.

334 To determine and quantify how well our results compared with those of the extant
335 studies using two-way coupled models, we compared our study with previous ones in
336 terms of the evaluation results of meteorology and air quality. We discussed meteorology
337 and air quality in this section and Section 4.1, respectively. We employed
338 box-and-whisker plots, and the 5th, 25th, 75th, and 95th percentiles were used as statistical
339 indicators. In the plots, the dashed lines in the boxes represent the mean values, and the
340 circles represent outliers. Previous studies mainly used WRF–Chem and WRF–CMAQ to
341 evaluate meteorology and air quality, whereas the WRF–NAQPMS and GRAPES–
342 CUACE barely had application potential. As mentioned in Section 1, previous
343 investigations of meteorology and air quality using WRF–CHIMERE with/without
344 aerosol feedbacks have not been conducted in ECR. Therefore, only the evaluation results
345 involving WRF–Chem and WRF–CMAQ were analyzed to study aerosol feedbacks.

346 Figure S8 shows the comparison between the statistical metrics, T2, RH2, Q2, and
347 WS10, in this study and the evaluation results of previous studies. Based on the number
348 of samples in the statistical metrics of each meteorological variable, most previous
349 studies mainly involved the simulation and evaluation of T2, WS10, and RH2, with only
350 a relatively few studies focusing on Q2. Compared with the evaluation results of the
351 extant studies, the ranges of our statistical metrics were roughly similar, although there
352 were some notable differences. The R-values of the WRF–CMAQ and WRF–Chem
353 models in our study were higher than those of the previous studies; MBs of T2 simulated
354 by WRF–CMAQ were smaller, whereas those of T2 simulated by WRF–Chem were
355 larger; and RMSEs of the WRF–CMAQ simulation were larger, whereas those of the
356 WRF–Chem simulation were smaller. For RH2, the R-values for our WRF–CMAQ and
357 WRF–Chem were larger than the average level of the previous studies, whereas MBs and
358 RMSEs for WRF–CMAQ were larger. Those for WRF–Chem were smaller than the
359 average reported in previous studies. For Q2, the model performance of WRF–CMAQ in
360 this study was generally better than the average level reported in previous studies,
361 although the R-value between the WRF–Chem simulation results and observed values
362 was higher (and MB and RMSE were lower) than the average level reported in previous
363 studies. We also conclude that the simulation results of our WRF–CMAQ and WRF–
364 Chem better reproduced the variations in WS10 compared with the simulation reported
365 by previous studies.

366

367 3.2 Satellite-borne observations

368 To further evaluate the performances of WRF–CMAQ, WRF–Chem, and WRF–
369 CHIMERE against the satellite observations, we analyzed the annual and seasonal
370 statistical metrics of SW and LW radiations at the surface, PREC, cloud cover, and LWP
371 simulated by the three coupled models with and without aerosol feedbacks by
372 comparisons the simulations with the satellite-borne observations (Table 3 and Figs. 5, S9,
373 and S12–S14). Additionally, evaluations of SW and LW radiation at TOA are presented in

374 Section 1.2 of SI.

375 As shown in Table 3 and Fig. 5, the three coupled models exhibited relatively poor
376 performances for SSR, with annual MBs of 8.21–30.74 W m⁻² and correlations of 0.61–
377 0.78. A similar poor performance for SW radiation was also reported in the United States
378 using the coupled WRF–CMAQ and offline WRF models (Wang et al., 2021). The
379 overall seasonal characteristics of SSR were reproduced by the three coupled models (Fig.
380 S10). Concurrently, regardless of whether aerosol feedbacks were enabled or not, the
381 three models overestimated seasonal SSR (except WRF–Chem_ARI in winter), obtaining
382 higher MBs in spring and summer than in autumn and winter. The seasonal SSR
383 overestimations might be directly due to the underestimation of the calculated AOD when
384 examining the ARI effects (Wang et al., 2021). Compared with SSR, the three coupled
385 models predicted the surface LW radiation variables (SLR) well (R-values were up to
386 0.99), with annual domain-average MBs of –9.97 to –6.05 W m⁻². Furthermore,
387 significant seasonal differences were observed in the simulated LW radiation by the three
388 coupled models; the WRF–CMAQ and WRF–CHIMERE scenarios yielded
389 underestimations, with maximum and minimum SLR values in winter and summer,
390 respectively, whereas the maximum underestimations of WRF–Chem were recorded in
391 autumn, particularly for WRF–Chem_BOTH (Fig. S9).

392 As the three coupled models adopted the same grid resolution (27 × 27 km) as well
393 as SW and LW radiation schemes (RRTMG), the above analysis demonstrated that the
394 configuration differences among the aerosol components, size distributions, and
395 mechanisms contributed to the diverse seasonal MBs (Tables 1 and S2). Moreover, the
396 three two-way coupled models with ARI feedbacks effectively improved the
397 performances of annual and seasonal SSR; however, for SLR, the performance
398 improvements were much more variable across the three coupled models and different
399 scenarios with and without ARI and/or ACI feedbacks enabled (Table S4). When the ARI
400 effects were enabled, the diverse refractive indices of the aerosol species groups caused
401 discrepancies in the online calculated aerosol optical properties in different SW and LW
402 bands in the RRTMG SW/LW radiation schemes of WRF–CMAQ, WRF–Chem, and
403 WRF–CHIMERE (Tables S5–S6). The online calculated cloud optical properties induced
404 by aerosol absorption in the RRTMG radiation schemes differed regarding their
405 treatments of the aerosol species groups in the three coupled models. With the ACI effects
406 enabled, the activation of cloud droplets from aerosols based on the Köhler theory was
407 considered in WRF–Chem and WRF–CHIMERE compared with the simulations without
408 aerosol feedbacks (Table S7). The treatments of prognostic ice-nucleating particles (INP)
409 formed via the heterogeneous nucleation of dust particles (diameters > 0.5 μm) and
410 homogeneous freezing of hygroscopic aerosols (diameters > 0.1 μm) were only
411 investigated in WRF–CHIMERE, whereas the prognostic INP were not included in
412 WRF–CMAQ and WRF–Chem. These discrepancies eventually contributed to the
413 differences in the simulated radiation changes caused by aerosols.

414 From IPCC 2007–2021, the effects of aerosol feedbacks (particularly with the ACI

415 effects enabled) on PREC and cloud processes remained unclear. In this study, we further
416 assessed the annual and seasonal simulated PREC, cloud cover, and LWP in ECR with
417 high aerosol loadings against the satellite observations (Table 3 and Figs. S12–S14) to
418 provide new insights into enabling online feedbacks in two-way coupled modeling
419 simulations from a yearly perspective.

420 The results indicated that PREC simulated by WRF–CMAQ (0.51–0.89) exhibited
421 higher correlations than those simulated by WRF–Chem (0.61–0.73) and WRF–
422 CHIMERE (0.54–0.70). WRF–CMAQ demonstrated the best correlation in winter,
423 whereas WRF–Chem and WRF–CHIMERE had the best correlation in spring; the three
424 models presented their worst correlations in summer, as the numerical models struggled
425 to effectively capture enhanced convective activities in summer. Huang and Gao (2018)
426 also revealed that the accurate representations of lateral boundaries were crucial to
427 improving PREC simulations in China during summer. WRF–CMAQ underestimated
428 annual PREC, with MBs of -76.49 to -51.93 mm, whereas WRF–Chem and WRF–
429 CHIMERE produced large PREC overestimations ranging from $+108.04$ to $+207.05$ mm
430 (Table 3), particularly in southern China regions (Fig. S11). WRF–CMAQ also produced
431 negative biases (-27.89 to $+42.08$ mm) for seasonal PREC, except for WRF–
432 CMAQ_ARI in winter. WRF–Chem and WRF–CHIMERE only underestimated seasonal
433 PREC in autumn (-31.39 to -26.89 mm) and winter (-7.12 to -4.43 mm), respectively
434 (Fig. S12). The variations in the annual and seasonal MBs of PREC were consistent with
435 the changes in CF and LWP (Zhang et al., 2016), and these changes will be discussed in
436 detail below.

437 By considering aerosol feedbacks, the ARI-induced decrease in annual MBs of
438 PREC for WRF–CMAQ, WRF–Chem, and WRF–CHIMERE were 24.56, 12.11, and
439 4.70 mm, respectively. WRF–Chem_BOTH (24.9 mm) and WRF–CHIMERE_BOTH
440 (3.41 mm) enhanced the overestimation of annual PREC compared with WRF–
441 Chem_ARI and WRF–CHIMERE_ARI, respectively. Significant increases ($+53.15$ mm)
442 and decreases (-6.3 to -3.41 mm) in MBs were facilitated by WRF–CMAQ and the other
443 two models with ARI effects enabled compared with those without feedbacks,
444 respectively. WRF–Chem and WRF–CHIMERE with ARI and ACI effects enabled
445 produced larger MB enhancements ($+3.54$ to $+7.46$ mm) on the seasonal scale (Fig. S12).
446 Notably, the discrepancies in simulated PREC were mainly attributable to the selection of
447 different microphysics and cumulus schemes in WRF–CMAQ (Morrison and Kain–
448 Fritsch), WRF–Chem (Morrison and Grell–Freitas), and WRF–CHIMERE (Thompson
449 and Grell–Freitas).

450 CF and LWP can significantly influence the spatiotemporal distributions of PREC;
451 our simulated results of annual and seasonal CFs in ECR are presented in Table 3 and Fig.
452 S13. Overall, WRF–CMAQ performed best in simulating CF. The R-values of WRF–
453 Chem during summer (0.69) and winter (0.70) were larger than those of WRF–CMAQ
454 (0.59 and 0.64) and WRF–CHIMERE (0.56 and 0.66), whereas WRF–CMAQ and WRF–
455 CHIMERE obtained better simulation results in winter and autumn, with correlations of

456 up to 0.89 and 0.67, respectively. The three coupled models underestimated annual and
457 seasonal CFs, with MBs of -16.83% to -6.18% and -21.13% to -4.13% , respectively;
458 these results were consistent with those of previous two-way coupled modeling studies
459 using WRF-CMAQ (-19.7%) and WRF-Chem (-32% to -9%) in China (Hong et al.,
460 2017; Zhao et al., 2017). The models reasonably simulated the annual LWP in ECR, with
461 R-values of >0.55 and negative biases varying from -57.36 to -31.29 g m^{-2} . These
462 underestimations were closely related to missing cloud homogeneity (Wang et al., 2015;
463 Dionne et al., 2020) and the excessive conversion of liquid water to ice in the selected
464 cloud microphysics schemes (Klein et al., 2009). As shown in Fig. S14, the models
465 performed best in simulating LWP in spring ($R = 0.51-0.79$), and their highest
466 underestimations were observed in winter (MBs = -54.82 to -40.89 g m^{-2}), except for
467 WRF-Chem, which obtained its maximum bias in autumn.

468 To quantitatively determine the impacts of aerosol feedbacks on CF and LWP, the
469 simulated scenarios revealed that WRF-CMAQ_ARI overwhelmingly decreased the
470 annual and seasonal underestimations of CF ($0.48\%-1.05\%$) and LWP ($3.03-4.29$ g m^{-2}),
471 whereas in WRF-Chem_ARI and WRF-CHIMERE_ARI slightly increased the
472 underestimations (CF: $0.02\%-0.39\%$; LWP: $0.03-0.58$ g m^{-2}). Compared with WRF-
473 CHIMERE_ARI, WRF-CHIMERE_BOTH produced larger variations in the annual and
474 seasonal MBs of CF ($0.23\%-0.93\%$) and LWP (-2.96 to 7.38 g m^{-2}). WRF-
475 Chem_BOTH and WRF-Chem_ARI exhibited equivalent variations (CF: $0.03\%-0.71\%$;
476 LWP: $0.02-2.89$ g m^{-2}). This could be explained by the different parameterization
477 treatments of the cloud droplet number concentration (CDNC) simulated by the three
478 coupled models with/without enabling the ACI effects. The cloud condensation nuclei
479 (CCN) activated by the aerosol particles can increase CDNC and impact LWP and CF.
480 Without enabling any aerosol feedbacks or by enabling only ARI, CDNC is, by default,
481 prescribed as a constant value of 250 cm^{-3} in the Morrison schemes of WRF-CMAQ and
482 WRF-Chem and 300 cm^{-3} in the Thompson schemes of WRF-CHIMERE. With enabling
483 only ACI or both ARI and ACI effects, prognostic CDNC is online calculated in the
484 two-way coupled WRF-Chem and WRF-CHIMERE models when cloud maximum
485 supersaturation is greater than aerosol critical supersaturation (Abdul-Razzak and Ghan,
486 2002; Chapman et al., 2009; Tuccella et al., 2019). Although we have obtained
487 preliminary quantitative results of the ACI effects on regional PREC, CF, and LWP, we
488 acknowledge that several limitations still exist regarding the representation of the ACI
489 effects in state-of-the-art two-way coupled models. These limitations include a lack of
490 consideration for the responses of convective clouds to ACI (Tuccella et al., 2019),
491 numerical descriptions for giant CCN (Wang et al., 2021) and heterogeneous ice nuclei
492 (Keita et al., 2020).

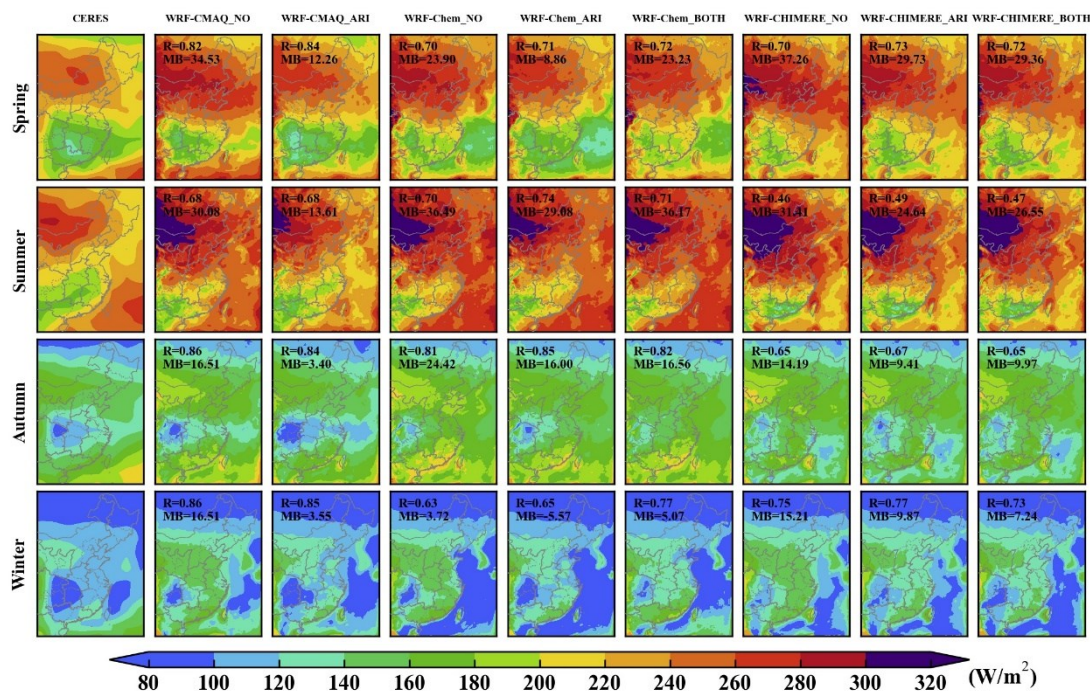
493

494 Table 3. Statistical metrics (R, MB, NMB, NGE, and RMSE) between the annual
495 simulations and satellite retrievals of SSR and SLR, TOA SW and LW radiation, PREC,
496 CF, and LWP in ECR. The best results are captured in bold fonts, and the mean

497 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
Surface shortwave radiation (172.74 W m ⁻²)	Mean_sim	<i>197.15</i>	<i>180.94</i>	<i>203.48</i>	<i>194.52</i>	<i>201.45</i>	<i>197.39</i>	<i>191.34</i>	<i>195.58</i>
	R	0.76	0.75	0.73	0.78	0.75	0.61	0.64	0.66
	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	<i>316.25</i>	<i>315.83</i>	<i>312.96</i>	<i>312.60</i>	<i>312.32</i>	<i>313.33</i>	<i>314.60</i>	<i>314.47</i>
	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	<i>107.76</i>	<i>112.68</i>	<i>110.38</i>	<i>110.95</i>	<i>107.16</i>	<i>114.33</i>	<i>116.62</i>	<i>113.09</i>
	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	<i>231.54</i>	<i>232.26</i>	<i>234.34</i>	<i>233.96</i>	<i>234.39</i>	<i>232.52</i>	<i>232.17</i>	<i>233.18</i>
	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	<i>872.42</i>	<i>896.98</i>	<i>1069.06</i>	<i>1056.95</i>	<i>1081.84</i>	<i>1165.06</i>	<i>1160.35</i>	<i>1163.77</i>
	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	<i>52.51</i>	<i>53.32</i>	<i>48.18</i>	<i>47.80</i>	<i>47.46</i>	<i>58.12</i>	<i>57.98</i>	<i>58.55</i>
	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	<i>53.50</i>	<i>57.15</i>	<i>32.29</i>	<i>31.87</i>	<i>31.08</i>	<i>56.23</i>	<i>56.21</i>	<i>54.00</i>
	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



498

499 Figure 5. Spatial distributions of seasonal SSR between CERES observations and simulations using
500 WRF-CMAQ, WRF-Chem, and WRF-CHIMERE with and without aerosol feedbacks in ECR.

501

502 4 Multimodel air-quality evaluations

503 Similar to meteorology, to further determine the quantitative effects of enabling
504 aerosol feedbacks on the simulation accuracy of the air-quality variables in ECR,
505 ground-based and satellite-borne observations were adopted for comparisons in the
506 following evaluation analysis. The usage status of computing resources in each
507 simulation process was also assessed (Section 4.3).

508

509 4.1 Ground-based observations

510 Table 4 and Fig. 7 present the statistical metrics of the annual and seasonal air
511 pollutant concentrations (PM_{2.5}, O₃, NO₂, SO₂, and CO) simulated by the three coupled
512 models. The evaluations between the surface measurements and simulations of PM_{2.5} and
513 O₃ are presented below, and the performance assessments of the other gaseous pollutants
514 are presented in Section 2 of SI.

515 The R-values of the annual PM_{2.5} concentrations simulated by WRF-CMAQ (0.68)
516 were the highest, followed by those obtained by WRF-Chem (0.65–0.68) and WRF-
517 CHIMERE (0.52–0.53). The three models exhibited higher correlations in winter than in

518 the other seasons (Fig. 7). Table 4 and Figs. 6–7 reveal that WRF–CMAQ underestimated
519 the annual and seasonal (except for autumn) PM_{2.5} concentrations, with NMBs of –9.78%
520 to –6.39% and –17.68% to +5.17%, respectively. WRF–Chem overestimated and
521 underestimated PM_{2.5} on the annual and seasonal scales, with related NMBs varying from
522 –39.11% to +24.72. Concurrently, WRF–CHIMERE excessively overestimated the
523 annual and seasonal PM_{2.5} concentrations (NMB: +19.51% to +75.47%). These biases
524 could be related to the different aerosol and gas-phase mechanisms, dust and sea salt
525 emission schemes, chemical ICs and BCs, and the aerosol-size-distribution treatments
526 applied to the three two-way coupled models. Based on the NMB differences between the
527 simulations with ARI and those without aerosol feedbacks, the ARI-induced annual and
528 seasonal NMB variations in WRF–CMAQ_ARI and WRF–Chem_ARI ranged from
529 +3.01% to +4.21% and +3.07% to +5.02%, respectively, indicating that enabling ARI
530 feedbacks slightly reduced the annual and seasonal (except for autumn) underestimations
531 of PM_{2.5} concentrations. Notably, WRF–CHIMERE_ARI further overestimated the
532 annual and seasonal PM_{2.5} concentrations, with an NMB increase of up to 10.04%. The
533 increases in the PM_{2.5} concentrations due to the ARI effects were attributable to the
534 synergetic decreases in SSR, T2, WS10, and PBLH, as well as increases in RH2. With
535 ACI feedbacks further enabled, WRF–Chem_BOTH largely underestimated the annual
536 and seasonal PM_{2.5}, with NMBs varying from –24.15% to –14.44%, compared with
537 WRF–Chem_ARI. WRF–CHIMERE_BOTH tended to decrease (–2.1% to –0.51%) the
538 annual and autumn–winter NMBs and increase (+0.35% to +3.04%) the spring–summer
539 ones. A further comparison of the ARI- and ACI-induced NMB variations demonstrated
540 that the ARI-induced variations in the PM_{2.5} concentrations were smaller than the
541 ACI-induced ones in WRF–Chem, and that the reversed pattern proceeded in WRF–
542 CHIMERE. This might be explained by the incorporation of dust aerosols in WRF–
543 CHIMERE serving as IN, which was not included in WRF–Chem in this study.

544 For O₃, WRF–CHIMERE (R = 0.62) exhibited the highest correlation, followed by
545 WRF–CMAQ (R = 0.55) and WRF–Chem (R = 0.45) (Table 4 and Fig. S16). WRF–
546 CMAQ slightly underestimated the annual O₃ concentration, with NMBs and NGEs of
547 –12.57% to –11.52%; conversely, WRF–Chem and WRF–CHIMERE significantly
548 overestimated it, with NMBs of 47.82%–48.10% and 29.46%–29.75%, respectively. The
549 seasonal results of the statistical metrics displayed consistent patterns with the annual
550 simulations, and the O₃ pollution levels in summer were better simulated than in the other
551 seasons (Fig. 6). The models with enabling ARI feedbacks slightly decreased the annual
552 and seasonal O₃ NMBs and NGEs, ranging from –3.02% to +0.85% (the only positive
553 value of +0.85% was produced by WRF–CMAQ in summer) and –1.42% to –0.75%,
554 respectively. Concurrently, regarding the ACI effects, WRF–Chem and WRF–CHIMERE
555 exhibited increased annual O₃ NMBs and NGEs of 0.12%–0.65% and 0.40%–0.55%,
556 respectively. The ACI-induced seasonal NMB variations for WRF–Chem differed from
557 those for WRF–CHIMERE; WRF–Chem increased and decreased in spring–summer and
558 autumn–winter, respectively, whereas WRF–CHIMERE increased in all seasons except

559 winter (Fig. 7). Such diverse NMB and NGE variations can be explained by two aspect
 560 differences. Under the model-top BCs, the WRF-CMAQ and WRF-Chem models
 561 employed the parameterization scheme of O₃-potential vorticity, and WRF-CHIMERE
 562 employed the climatological data from LMDz-INCA. Regarding the gas-phase chemistry
 563 mechanisms, the three coupled models incorporated various photolytic reactions, with a
 564 more comprehensive discussion in Section 4.2.

565 Knote et al. (2015) comprehensively assessed the effects of seven gas-phase
 566 chemical mechanisms (RADM2, RADMKA, RACM-ESRL, CB05Clx, CB05-TUCL,
 567 CBMZ, and MOZART-4) on O₃ simulations using the three two-way coupled models
 568 (WRF-Chem, WRF-CMAQ, and COSMO-ART). They concluded that the O₃
 569 concentrations simulated by WRF-Chem using the CBMZ mechanism were closest to the
 570 mean values of multiple models for North America and Europe in spring and summer.
 571 However, dissimilar to North America and Europe, the two-way coupled WRF-Chem
 572 with CBMZ exhibited the lowest performance in spring for ECR. Additionally, the ARI
 573 and/or ACI effects contributed to atmospheric dynamics and stability (as mentioned in the
 574 PBLH evaluation part of Section 1.1 in SI), as well as photochemistry and heterogeneous
 575 reactions; thus, they eventually influenced O₃ formation (Xing et al., 2017; Qu et al.,
 576 2021; Zhu et al., 2021).

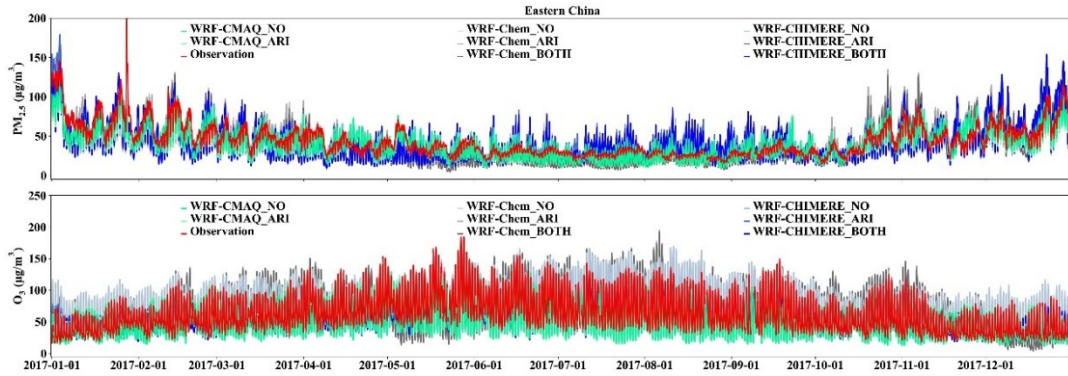
577

578 Table 4. Statistical metrics (R, MB, NMB, NGE, and RMSE) of the annual simulations
 579 and observations of surface PM_{2.5}, O₃, NO₂, SO₂, and CO in ECR. The best results are in
 580 bold, while the 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	<i>40.59</i>	<i>42.12</i>	<i>44.45</i>	<i>46.65</i>	<i>38.33</i>	<i>62.17</i>	<i>65.36</i>	<i>65.13</i>
	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
	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	<i>55.06</i>	<i>54.41</i>	<i>88.53</i>	<i>87.81</i>	<i>87.89</i>	<i>76.92</i>	<i>76.48</i>	<i>76.89</i>
	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
	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	<i>33.94</i>	<i>34.46</i>	<i>21.17</i>	<i>21.98</i>	<i>21.40</i>	<i>21.85</i>	<i>22.20</i>	<i>22.24</i>
	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
	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

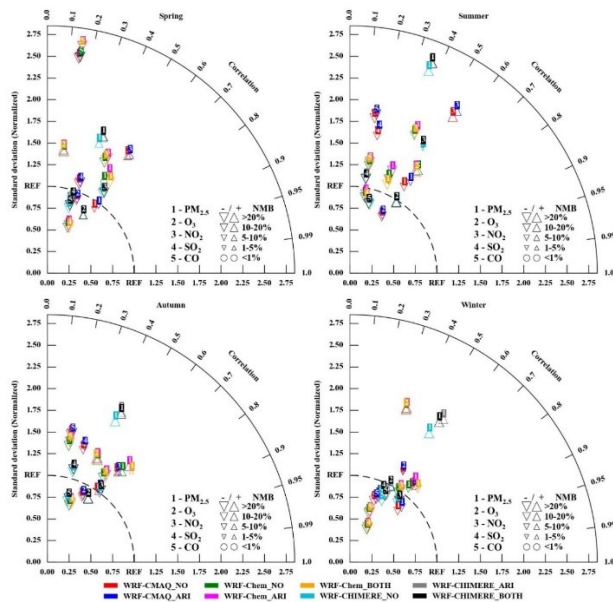
581



582

583 Figure 6. Time series of the observed and simulated hourly PM_{2.5} and O₃ concentrations by WRF-
584 CMAQ, WRF-Chem, and WRF-CHIMERE with/without aerosol feedbacks over ECR in 2017.

585



586

587 Figure 7. Taylor diagrams (R, normalized standard deviation, and NMB) of seasonal PM_{2.5}, O₃, NO₂,

588 SO₂, and CO using the three two-way coupled models (WRF-CMAQ, WRF-Chem, and WRF-
589 CHIMERE) with/without the ARI and/or ACI effects in ECR compared with the surface observations

590 Similar to the meteorological variables presented above, we conducted quality
591 assurance for the statistical metrics via further comparisons with the PM_{2.5} and O₃ results
592 in previous model evaluations (Fig. S20). In this study, the performances of WRF-
593 CMAQ and WRF-Chem in simulating PM_{2.5} were better than the average levels reported
594 by the previous studies on ECR. Regarding the simulation of the O₃ level, WRF-Chem
595 performed worse compared with the average level reported by the previous studies.
596 Although the R-values of O₃ simulated by WRF-CMAQ in this study were lower than
597 the average level reported in the previous studies, our RMSEs were smaller.

598 4.2 Satellite-borne observations

599 In this section, we further investigated the discrepancies among the different models
600 regarding the calculated AOD and column concentrations of the gases (O₃, NO₂, SO₂, CO,
601 and NH₃) and compared them with various satellite observations. Regarding NH₃, as the
602 output of simulated NH₃ concentrations was not set in WRF-CHIMERE, the discussion
603 here only includes the results from the WRF-CMAQ and WRF-Chem models.

604 Table 5 reveals that the annual AOD at 550 nm, TCO, NO₂, and CO simulated by the
605 three models agreed the most with the satellite observations, with R-values of 0.80–0.98;
606 these were followed by NH₃ (0.75–0.76), and SO₂ (0.50–0.53). WRF-CMAQ exhibited
607 negative biases for the annual AOD (–0.01), TCO (–5.92 Dobson Units (DU)), SO₂
608 (–0.03 to –0.02 DU), CO (-1.25×10^{17} molecules cm⁻²), and NH₃ (-2.95×10^{15}
609 molecules cm⁻²). Conversely, it exhibited a positive bias for NO₂ (1.09–1.21
610 petamolecules cm⁻²). Regarding AOD, WRF-Chem and WRF-CHIMERE produced
611 positive (+0.09) and negative (–0.06) MBs. WRF-Chem and WRF-CHIMERE
612 overestimated NO₂ (0.28–0.63 petamolecules cm⁻²) and CO ($0.93-1.21 \times 10^{17}$ molecules
613 cm⁻²) and underestimated O₃ (–10.99 to –3.63 DU) and SO₂ (–0.03 to –0.02 DU).
614 Similar to WRF-CMAQ, WRF-Chem underestimated NH₃ by approximately $-3.14 \times$
615 10^{15} molecules cm⁻².

616 Regarding the seasonal variations, we observed relatively high correlation
617 relationships (0.71–0.88) regarding AOD in autumn, with lower values (0.53–0.84) in the
618 other seasons (Fig. 8). WRF-CMAQ and WRF-Chem tended to underestimate (MBs of
619 –0.1 to –0.4) and overestimate (MBs of 0.01–0.05) AOD in summer and the other
620 seasons, respectively. WRF-CHIMERE exhibited positive (0.03–0.04) and negative
621 (–0.10 to –0.01) biases in winter and the other seasons, respectively. Regarding TCO (Fig.
622 S24), the performances of the WRF-CMAQ and WRF-Chem models in spring and
623 winter were slightly better than the performances in summer and autumn; however, the
624 R-values of all the seasons were above 0.89. WRF-CMAQ (–9.53 to –0.72 DU) and
625 WRF-Chem (–24.62 to +10.57 DU) exhibited negative biases in all the seasons (except
626 WRF-Chem in autumn). WRF-CHIMERE better captured TCO in spring and summer
627 (overestimations of +9.19 to +29.20 DU) than in autumn and winter (underestimations of
628 –33.75 to –19.40 DU). The R-values of the NO₂ columns for the three models were

629 slightly higher in autumn and winter (0.82–0.91) than in spring and summer (0.76–0.84).
630 Generally, WRF–CMAQ (–0.68 to –0.16 DU), WRF–Chem (–1.40 to –0.44 DU), and
631 WRF–CHIMERE (–1.31 to –0.19 DU) generally underestimated the seasonal NO₂
632 columns (Fig. S22). All the models overestimated the SO₂ column concentrations in
633 winter (by 0.01–0.03 DU) but underestimated them in the other seasons (–0.05 to –0.001
634 DU) (Fig. S23). Regarding NH₃, the only primary alkaline gas in the atmosphere, the
635 WRF–CMAQ and WRF–Chem models performed better in summer (R: 0.81–0.87; MB:
636 –3.42 to 2.07×10^{15} molecules cm^{–2}) (Fig. S25). The NH₃ emissions from fertilizers and
637 livestock have been substantially underestimated in China (Zhang et al., 2017), and the
638 peak values were obtained in spring and summer (Huang et al., 2012). Additionally, the
639 bidirectional exchanges of fertilizer-induced NH₃ were not considered in our simulations.
640 Compared with the above column variables, WRF–CMAQ, WRF–Chem, and WRF–
641 CHIMERE exhibited relatively poor performances (R: 0.68–0.79) in simulating the CO
642 columns during spring, summer, and autumn, respectively, than in simulating them in the
643 other seasons (Fig. S24). WRF–CMAQ and WRF–CHIMERE underestimated and
644 overestimated the CO columns in the other seasons, respectively, except for summer and
645 spring, with MBs of –3.29 to 0.31×10^{17} and –0.62 to 2.09×10^{17} molecules cm^{–2},
646 respectively. WRF–Chem obtained positive MBs in summer and autumn (4.03 – $5.12 \times$
647 10^{17} molecules cm^{–2}) and negative ones in spring and winter (–3.15 to -2.10×10^{17}
648 molecules cm^{–2}).

649 Moreover, after comparing the performances of the models for each pollutant
650 between Sections 4.1 and 4.2, the only disparity found between evaluations with
651 ground-based observations and those with satellite-borne observations was for CO. The
652 formation of CO via the oxidation of methane, an important source of CO emissions
653 (Stein et al., 2014), was not considered in the three coupled models, and the methane
654 emissions were not included in the MEIC inventory. Furthermore, the contribution of CO
655 to atmospheric oxidation capacity (OH radicals) was nonnegligible (e.g., the values were
656 approximately 20.54%–38.97% in Beijing (Liu et al., 2021) and 26%–31% in Shanghai
657 (Zhu et al., 2020)). In addition, these discrepancies in the model performances in
658 simulating AOD and column concentrations of gases can be explained by the differences
659 in the representations of the aerosol species groups, Fast-JX photolysis scheme, and
660 gas-phase mechanisms in the three coupled models. More detailed interpretations were
661 grouped into four aspects: (1) AODs are calculated via the Mie theory using the refractive
662 indices of different numbers (5, 6, and 10) of aerosol species groups in different coupled
663 models (WRF–CMAQ, WRF–Chem, and WRF–CHIMERE) (Tables S5–S6); (2) seven
664 (294.6, 303.2, 310.0, 316.4, 333.1, 382.0, and 607.7 nm), four (300, 400, 600, and 999
665 nm), and five (200, 300, 400, 600, and 999 nm) effective wavelengths were used to
666 calculate the actinic fluxes and photolysis rates in the Fast-JX photolysis modules of
667 WRF–CMAQ, WRF–Chem, and WRF–CHIMERE, respectively; (3) different methods
668 exist in the Fast-JX schemes of the three coupled models for calculating the aerosol and
669 cloud optical properties (Tables S1 and S5–S6); (4) 77, 52, and 40 gas-phase species

670 comprised 218, 132, and 120 gas-phase reactions under the CB6, CBMZ, and
 671 MELCHIOR2 mechanisms, respectively.

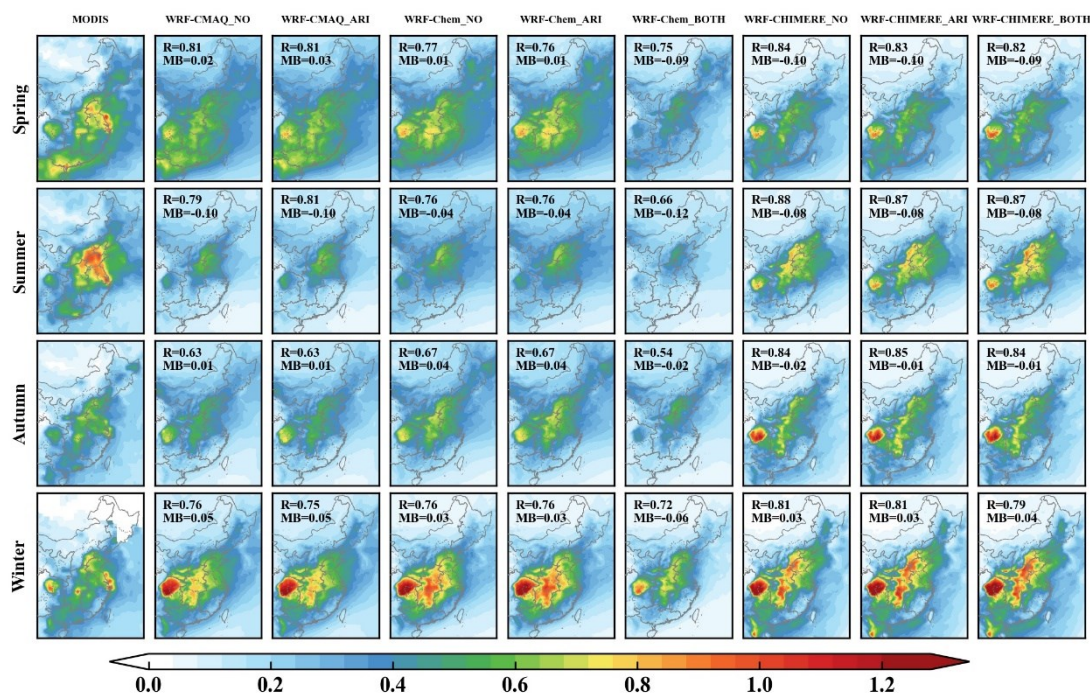
672 When the three models enabled only the ARI effects, relatively limited
 673 improvements were observed in the annual AOD and NO₂ columns simulated by these
 674 models. The AOD simulations improved in spring and summer but worsened in autumn
 675 and winter (Table 4 and Fig. 9). Larger ARI-induced variations in seasonal MBs of the
 676 NO₂ columns were observed in WRF-CMAQ (-0.18 to 0.13 petamolecules cm⁻²)
 677 compared with WRF-Chem and WRF-CHIMERE (0-0.01 petamolecules cm⁻²). When
 678 the ARI and ACI effects were enabled in WRF-Chem, the model performance for
 679 seasonal AOD simulations worsened considerably. The annual and seasonal NO₂
 680 simulations by WRF-Chem became slightly worse, whereas those by WRF-CHIMERE
 681 became slightly better. Dissimilar to AOD and the NO₂ column concentrations, the
 682 improvements in the annual and seasonal column simulations of total ozone, PBL SO₂,
 683 and NH₃ by all the two-way coupled models were limited when one or both of ARI and
 684 ACI were enabled.

685 Table 5. Statistical metrics (R, MB, NMB, NGE, and RMSE) of the simulated and
 686 satellite-retrieved AOD, TCO, tropospheric column NO₂, PBL column SO₂, total column
 687 CO, and total column density of NH₃ in ECR. The best results are captured in bold fonts,
 688 and the 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 (<i>0.27</i>)	Mean_sim	<i>0.26</i>	<i>0.27</i>	<i>0.35</i>	<i>0.36</i>	<i>0.25</i>	<i>0.21</i>	<i>0.22</i>	<i>0.22</i>
	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	Mean_sim	<i>306.15</i>	<i>306.15</i>	<i>300.77</i>	<i>300.73</i>	<i>300.46</i>	<i>307.69</i>	<i>307.47</i>	<i>307.75</i>
VCDs (<i>312.07</i> DU)	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 (<i>2.71 × 10¹⁵</i> molecules cm ⁻²)	Mean_sim	<i>3.80</i>	<i>3.91</i>	<i>3.07</i>	<i>3.08</i>	<i>3.06</i>	<i>2.62</i>	<i>2.63</i>	<i>2.63</i>
	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 (<i>0.09</i> DU)	Mean_sim	<i>0.07</i>	<i>0.07</i>	<i>0.09</i>	<i>0.09</i>	<i>0.06</i>	<i>0.06</i>	<i>0.06</i>	<i>0.06</i>
	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	<i>20.34</i>	<i>20.35</i>	<i>22.20</i>	<i>22.20</i>	<i>22.21</i>	<i>22.34</i>	<i>22.36</i>	<i>22.35</i>
	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	<i>13.06</i>	<i>13.15</i>	<i>12.31</i>	<i>12.27</i>	<i>8.63</i>	N/A	N/A	N/A
	R	0.76	0.76	0.73	0.73	0.76	N/A	N/A	N/A
	MB	-3.00	-2.90	-3.27	-3.32	-3.34	N/A	N/A	N/A
	NMB (%)	-18.66	-18.08	-21.01	-21.28	-21.41	N/A	N/A	N/A
	NGE (%)	47.69	48.09	50.84	50.80	50.99	N/A	N/A	N/A
	RMSE	9.26	9.47	9.48	9.46	9.61	N/A	N/A	N/A

689 N/A indicates that the outputs of the NH₃ column concentrations were not extracted from WRF-CHIMERE simulations
 690 with/without aerosol feedbacks.



691

692 Figure 8. Spatial distributions of seasonal AOD between MODIS observations and simulations using
 693 the WRF-CMAQ, WRF-Chem, and WRF-CHIMERE models with and without aerosol feedbacks in
 694 ECR.

695

696 4.3 Computational performance

697 Table 5 presents a summary of the comparative results of the time consumption by
 698 the central processing unit (CPU) per simulation day using WRF-CMAQ, WRF-Chem,
 699 and WRF-CHIMERE with and without aerosol feedbacks in 2017. The results indicated
 700 that WRF-CMAQ consumed the shortest CPU time simulating one-day meteorology and
 701 air quality with or without enabling aerosol feedbacks. This CPU time consumption was
 702 followed by WRF-CHIMERE and WRF-Chem. Compared with the simulations without
 703 aerosol feedbacks, the processing time of WRF-CMAQ with ARI increased by 0.22–0.34
 704 h per day. The increases in the running time of WRF-Chem and WRF-CHIMERE were
 705 insignificant (0.02–0.03 h per day). The CPU times for WRF-Chem and WRF-
 706 CHIMERE with the ARI and ACI effects enabled increased slightly, and the increase in
 707 the CPU time for the former (0.25 h per day) was higher than that for the latter (0.11 h
 708 per day). Compared with WRF-CMAQ and WRF-Chem, the CPU time consumed by
 709 WRF-CHIMERE exhibited clear seasonal differences, with the CPU times in winter and
 710 spring being significantly longer than those in summer and autumn. These differences can
 711 be partially explained by the choice of the main configurations, including the model
 712 resolution, model version, and parametrization schemes (cloud microphysics, PBL,
 713 cumulus, surface layer, land surface, gas-phase chemistry, and aerosol mechanisms).

714 Table 5. Summary of the running time for the different coupled models.

Month	WRF-CMAQ (h)		WRF-Chem (h)			WRF-CHIMERE (h)		
	NO	ARI	NO	ARI	BOTH	NO	ARI	BOTH
Jan.	0.37	0.59	0.69	0.71	0.96	0.67	0.70	0.77
Feb.	0.35	0.60	0.68	0.70	0.93	0.64	0.67	0.73
Mar.	0.39	0.65	0.70	0.72	1.00	0.59	0.62	0.72
Apr.	0.37	0.67	0.67	0.69	0.92	0.54	0.57	0.65
May	0.39	0.71	0.61	0.66	0.86	0.52	0.55	0.62
June	0.40	0.74	0.66	0.67	0.95	0.48	0.51	0.63
July	0.36	0.69	0.65	0.67	0.86	0.49	0.50	0.58
Aug.	0.38	0.68	0.66	0.68	0.90	0.49	0.52	0.61
Sept.	0.37	0.63	0.64	0.65	0.89	0.48	0.52	0.63
Oct.	0.38	0.62	0.66	0.68	0.94	0.53	0.56	0.69
Nov.	0.36	0.58	0.68	0.70	0.91	0.64	0.67	0.72
Dec.	0.35	0.57	0.63	0.66	0.87	0.67	0.70	0.74

715

716 5 Conclusions

717 Two-way coupled meteorology and air-quality models have been deployed in ECR
 718 in recent years. However, no study comprehensively assessed multiple coupled models in
 719 this region. To the best of our knowledge, this is the first study to perform comprehensive

720 intercomparisons of the open-sourced two-way coupled meteorology and air-quality
721 models (WRF–CMAQ, WRF–Chem, and WRF–CHIMERE). Here, we systemically
722 evaluated the hindcast simulations for 2017 and explored the impacts of ARI and/or ACI
723 on the model performance and computational efficiency in ECR.

724 After detailed comparisons with ground-based and satellite-borne observations, the
725 evaluation results revealed that the three coupled models performed well for meteorology
726 and air quality, particularly for surface temperature (with an R-value of up to 0.97) and
727 PM_{2.5} concentrations (with an R-value of up to 0.68). The effects of aerosol feedbacks on
728 the model performance varied with the two-way coupled models, variables, and time
729 scales. The computational time increased by 20%–70% when these two-way coupled
730 models enabled aerosol feedbacks compared with when the simulations proceeded
731 without aerosol–radiation–cloud interactions. Notably, the three coupled models could
732 effectively reproduce the spatiotemporal distributions of the satellite-retrieved CO
733 column concentrations but not for ground-observed CO concentrations.

734 The intercomparisons revealed some uncertainty sources in the evaluation of the
735 aerosol feedback effects. As numerous schemes can be combined with the configurations
736 of different coupled models, we only evaluated the simulations with specific settings.
737 Future comparisons considering more combinations of multiple schemes within the same
738 or different coupled models are desired. Among the three coupled models, the numerical
739 representations for specific variables in the same scheme are diverse, e.g., the treatments
740 of cloud cover and cloud optical properties in the Fast-JX photolysis scheme. More
741 accurate representations of photolysis processes must be considered to reduce evaluation
742 uncertainties. Additionally, the FDDA nudging technique can attenuate the ARI effects
743 during severe air pollution episodes, and optimal nudging coefficients among different
744 regions must be determined. Finally, the actual mechanisms underlying the ACI effects
745 are still unclear, and the new advances in the measurements and parameterizations of
746 CCN/IN activations and PREC must be duly incorporated in coupled models.

747

748 Code availability

749 The source codes of the two-way coupled WRF v4.1.1-CMAQ v5.3.1, WRF-Chem
750 v4.1.1, and WRF v3.7.1-CHIMERE v2020r1 models are obtained from
751 <https://github.com/USEPA/CMAQ>, <https://github.com/wrf-model/WRF>, and
752 <https://www.lmd.polytechnique.fr/chimere>, respectively (last access: November 2020).
753 The related source codes, configuration information, namelist files and automated run
754 scripts of these three two-way coupled models are archived at Zenodo with the associated
755 DOI: <https://doi.org/10.5281/zenodo.7901682> (Gao et al., 2023a; link:
756 <https://zenodo.org/record/7901682>).

757

758 Data availability

759 The meteorological ICs and BCs used for three coupled models can be obtained at

760 <https://doi.org/10.5281/zenodo.7925012> (Gao et al., 2023b; link:
761 <https://zenodo.org/record/7925012>). The Chemical ICs and BCs used for WRF-CMAQ,
762 WRF-Chem and WRF-CHIMERE are available at
763 <https://doi.org/10.5281/zenodo.7932390> (Gao et al., 2023c; link:
764 <https://zenodo.org/record/7932390>), <https://doi.org/10.5281/zenodo.7932936> (Gao et al.,
765 2023d; link: <https://zenodo.org/record/7932936>), and
766 <https://doi.org/10.5281/zenodo.7933641> (Gao et al., 2023e; link:
767 <https://zenodo.org/record/7933641>), respectively. The emission data used for
768 WRF-CMAQ, WRF-Chem and WRF-CHIMERE can be downloaded from
769 <https://doi.org/10.5281/zenodo.7932430> (Gao et al., 2023f; link:
770 <https://zenodo.org/record/7932430>), <https://doi.org/10.5281/zenodo.7932734> (Gao et al.,
771 2023g; link: <https://zenodo.org/record/7932734>), and
772 <https://doi.org/10.5281/zenodo.7931614> (Gao et al., 2023h; link:
773 <https://zenodo.org/record/7931614>), respectively. The DOIs and links regarding the
774 output data of each simulation scenario are presented in Table S9. All data used to create
775 figures and tables in this study are provided in an open repository on Zenodo
776 (<https://doi.org/10.5281/zenodo.7750907>, Gao et al., 2023i; link:
777 <https://zenodo.org/record/7750907>).

778

779 Author contributions

780 CG, ZX, AX performed the majority of the source code configuration of
781 WRF-CMAQ, WRF-Chem and WRF-CHIMERE, designed the numerical simulations to
782 carry them out, related analysis, figure plotting, and paper writing. QT, HZ, SZ, GY, MZ
783 and XS were involved with the original research plan and made suggestions for the paper
784 writing.

785

786 Competing interests

787 The contact author has declared that neither they nor their co-authors have any
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789

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