



*Supplement of*

**Intercomparison of multiple two-way coupled meteorology and air quality models (WRF v4.1.1–CMAQ v5.3.1, WRF-Chem v4.1.1, and WRF v3.7.1–CHIMERE v2020r1) in eastern China**

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1      Supplement

2      S1. Evaluations of other meteorological variables

3      S1.1. Ground-based observations

4      For Q2, RMSEs between WRF-CMAQ, WRF-Chem, and WRF-CHIMERE  
5      simulations and surface observation were consistently below  $3 \text{ g kg}^{-1}$ , as illustrated in  
6      Table S3 and Fig. S2. Most models exhibited a tendency to underestimate annual and  
7      seasonal Q2, with MBs ranging from  $-0.57$  to  $-0.18 \text{ g kg}^{-1}$  and  $-1.16$  to  $+0.20 \text{ g kg}^{-1}$   
8      in WRF-Chem and WRF-CHIMERE, respectively. The more obvious underestimations  
9      appeared in summer. In the MICS-Asia III project, Gao et al. (2018) reported that all  
10     the seven included two-way coupled models produced slightly positive values for Q2  
11     during January 2010 over the North China Plain. In contrast to simulations without  
12     enabling aerosol feedbacks, the negative biases in annual and seasonal Q2 simulated by  
13     WRF-CMAQ\_ARI and WRF-CHIMERE\_ARI were amplified, and the WRF-  
14     CMAQ\_ARI simulations exhibited bigger negative biases (see Fig. 3 and Table S3).  
15     The changes in annual, summer, and autumn MBs for WRF-Chem\_ARI were consistent  
16     with the trend of WRF-CMAQ\_ARI, except for spring and winter.

17     The annual and seasonal correlation coefficients of precipitation were 0.56–0.69,  
18     0.46–0.63, and 0.25–0.55 for WRF-CMAQ, WRF-Chem, and WRF-CHIMERE,  
19     respectively (Table S3 and Fig. S5). All simulated results presented the highest  
20     correlations in winter and the lowest in summer and the possible reasons are due to the  
21     much more convective activities in summertime, which are not accurately captured in  
22     all coupled models. WRF-CMAQ and WRF-CHIMERE exhibited underestimation and  
23     overestimation in annual and seasonal precipitation, respectively. At the annual and  
24     seasonal scales, WRF-Chem and WRF-CHIMERE overestimated the daily  
25     precipitation magnitude by more than  $1 \text{ mm day}^{-1}$ , and WRF-CMAQ underestimated it  
26     by approximately  $0.5 \text{ mm day}^{-1}$ . A similar conclusion was obtained for North America  
27     during 2010, with the magnitude of precipitation MBs being higher in WRF-Chem  
28     compared to WRF-CMAQ (refer to Fig. 11 in Makar et al., 2015). The largest  
29     precipitation MBs simulated by the three models occurred in summer and varied from  
30      $-0.70$  to  $+1.39 \text{ mm day}^{-1}$ . The RMSE was highest in WRF-CHIMERE, followed by  
31     WRF-Chem, and WRF-CMAQ, and all models had the largest ( $> 10 \text{ mm day}^{-1}$ ) and  
32     smallest (approximately  $2.5 \text{ mm day}^{-1}$ ) values in summer and winter, respectively.  
33     Considering the ARI effects, WRF-CMAQ\_ARI simulations amplified the  
34     underestimations of annual and seasonal precipitation in eastern China. In contrast,  
35     WRF-Chem\_ARI (except for autumn) and WRF-CHIMERE\_ARI simulations  
36     mitigated the overestimations of precipitation. The effects of ARI on summer MBs were  
37     larger in all three coupled models compared to other seasons. When ACI effects were  
38     further included, WRF-Chem\_BOTH demonstrated only marginal improvement in  
39     precipitation overestimation compared to WRF-Chem\_NO, while WRF-  
40     CHIMERE\_BOTH gave out certain enhancement of precipitation overestimation. This  
41     can be interpreted as follows: WRF-CHIMERE has the ability to simulate the activation  
42     of aerosol particles into cloud ice via heterogeneous ice nucleation and homogeneous  
43     freezing, whereas WRF-Chem lacks this capability.

44     Overall, the PBLH was not well simulated by any of the three coupled models,

which may be a result of the adoption of low resolution sounding data in evaluations (Brunner et al., 2015) and the different settings of Richardson number thresholds in the calculation of observed PBLH (Guo et al., 2016). At 08:00 and 20:00 local time (LT), the simulated PBLHs in WRF-CMAQ have lower correlations only ranging from 0.21 to 0.40 and largest negative MBs varying from  $-400$  to  $-133$  m. These poor performances were mainly caused by: 1) different configurations of the PBL scheme were employed in this study, namely, WRF-CMAQ adopted the ACM2 scheme with hybrid local–nonlocal closure, while WRF-Chem and WRF-CHIMERE adopted the YSU scheme with non-local closure (Table 1); 2) Richardson number threshold was set to different values for unstable atmospheric conditions, i.e., the YSU and ACM2 schemes using the thresholds of 0 and 0.25, respectively (Xie et al., 2012); 3) different to the YSU scheme, the ACM2 scheme considers the entrainment layer in the PBLH calculations (Xie et al., 2012).

Meanwhile, all correlations of PBLH simulated by the three coupled models at 20:00 LT ( $R = 0.3\text{--}0.4$ ) were better than those at 08:00 LT ( $R = 0.1\text{--}0.2$ ), which indicated that the PBL schemes in these model were able to calculate PLBH after PBL collapsing a little better than before PBL developing and more observation with better spatiotemporal resolutions are needed to further evaluate the models' performance. In addition, the RMSEs of PBLH in autumn (369.89–388.79 m) and winter (347.48–392.38 m) were smaller than those in spring (405.61–622.37 m) and summer (348.80–570.16 m) for all three models.

As shown in Fig. 3 and Table S3, the changes of MB and RMSE of simulated PBLH induced by the effects of aerosol feedbacks were greater than those of R. Meanwhile, the MBs were further analyzed. For WRF-CMAQ, ARI effects induced an increase ( $-1.93$  m) and decrease ( $+6.66$  m) in the annual underestimations of PBLH at 8:00 and 20:00 LT, respectively (Table S3). The negative MBs for WRF-Chem\_ARI and WRF-Chem\_BOTH showed an enhancement (08:00 LT:  $-25.25$  m, 20:00 LT:  $-25.60$  m) and reduction (08:00 LT:  $+19.65$  m, 20:00 LT:  $+14.09$  m) compared to those for WRF-Chem\_NO and WRF-Chem\_ARI, respectively. Both the ARI ( $-6.17$  and  $-3.34$  m) and ACI ( $-0.65$  and  $-1.11$  m) effects further underestimated annual PBLH at 08:00 and 20:00 LT for WRF-CHIMERE. Note that the variations in MBs induced by aerosol feedbacks for the three coupled models at the annual scale were similar to those at the seasonal scale.

### S1.2. Satellite-borne observations

As indicated in Table 3, the three coupled models demonstrated good performance in simulating the shortwave radiation at *the top of the atmosphere* (SRTOA) and longwave radiation at *the top of the atmosphere* (LRTOA). The annual MBs for SRTOA and LRTOA are ranging from  $-4.40$  to  $+5.42$   $\text{W m}^{-2}$  and  $-2.14$  to  $0.66$   $\text{W m}^{-2}$ , respectively. Seasonal SRTOA was also well simulated by all three models, especially in winter (Figure S10). For seasonal LRTOA, the WRF-CMAQ and WRF-Chem model performances were better than that of WRF-CHIMERE for all seasons except autumn (Figure S11). No matter whether ARI and/or ACI effects were enabled or not, simulations by WRF-CMAQ exhibited negative MBs in all seasons and WRF-

89 CHIMERE displayed negative MBs in all seasons except for spring. For WRF-Chem,  
90 it produced underestimations and overestimations of SROTA in spring–summer and  
91 autumn–winter, respectively.

92

### 93 S2. Evaluations of other air quality variables

94 According to the annual statistical results (Table 4 and Fig. S17), the NO<sub>2</sub>  
95 simulated by all three models had comparable correlations (0.50–0.60) with ground-  
96 based observations. WRF-CMAQ slightly overestimated NO<sub>2</sub> (MBs of +2.74 to +3.26  
97  $\mu\text{g m}^{-3}$ , and NMBs of +8.77% to +10.44%). In contrast, WRF-Chem (MBs of −10.03  
98 to −9.22  $\mu\text{g m}^{-3}$ , and NMBs of −32.14% to −29.55%) and WRF-CHIMERE (MBs of  
99 −9.35 to −8.96  $\mu\text{g m}^{-3}$ , and NMBs of −29.96% to −28.73%) tended to significantly  
100 underestimate NO<sub>2</sub> in eastern China. For seasonal variations (Fig. 7), WRF-CMAQ  
101 showed the best performance in winter, and generally overestimated NO<sub>2</sub> in all seasons  
102 with the NMBs ranging from −2.21% to 34.34%. Both WRF-Chem and WRF-  
103 CHIMERE had maximum R and NMB values (0.42 to 0.50 and −13.09% to −3.23%,  
104 respectively) in winter, and minimum values (0.57 to 0.62 and −41.57% to −38.05%,  
105 respectively) in summer. The annual and seasonal positive biases of WRF-CMAQ are  
106 partially caused by lack of incorporation of heterogeneous reactions of NO<sub>2</sub> that  
107 occurred on ground and aerosol surfaces (Spataro et al., 2013; Li et al., 2018; Liu et al.,  
108 2019). Recently, Zhang et al. (2021) addressed these gaps in CMAQ v5.3 but related  
109 modules had not been integrated into the latest officially released version (version 5.4).  
110 For WRF-Chem and WRF-CHIMERE, underestimations of NO<sub>2</sub> were consistent with  
111 overestimations of O<sub>3</sub>, as the NO<sub>x</sub> depletions were dominated by O<sub>3</sub> titrations. In  
112 addition, subtle differences existed in the default settings of reaction rate constants for  
113 specific chemical reactions referring to NO<sub>x</sub> in WRF-CMAQ, WRF-Chem, and WRF-  
114 CHIMERE. More detailed information can be found in the source code files of  
115 mech\_cb6r3\_ae6\_aq.def, module\_cbmz.F, and rates.F, respectively. With ARI  
116 feedbacks enabled, the annual and seasonal R values of NO<sub>2</sub> simulated by WRF-CMAQ  
117 improved, but the NMBs worsened. In contrast, both WRF-Chem and WRF-CHIMERE  
118 presented improvements. Our results showed that ARI effects tended to amplify NO<sub>2</sub>  
119 overestimations in WRF-CMAQ, and alleviate underestimations in WRF-Chem and  
120 WRF-CHIMERE. This can be explained by the ARI-induced NO<sub>2</sub> reductions being  
121 associated with slower photochemical reactions, strengthened atmospheric stability and  
122 O<sub>3</sub> titration, and vice versa. The inclusion of ACI effects in WRF-Chem and WRF-  
123 CHIMERE resulted in relatively limited improvements in model performances.

124 All models had the poorest performance in the annual and seasonal SO<sub>2</sub> and CO  
125 simulations over eastern China (Table 4 and Fig. 6). For SO<sub>2</sub>, annual correlations were  
126 comparable for all models ranging from 0.39 to 0.41. All three models underestimated  
127 SO<sub>2</sub>. WRF-CMAQ showed the smallest MB of −4.31  $\mu\text{g m}^{-3}$ , while WRF-Chem had  
128 the largest of −10.30  $\mu\text{g m}^{-3}$ . Gao et al. (2018) also demonstrated that all two-way  
129 coupled models, except the WRF-Chem version from the University of Iowa *modelling*  
130 group, tended to underestimate SO<sub>2</sub> (−54.77 to 4.50  $\mu\text{g m}^{-3}$ ) over the North China Plain  
131 during January 2013. The R values for all models were highest in autumn and winter  
132 (0.31–0.46) and lowest in spring and summer (0.16–0.38), while NMBs showed the

133 opposite trend. As concluded by Liu et al. (2010), the larger underestimations of  
 134 seasonal SO<sub>2</sub> concentrations were caused by the weaker solar radiation and lower  
 135 amount of precipitation in winter compared to summer. These conditions slowed down  
 136 the photochemical conversion of SO<sub>2</sub> to SO<sub>4</sub><sup>2-</sup>, wet scavenging, and aqueous-phase  
 137 oxidation rates of SO<sub>2</sub>.

138 For CO (Table 4), WRF-CHIMERE (0.47–0.48) had higher correlation  
 139 coefficients than those of WRF-CMAQ (0.23–0.24) and WRF-Chem (0.21–0.22). All  
 140 three models underestimated CO concentrations, with MBs ranging from –0.52 to  
 141 –0.39 mg m<sup>-3</sup>. These underestimations were partly attributed to uncertainties in the  
 142 vertical allocation of CO emissions (He et al., 2017). WRF-CMAQ and WRF-Chem  
 143 both produced spring-minimum (0.15) and winter-maximum (0.36) seasonal cycles of  
 144 R values (Fig. 6), while WRF-CHIMERE presented high (0.47) and low (0.26)  
 145 correlations in winter and summer, respectively. Negative seasonal NMBs varied from  
 146 –56.94% to –33.18% in all coupled models. When ARI effects were considered, annual  
 147 and seasonal SO<sub>2</sub> and CO model performances in all three models showed slight  
 148 improvement (R increased approximately 0.01 and NMB enhanced from 0.98% to  
 149 1.71%). Moreover, the enhancements in the simulation accuracies of SO<sub>2</sub> and CO for  
 150 the two-way coupled WRF-Chem and WRF-CHIMERE were dominated by ARI effects  
 151 rather than ACI effects.

152

### 153 S3. Statistical metrics

154 The correlation coefficient (R), mean bias (MB), normalized mean bias (NMB),  
 155 normalized gross error (NGE) and root mean square error (RMSE) were adopted to  
 156 assess the accuracy of coupled models in simulating meteorological and air quality  
 157 parameters against the ground-based and satellite observations with the following  
 158 equations:

159

$$160 \quad R = \frac{\sum_{i=1}^N (p_i - \bar{p})(o_i - \bar{o})}{\sqrt{\sum_{i=1}^N (p_i - \bar{p})^2} \sqrt{\sum_{i=1}^N (o_i - \bar{o})^2}} \quad (S1)$$

$$161 \quad MB = \frac{1}{N} \sum_{i=1}^N (p_i - o_i) \quad (S2)$$

$$162 \quad NMB = \frac{\sum_{i=1}^N (p_i - o_i)}{\sum_{i=1}^N (o_i)} \quad (S3)$$

$$163 \quad NGE = \frac{\sum_{i=1}^N |p_i - o_i|}{\sum_{i=1}^N (o_i)} \quad (S4)$$

$$164 \quad RMSE = \left[ \frac{1}{N} \sum_{i=1}^N (p_i - o_i)^2 \right]^{1/2} \quad (S5)$$

165 where p<sub>i</sub> and o<sub>i</sub> are the simulated and observed parameters, respectively, n is the  
 166 total number of the values used for evaluation, and  $\bar{p}$  and  $\bar{o}$  are the averages of the  
 167 simulation and observation, respectively.

168

169    Table S1. Summary of representations of cloud cover and cloud optical properties in  
 170    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 <sup>a</sup> 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 Stammes (1993) and Fu (1996)
WRF-Chem	1. CF=0 if CLWC <sup>b</sup> =0 2. CF=1 if CIC>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 radii and cloud liquid water/ice content

171    <sup>a</sup>CF is cloud fraction. <sup>b</sup>CLWC is cloud liquid water content. <sup>c</sup>CIC is cloud ice content.

172

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

Model	Aerosol mechanism	Modal approach					
		Aitken		Accumulation		Coarse	
WRF-CMAQ	AERO6	BC, OC, sulfate, nitrate, ammonium, PMOTHR <sup>d</sup> , PNCOM <sup>e</sup> water, metals		BC, OC, sulfate, nitrate, ammonium, PMOTHR, PNCOM, water, metals, sea salt, dust		PMC <sup>f</sup> , sea salt, dust	
<b>Sectional approach</b>							
WRF-Chem	MOSAIC <sup>a</sup>	Bin 1  0.039–0.156 µm  <i>Black carbon (BC), OC, sulfate, nitrate, sea salt<sup>d</sup></i>	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  <i>Dust, sea salt, OIN<sup>g</sup></i>		
WRF-CHIMERE	SAM <sup>b</sup>	Bin 1  0.039–0.078 µm  BC, OC, sulfate, PPM <sup>c</sup>	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, dust, sea salt	Bin 6  1.25–2.5 µm  BC, OC, sulfate, dust, sea salt
						Bin 7  2.5–5.0 µm  <i>Dust, sea salt</i>	Bin 8  5.0–10.0 µm  <i>BC, OC, PPM, dust, sea salt</i>
						Bin 9  10.0–20.0 µm  <i>BC, OC, PPM, dust, sea salt</i>	Bin 10  20.0–40.0 µm  <i>Dust, sea salt</i>

176    <sup>a</sup>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.

177    <sup>b</sup>SAM is the sectional aerosol mechanism.

178    <sup>c</sup>PPM is the primary particulate matter.

179    <sup>d</sup>PMOTHR is the remaining particulate matter that *can not be speciated into fine mode*, and more detailed information is at

180    [https://www.airqualitymodeling.org/index.php/CMAQv5.0\\_PM\\_emitted\\_species\\_list](https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list).

181    <sup>e</sup>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](https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list).

182    <sup>f</sup>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](https://www.airqualitymodeling.org/index.php/CMAQv5.0_PM_emitted_species_list).

183    <sup>g</sup>OIN is the other inorganic matter.

184

185    Table S3. Statistical metrics (R, MB, NMB, NGE, and RMSE) between simulated and  
 186    observed annual SSR, T2, RH2, Q2, WS10, WD10, precipitation, and PBLH at 08:00  
 187    and 20:00 LT) in eastern China. The best results are in bold, while mean simulations  
 188    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
(155.22 W m <sup>-2</sup> )	Mean_sim	<i>191.12</i>	<i>171.14</i>	<i>194.52</i>	<i>180.04</i>	<i>191.71</i>	<i>197.88</i>	<i>188.63</i>	<i>189.54</i>
	R	0.88	<b>0.89</b>	0.88	<b>0.89</b>	0.88	0.85	0.85	0.85
	MB	35.89	<i>15.91</i>	39.30	24.82	36.48	42.65	33.41	34.32
	NMB (%)	23.12	<b>10.25</b>	25.32	15.99	23.50	27.48	21.52	22.11
	NGE (%)	206.62	<i>170.85</i>	202.41	<b>170.70</b>	208.05	242.53	221.67	226.29
T2 (13.68 °C)	RMSE	133.05	<b>120.60</b>	134.16	123.94	134.45	154.71	147.73	148.57
	Mean_sim	<i>12.81</i>	<i>12.61</i>	<i>12.99</i>	<i>12.84</i>	<i>12.96</i>	<i>11.84</i>	<i>11.68</i>	<i>11.69</i>
R		<b>0.97</b>	<b>0.97</b>	<b>0.97</b>	<b>0.97</b>	<b>0.97</b>	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	<b>-4.97</b>	-6.09	-5.21	-13.39	-14.60	-14.50
	NGE (%)	<b>10.58</b>	10.76	10.79	10.95	10.86	17.00	17.65	17.60
	RMSE	<b>2.88</b>	2.94	3.05	3.07	3.05	3.87	3.94	3.97
Q2 (8.87 g kg <sup>-1</sup> )	Mean_sim	8.69	8.51	8.57	8.54	8.58	8.35	8.30	8.30
	R	<b>0.90</b>	<b>0.90</b>	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 (%)	<b>-2.00</b>	-3.98	-3.36	-3.66	-3.19	-5.84	-6.37	-6.35
	NGE (%)	<b>16.80</b>	16.85	19.70	19.66	19.77	20.55	20.65	20.62
	RMSE	<b>2.93</b>	2.95	3.09	3.09	3.10	3.17	3.18	3.18
RH2 (67.48 %)	Mean_sim	71.03	70.51	70.01	70.33	70.13	70.41	70.58	70.46
	R	<b>0.73</b>	<b>0.73</b>	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	<b>3.74</b>	4.22	3.92	4.34	4.59	4.41
	NGE (%)	<b>19.90</b>	19.91	23.45	23.71	23.71	24.77	24.88	24.90
	RMSE	<b>18.92</b>	18.98	19.78	19.79	19.84	20.81	20.82	20.84
WS10 (2.81 m s <sup>-1</sup> )	Mean_sim	3.27	3.23	<b>3.30</b>	3.29	<b>3.30</b>	<b>3.85</b>	<b>3.83</b>	<b>3.83</b>
	R	<b>0.62</b>	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	<b>14.98</b>	17.45	17.11	17.53	36.98	36.27	36.34
	NGE (%)	96.20	<b>95.00</b>	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 (175.27 °)	Mean_sim	177.13	176.62	177.87	177.82	<b>178.11</b>	<b>171.97</b>	<b>171.53</b>	<b>171.68</b>
	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	<b>94.00</b>	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 (PREC)	Mean_sim	2.46	2.31	3.24	3.19	3.26	3.31	3.24	3.21
	R	<b>0.59</b>	<b>0.59</b>	0.50	0.50	0.50	0.35	0.34	0.34
(2.72 mm d <sup>-1</sup> )	MB	-0.27	-0.42	0.51	0.46	0.53	0.59	0.52	0.48
	NMB (%)	<b>-9.80</b>	-15.35	18.86	16.83	19.43	21.46	18.96	17.63
	NGE (%)	310.71	<b>283.10</b>	442.60	428.11	445.89	573.24	565.36	557.56
	RMSE	8.03	<b>7.96</b>	10.32	10.26	10.33	10.87	10.85	10.93
PBLH00 (432.13 m)	Mean_sim	253.54	251.61	288.41	263.16	282.81	<b>276.45</b>	270.28	269.63
	R	<b>0.21</b>	<b>0.21</b>	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	<b>-33.26</b>	-39.10	-34.55	-36.03	-37.45	-37.61
	NGE (%)	58.89	58.75	<b>54.37</b>	56.96	54.51	57.20	57.63	57.28
	RMSE	380.23	378.79	<b>371.27</b>	379.72	372.14	373.78	375.85	374.52
PBLH12 (547.02 m)	Mean_sim	230.14	236.80	358.05	332.45	<b>346.54</b>	<b>363.47</b>	<b>360.13</b>	<b>359.03</b>
	R	<b>0.40</b>	<b>0.40</b>	0.39	<b>0.40</b>	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	<b>-33.56</b>	-34.16	-34.37

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NGE (%)	65.84	65.23	59.55	59.05	59.49	59.65	<b>59.32</b>	59.66
RMSE	505.64	502.24	459.64	460.51	<b>459.50</b>	470.39	467.90	469.19

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Table S4. Effects of aerosol feedbacks (ARI and/or ACI) considered in different coupled models on *statistical* metrics between annual and seasonal meteorological and air quality simulations and observations in eastern China.

Surface observations		WRF-CMAQ_ARI	WRF-Chem_ARI	WRF-Chem_ACI	WRF-Chem_BOTH	WRF-CHIMERE_ARI	WRF-CHIMERE_ACI	WRF-CHIMERE_BOTH
SSR	Annual	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)
	Spring	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)
	Summer	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↓)
	Autumn	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)
	Winter	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)
T2	Annual	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Spring	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Summer	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↑)
	Autumn	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)
	Winter	R(↑), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↑), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)
SH2	Annual	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Spring	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)
	Summer	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Autumn	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↑), MB(↑), RMSE(↑)
	Winter	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
Q2	Annual	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)
	Spring	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↑), RMSE(↑)
	Summer	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↓)
	Autumn	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Winter	R(↓), MB(↓), RMSE(↓)	R(↑), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↑), RMSE(↑)
WS10	Annual	R(↓), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↓)	R(↑), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↓)
	Spring	R(↓), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Summer	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↑), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↑), MB(↓), RMSE(↓)
	Autumn	R(↓), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)
	Winter	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↑)	R(↑), MB(↓), RMSE(↓)	R(↓), MB(↑), RMSE(↑)	R(↓), MB(↓), RMSE(↓)









1   **Table S5. Radiation variables used in the two-way coupled WRF-CMAQ, WRF-Chem  
2   and WRF-CHIMERE models with only enabling ARI compared to without aerosol  
3   feedbacks.**

Model	SW/LW radiation schemes	Turning off feedback	Turning on ARI feedback	
			Direct effects	Semi-direct effects
WRF-CMAQ	RRTMG/RRTMG	Aerosol optical properties are not calculated	Aerosol extinction, single scattering albedo ( $\omega_0$ ), and asymmetry factor (g) 14 shortwave bands and 5 longwave bands (Wong et al., 2012)	1. Solar uv and ir fluxes 2. Radiative heating rate for the ttenld variable
WRF-Chem	RRTMG/RRTMG	Aerosol optical properties are not calculated	$\omega_0$ (300 nm, 400 nm, 600 nm, 999 nm), g (300 nm, 400 nm, 600 nm, 999 nm), AOD ( $\tau$ ) (300 nm, 400 nm, 600 nm, 999 nm, 16 bands 3400 nm to 55600 nm) (Zhao et al., 2011)	1. Solar uv and ir fluxes 2. Radiative heating rate for the ttenld variable
WRF-CHIMERE	RRTMG/RRTMG	Aerosol optical properties are not calculated	$\omega_0$ (400 nm, 600 nm), g (400 nm, 600 nm), AOD (300 nm, 400 nm, 999 nm, 16 bands 3400 nm to 55600 nm) (Briant et al., 2017)	1. Solar uv and ir fluxes 2. Radiative heating rate for the ttenld variable

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5   **Table S6. Description of refractive indices and radiation schemes used in the WRF-  
6   CMAQ, WRF-Chem and WRF-CHIMERE models.**

Model	Refractive indices of aerosol species groups		
	SW	LW	
WRF-CMAQ	1. Water (1.408+1.420×10 <sup>-3</sup> i, 1.324+1.577×10 <sup>-4</sup> i, 1.277+1.516×10 <sup>-5</sup> i, 1.302+1.159×10 <sup>-5</sup> i, 1.312+2.360×10 <sup>-4</sup> i, 1.321+1.713×10 <sup>-4</sup> i, 1.323+2.425×10 <sup>-5</sup> i, 1.327+3.125×10 <sup>-5</sup> i, 1.331+3.405×10 <sup>-5</sup> i, 1.334+1.639×10 <sup>-6</sup> i, 1.340+2.955×10 <sup>-6</sup> i, 1.349+1.635×10 <sup>-6</sup> i, 1.362+3.350×10 <sup>-6</sup> i, 1.260+6.220×10 <sup>-7</sup> i) 2. Water-soluble (1.443+5.718×10 <sup>-4</sup> i, 1.420+1.777×10 <sup>-4</sup> i, 1.420+1.060×10 <sup>-4</sup> i, 1.420+8.368×10 <sup>-5</sup> i, 1.463+1.621×10 <sup>-5</sup> i, 1.510+2.198×10 <sup>-5</sup> i, 1.510+1.929×10 <sup>-5</sup> i, 1.520+1.564×10 <sup>-5</sup> i, 1.530+7.000×10 <sup>-6</sup> i, 1.530+5.666×10 <sup>-6</sup> i, 1.530+5.000×10 <sup>-6</sup> i, 1.530+8.440×10 <sup>-6</sup> i, 1.530+3.000×10 <sup>-7</sup> i, 1.710+1.100×10 <sup>-7</sup> i) 3. BC (2.089+1.070i, 2.014+0.939i, 1.962+0.843i, 1.950+0.784i, 1.940+0.760i, 1.930+0.749i, 1.905+0.737i, 1.870+0.726i, 1.850+0.710i, 1.850+0.710i, 1.850+0.710i, 1.850+0.710i, 1.850+0.710i, 2.589+1.771i) 4. Insoluble (1.272+1.165×10 <sup>-4</sup> i, 1.168+1.073×10 <sup>-5</sup> i, 1.208+8.650×10 <sup>-5</sup> i, 1.253+8.092×10 <sup>-5</sup> i, 1.329+8.000×10 <sup>-5</sup> i, 1.418+8.000×10 <sup>-6</sup> i, 1.456+8.000×10 <sup>-6</sup> i, 1.518+8.000×10 <sup>-6</sup> i, 1.530+8.000×10 <sup>-6</sup> i, 1.530+8.000×10 <sup>-7</sup> i, 1.530+8.000×10 <sup>-7</sup> i, 1.530+8.440×10 <sup>-7</sup> i, 1.530+3.000×10 <sup>-7</sup> i, 1.470+9.000×10 <sup>-8</sup> i) 5. Sea-salt (1.480+1.758×10 <sup>-4</sup> i, 1.534+7.462×10 <sup>-5</sup> i, 1.437+2.950×10 <sup>-5</sup> i, 1.448+1.276×10 <sup>-5</sup> i, 1.450+7.944×10 <sup>-5</sup> i, 1.462+5.382×10 <sup>-6</sup> i, 1.469+3.754×10 <sup>-6</sup> i, 1.470+1.498×10 <sup>-6</sup> i, 1.490+2.050×10 <sup>-6</sup> i, 1.500+1.184×10 <sup>-6</sup> i, 1.502+9.938×10 <sup>-6</sup> i, 1.510+2.060×10 <sup>-6</sup> i, 1.510+5.000×10 <sup>-6</sup> i, 1.510+1.000×10 <sup>-7</sup> i) in terms of 14 wavelengths at 3.4615, 2.7885, 2.325, 2.046, 1.784, 1.4625, 1.2705, 1.0101, 0.7016, 0.53325, 0.38815, 0.299, 0.2316, 8.24 $\mu\text{m}$	1. Water (1.160+0.321i, 1.140+0.117i, 1.232+0.047i, 1.266+0.038i, 1.300+0.034i) 2. Water-soluble (1.570+0.069i, 1.700+0.055i, 1.890+0.128i, 2.233+0.334i, 1.220+0.066i) 3. BC (1.570+2.200i, 1.700+2.200i, 1.890+2.200i, 2.233+2.200i, 1.220+2.200i) 4. Insoluble (1.482+0.096i, 1.600+0.107i, 1.739+0.162i, 1.508+0.117i, 1.175+0.042i) 5. Sea-salt (1.410+0.019i, 1.490+0.014i, 1.560+0.017i, 1.600+0.029i, 1.402+0.012i) in terms of 5 thermal windows at 13.240, 11.20, 9.73, 8.870, 7.830 $\mu\text{m}$	
WRF-Chem	1. Water (1.35+1.524×10 <sup>-4</sup> i, 1.34+2.494×10 <sup>-4</sup> i, 1.33+1.638×10 <sup>-4</sup> i, 1.33+3.128×10 <sup>-4</sup> i) 2. Dust (1.55+0.003i, 1.550+0.003i, 1.550+0.003i, 1.550+0.003i) 3. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 4. OC (1.45+0i, 1.45+0i, 1.45+0i, 1.45+0i) 5. Sea salt (1.51+8.66×10 <sup>-7</sup> i, 1.5+7.019×10 <sup>-8</sup> i, 1.5+1.184×10 <sup>-8</sup> i, 1.47+1.5×10 <sup>-8</sup> i) 6. Sulfate (1.52+1.00×10 <sup>-6</sup> i, 1.52+1.00×10 <sup>-6</sup> i, 1.52+1.00×10 <sup>-6</sup> i, 1.52+1.75×10 <sup>-6</sup> i) in terms of 4 spectral intervals in 0.25-0.35, 0.35-0.45, 0.55-0.65, 0.998-1.000 $\mu\text{m}$	1. Water (1.532+0.336i, 1.524+0.360i, 1.420+0.426i, 1.274+0.403i, 1.161+0.321i, 1.142+0.115i, 1.232+0.0471i, 1.266+0.039i, 1.296+0.034i, 1.321+0.0344i, 1.342+0.092i, 1.315+0.012i, 1.330+0.013i, 1.339+0.011i, 1.350+0.0049i, 1.408+0.0142i) 2. Dust (2.34+0.7i, 2.904+0.857i, 1.748+0.462i, 1.508+0.263i, 1.911+0.319i, 1.822+0.26i, 2.917+0.65i, 1.557+0.373i, 1.242+0.093i, 1.447+0.105i, 1.432+0.061i, 1.473+0.0245i, 1.495+0.011i, 1.5+0.008i) 3. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 4. OC (1.86+0.5i, 1.91+0.268i, 1.988+0.185i, 1.439+0.198i, 1.606+0.059i, 1.7+0.0488i, 1.888+0.11i, 2.489+0.3345i, 1.219+0.065i, 1.419+0.058i, 1.426+0.0261i, 1.446+0.0142i, 1.457+0.013i, 1.458+0.011i) 5. Sea salt (1.74+0.1978i, 1.76+0.1978i, 1.78+0.129i, 1.456+0.038i, 1.41+0.019i, 1.48+0.014i, 1.56+0.016i, 1.63+0.03i, 1.4+0.012i, 1.43+0.0064i, 1.56+0.0196i, 1.45+0.0029i, 1.485+0.0017i, 1.486+0.0014i) 6. Sulfate (1.89+0.22i, 1.91+0.152i, 1.93+0.0846i, 1.586+0.2225i, 1.678+0.195i, 1.758+0.441i, 1.855+0.696i, 1.597+0.695i, 1.15+0.459i, 1.26+0.161i, 1.42+0.172i, 1.35+0.14i, 1.379+0.12i, 1.385+0.122i) in terms of 16 spectral intervals in 10-350, 350-500, 500-630, 630-700, 700-820, 820-980, 980-1080, 1080-1180, 1180-1390, 1390-1480, 1480-1800, 1800-2080, 2080-2250, 2250-2390, 2390-2600, 2600-3250 $\mu\text{m}$ 7. Water (1.42+0.02i, 1.35+0.0047i, 1.34+0.0085i, 1.33+0.015i, 1.32+0.01i, 1.32+0.13i, 1.32+0.032i, 1.3+0.034i, 1.27+0.039i, 1.23+0.047i, 1.15+0.1i, 1.16+0.32i, 1.27+0.4i, 1.41+0.43i, 1.52+0.37i, 1.65+0.55i) 8. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 9. OC (1.43+1.42i, 1.46+1.43i, 1.46+1.25i, 1.46+2.67i, 1.45+1.89i, 1.42+1.71i, 1.43+1.71i, 1.25+0.07i, 2.67+0.005i, 1.89+0.01i, 1.71+0.013i, 1.43+0.014i, 1.46+0.025i, 1.46+0.062i, 1.46+0.064i, 1.45+0.031i) 10. NH <sub>3</sub> (1.53+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i) in terms of 5 wavelengths at 0.2, 0.3, 0.4, 0.6, 0.999 $\mu\text{m}$	1. Water (1.532+0.336i, 1.524+0.360i, 1.420+0.426i, 1.274+0.403i, 1.161+0.321i, 1.142+0.115i, 1.232+0.0471i, 1.266+0.039i, 1.296+0.034i, 1.321+0.0344i, 1.342+0.092i, 1.315+0.012i, 1.330+0.013i, 1.339+0.011i, 1.350+0.0049i, 1.408+0.0142i) 2. Dust (2.34+0.7i, 2.904+0.857i, 1.748+0.462i, 1.508+0.263i, 1.911+0.319i, 1.822+0.26i, 2.917+0.65i, 1.557+0.373i, 1.242+0.093i, 1.447+0.105i, 1.432+0.061i, 1.473+0.0245i, 1.495+0.011i, 1.5+0.008i) 3. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 4. OC (1.86+0.5i, 1.91+0.268i, 1.988+0.185i, 1.439+0.198i, 1.606+0.059i, 1.7+0.0488i, 1.888+0.11i, 2.489+0.3345i, 1.219+0.065i, 1.419+0.058i, 1.426+0.0261i, 1.446+0.0142i, 1.457+0.013i, 1.458+0.011i) 5. Sea salt (1.74+0.1978i, 1.76+0.1978i, 1.78+0.129i, 1.456+0.038i, 1.41+0.019i, 1.48+0.014i, 1.56+0.016i, 1.63+0.03i, 1.4+0.012i, 1.43+0.0064i, 1.56+0.0196i, 1.45+0.0029i, 1.485+0.0017i, 1.486+0.0014i) 6. Sulfate (1.89+0.22i, 1.91+0.152i, 1.93+0.0846i, 1.586+0.2225i, 1.678+0.195i, 1.758+0.441i, 1.855+0.696i, 1.597+0.695i, 1.15+0.459i, 1.26+0.161i, 1.42+0.172i, 1.35+0.14i, 1.379+0.12i, 1.385+0.122i) in terms of 16 spectral intervals in 10-350, 350-500, 500-630, 630-700, 700-820, 820-980, 980-1080, 1080-1180, 1180-1390, 1390-1480, 1480-1800, 1800-2080, 2080-2250, 2250-2390, 2390-2600, 2600-3250 $\mu\text{m}$ 7. Water (1.42+0.02i, 1.35+0.0047i, 1.34+0.0085i, 1.33+0.015i, 1.32+0.01i, 1.32+0.13i, 1.32+0.032i, 1.3+0.034i, 1.27+0.039i, 1.23+0.047i, 1.15+0.1i, 1.16+0.32i, 1.27+0.4i, 1.41+0.43i, 1.52+0.37i, 1.65+0.55i) 8. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 9. OC (1.43+1.42i, 1.46+1.43i, 1.46+1.25i, 1.46+2.67i, 1.45+1.89i, 1.42+1.71i, 1.43+1.71i, 1.25+0.07i, 2.67+0.005i, 1.89+0.01i, 1.71+0.013i, 1.43+0.014i, 1.46+0.025i, 1.46+0.062i, 1.46+0.064i, 1.45+0.031i) 10. NH <sub>3</sub> (1.53+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i) in terms of 5 wavelengths at 0.2, 0.3, 0.4, 0.6, 0.999 $\mu\text{m}$
WRF-CHIMERE	1. Water (1.35+2.0×10 <sup>-4</sup> i, 1.34+2.0×10 <sup>-4</sup> i, 1.34+1.8×10 <sup>-8</sup> i, 1.33+3.4×10 <sup>-8</sup> i, 1.33+3.9×10 <sup>-7</sup> i) 2. Dust (1.53+0.0055i, 1.53+0.0055i, 1.53+0.0024i, 1.53+8.9-4i, 1.53+7.6-4i) 3. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 4. OC (1.53+0.09i, 1.53+0.008i, 1.53+0.005i, 1.53+0.0063i, 1.52+0.016i) 5. Sea salt (1.38+8.7×10 <sup>-7</sup> i, 1.38+3.5×10 <sup>-7</sup> i, 1.37+6.6×10 <sup>-8</sup> i, 1.36+1.2×10 <sup>-8</sup> i, 1.35+2.6×10 <sup>-8</sup> i) 6. PPM (1.53+0.008i, 1.52+0.008i, 1.52+0.008i, 1.51+0.008i, 1.5+0.008i) 7. SOA (1.56+0.0003i, 1.56+0.0003i, 1.56+0.0003i, 1.56+0.0003i) 8. H <sub>2</sub> SO <sub>4</sub> (1.5+1.0×10 <sup>-8</sup> i, 1.47+1.0×10 <sup>-8</sup> i, 1.44+1.0×10 <sup>-8</sup> i, 1.43+1.3×10 <sup>-8</sup> i, 1.42+1.2×10 <sup>-8</sup> i) 9. HNO <sub>3</sub> (1.53+0.006i, 1.53+0.006i, 1.53+0.006i, 1.53+0.006i) 10. NH <sub>3</sub> (1.53+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i, 1.52+0.0005i) in terms of 5 wavelengths at 0.2, 0.3, 0.4, 0.6, 0.999 $\mu\text{m}$	1. Water (1.42+0.02i, 1.35+0.0047i, 1.34+0.0085i, 1.33+0.015i, 1.32+0.01i, 1.32+0.13i, 1.32+0.032i, 1.3+0.034i, 1.27+0.039i, 1.23+0.047i, 1.15+0.1i, 1.16+0.32i, 1.27+0.4i, 1.41+0.43i, 1.52+0.37i, 1.65+0.55i) 2. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i) 3. OC (1.43+1.42i, 1.46+1.43i, 1.46+1.25i, 1.46+2.67i, 1.45+1.89i, 1.42+1.71i, 1.43+1.71i, 1.25+0.07i, 2.67+0.005i, 1.89+0.01i, 1.71+0.013i, 1.43+0.014i, 1.46+0.025i, 1.46+0.062i, 1.46+0.064i, 1.45+0.031i) 4. Salt (1.43+0.019i, 1.37+0.0043i, 1.36+0.0084i, 1.36+0.011i, 1.34+0.01i, 1.35+0.083i, 1.34+0.029i, 1.31+0.03i, 1.33+0.037i, 1.29+0.042i, 1.2+0.09i, 1.2+0.27i, 1.3+0.34i, 1.47+0.37i, 1.56+0.03i, 1.51+0.09i) 5. PPM (1.45+0.001i, 1.45+0.01i, 1.45+0.01i, 1.45+0.01i, 1.45+0.01i, 1.45+0.01i, 1.45+0.05i, 1.45+0.05i, 1.45+0.05i, 2.6+0.2i, 1.7+0.2i, 1.7+0.2i, 1.7+0.2i, 1.7+0.2i, 1.7+0.2i) 6. SOA (1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i, 1.56+0.003i)	

7.  $\text{H}_2\text{SO}_4$  ( $1.35 \pm 0.16$ i,  $1.4 \pm 0.13$ i,  $1.39 \pm 0.12$ i,  $1.38 \pm 0.12$ i,  $1.35 \pm 0.15$ i,  $1.42 \pm 0.18$ i,  $1.26 \pm 0.16$ i,  
 1.15  $\pm 0.44$ i,  $1.57 \pm 0.73$ i,  $1.83 \pm 0.7$ i,  $1.71 \pm 0.46$ i,  $1.68 \pm 0.2$ i,  $1.59 \pm 0.21$ i,  $1.87 \pm 0.48$ i,  $1.89 \pm 0.27$ i,  
 1.86  $\pm 0.31$ i)  
 8.  $\text{HNO}_3$  ( $1.45 \pm 0.01$ i,  $1.45 \pm 0.05$ i,  
 1.0*S<sub>i</sub>*,  $1 \pm 0.2$ i,  $2.6 \pm 0.2$ i,  $1.7 \pm 0.2$ i)  
 9.  $\text{NH}_3$  ( $1.45 \pm 0.01$ i,  $1.45 \pm 0.05$ i,  $1 \pm 0.5$ i,  
 1.0*S<sub>i</sub>*,  $2.6 \pm 0.2$ i,  $1.7 \pm 0.2$ i) in term of 16  
 wavelengths at  $3.4, 4, 4.3, 4.6, 5.2, 6.1, 7.0, 7.8, 8.8, 9.7, 11.1, 13.2, 15.0, 17.7, 23.5, 55.6 \mu\text{m}$

1

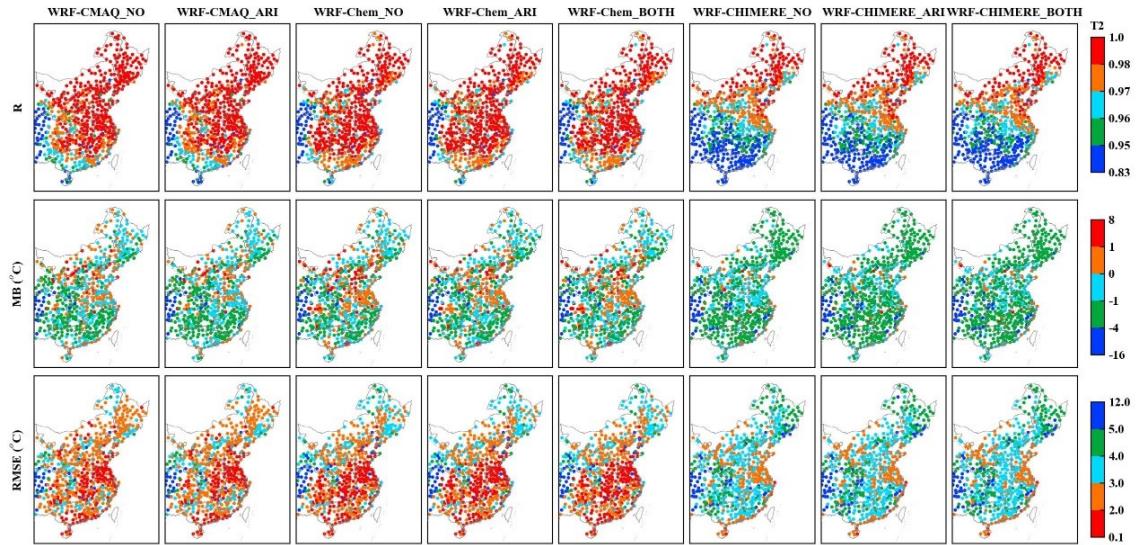
2 **Table S7. Microphysics variables used in the two-way coupled WRF-CMAQ, WRF-  
 3 Chem and WRF-CHIMERE models with enabling ACI effects compared to those  
 4 without aerosol feedbacks.**

Model	Microphysics scheme	Turning off feedback	Turning on ACI feedback	Second indirect effects
			First indirect effects	
WRF-CMAQ	Morrison	1. Prescribed constant CDNC value of $250 \text{ cm}^{-3}$	None	None
WRF-Chem	Morrison	1. Prescribed constant CDNC value of $250 \text{ cm}^{-3}$ 2. Constant cloud droplet effective radius with $10 \mu\text{m}$ 3. Cloud droplet extinction coefficient, single scattering albedo, and asymmetry factor 4. Prescribed ice nucleating particle (INP) concentration based on empirical formula (Rasmussen et al., 2002)	1. Hygroscopicity 2. Prognostic CDNC based on Köhler theory 3. Prognostic cloud droplet effective radius 4. Prognostic cloud droplet extinction coefficient, single scattering albedo, and asymmetry factor 5. Prescribed INP	1. Prognostic cloud-to-rain autoconversion rate
WRF-CHIMERE	Thompson	1. Prescribed constant CDNC values of $300 \text{ cm}^{-3}$ 2. Prescribed INP from heterogeneous ice nucleation in iceDeMott subroutine module_mp_thompson.F file using climatological dust concentration (dimeters $> 0.5 \mu\text{m}$ ) (DeMott et al., 2015) and homogeneous freezing (Thompson and Eidhammer, 2014) with climatological hygroscopic aerosol concentrations (dimeters $> 0.1 \mu\text{m}$ ) generated by QNWFA_QNIFA_Monthly_GFS file 3. Prescribed cloud droplet and ice effective radius 4. Prescribed extinction coefficient, single scattering albedo, and asymmetry factor of cloud droplet and ice	1. Hygroscopicity 2. Prognostic CDNC based on Köhler theory 3. Prognostic INP from heterogeneous ice nucleation based on online dust calculation (dimeters $> 0.5 \mu\text{m}$ ) and homogeneous freezing with prognostic hygroscopic aerosol concentrations (dimeters $> 0.1 \mu\text{m}$ ) (Tuccella et al., 2019) 4. Prognostic cloud droplet and ice effective radius 5. Prognostic extinction coefficient, single scattering albedo, and asymmetry factor of cloud droplet and ice	1. Prognostic cloud-to-rain autoconversion rate

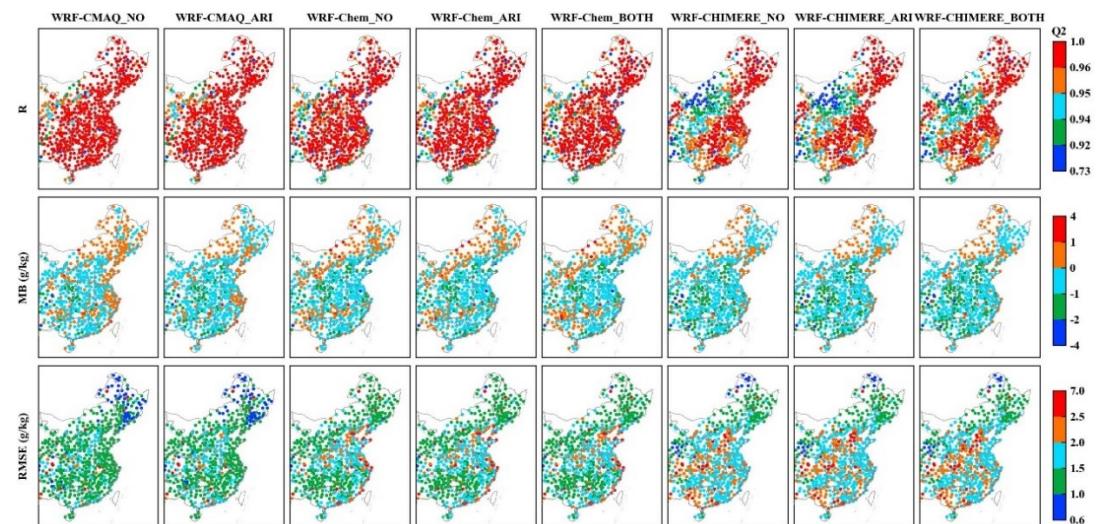
5

6 **Table S8. Summary of download information on model output of each simulation  
 7 scenario.**

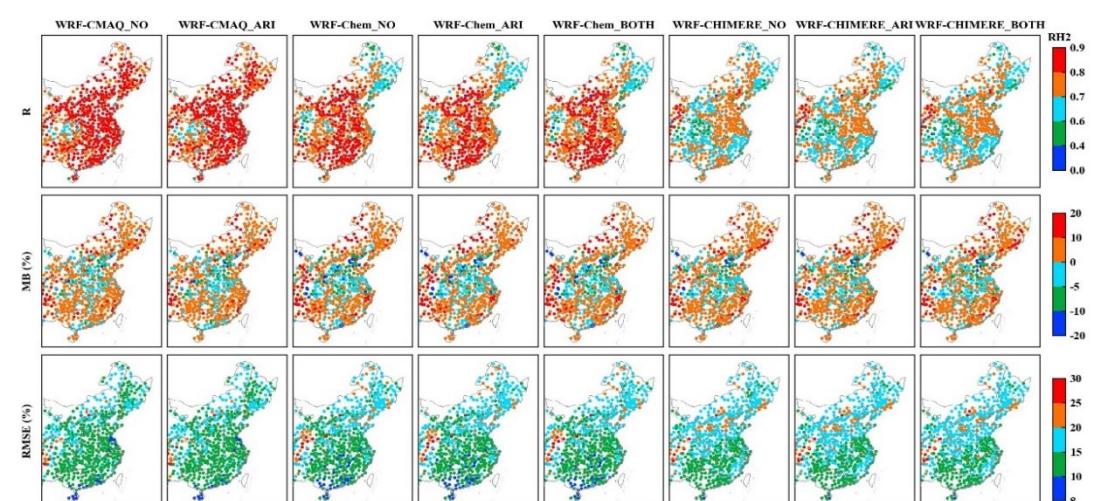
Scenario	DOI	Link	Reference
WRF-CMAQ_NO	<a href="https://doi.org/10.5281/zenodo.7951404">https://doi.org/10.5281/zenodo.7951404</a> <a href="https://doi.org/10.5281/zenodo.7951467">https://doi.org/10.5281/zenodo.7951467</a> <a href="https://doi.org/10.5281/zenodo.7951475">https://doi.org/10.5281/zenodo.7951475</a>	<a href="https://zenodo.org/record/7951404">https://zenodo.org/record/7951404</a> <a href="https://zenodo.org/record/7951467">https://zenodo.org/record/7951467</a> <a href="https://zenodo.org/record/7951475">https://zenodo.org/record/7951475</a>	Gao et al., 2023i_part1 Gao et al., 2023i_part2 Gao et al., 2023i_part3
WRF-CMAQ_ARI	<a href="https://doi.org/10.5281/zenodo.7949895">https://doi.org/10.5281/zenodo.7949895</a> <a href="https://doi.org/10.5281/zenodo.7950644">https://doi.org/10.5281/zenodo.7950644</a> <a href="https://doi.org/10.5281/zenodo.7950830">https://doi.org/10.5281/zenodo.7950830</a>	<a href="https://zenodo.org/record/7949895">https://zenodo.org/record/7949895</a> <a href="https://zenodo.org/record/7950644">https://zenodo.org/record/7950644</a> <a href="https://zenodo.org/record/7950830">https://zenodo.org/record/7950830</a>	Gao et al., 2023j_part1 Gao et al., 2023j_part2 Gao et al., 2023j_part3
WRF-Chem_NO	<a href="https://doi.org/10.5281/zenodo.7943804">https://doi.org/10.5281/zenodo.7943804</a> <a href="https://doi.org/10.5281/zenodo.7945383">https://doi.org/10.5281/zenodo.7945383</a> <a href="https://doi.org/10.5281/zenodo.7946944">https://doi.org/10.5281/zenodo.7946944</a> <a href="https://doi.org/10.5281/zenodo.7947169">https://doi.org/10.5281/zenodo.7947169</a>	<a href="https://zenodo.org/record/7943804">https://zenodo.org/record/7943804</a> <a href="https://zenodo.org/record/7945383">https://zenodo.org/record/7945383</a> <a href="https://zenodo.org/record/7946944">https://zenodo.org/record/7946944</a> <a href="https://zenodo.org/record/7947169">https://zenodo.org/record/7947169</a>	Gao et al., 2023k_part1 Gao et al., 2023k_part2 Gao et al., 2023k_part3 Gao et al., 2023k_part4
WRF-Chem_ARI	<a href="https://doi.org/10.5281/zenodo.7947050">https://doi.org/10.5281/zenodo.7947050</a> <a href="https://doi.org/10.5281/zenodo.7948216">https://doi.org/10.5281/zenodo.7948216</a> <a href="https://doi.org/10.5281/zenodo.7949410">https://doi.org/10.5281/zenodo.7949410</a> <a href="https://doi.org/10.5281/zenodo.7949561">https://doi.org/10.5281/zenodo.7949561</a>	<a href="https://zenodo.org/record/7947050">https://zenodo.org/record/7947050</a> <a href="https://zenodo.org/record/7948216">https://zenodo.org/record/7948216</a> <a href="https://zenodo.org/record/7949410">https://zenodo.org/record/7949410</a> <a href="https://zenodo.org/record/7949561">https://zenodo.org/record/7949561</a>	Gao et al., 2023l_part1 Gao et al., 2023l_part2 Gao et al., 2023l_part3 Gao et al., 2023l_part4
WRF-Chem_BOTH	<a href="https://doi.org/10.5281/zenodo.7939221">https://doi.org/10.5281/zenodo.7939221</a> <a href="https://doi.org/10.5281/zenodo.7943002">https://doi.org/10.5281/zenodo.7943002</a> <a href="https://doi.org/10.5281/zenodo.7943079">https://doi.org/10.5281/zenodo.7943079</a> <a href="https://doi.org/10.5281/zenodo.7943323">https://doi.org/10.5281/zenodo.7943323</a>	<a href="https://zenodo.org/record/7939221">https://zenodo.org/record/7939221</a> <a href="https://zenodo.org/record/7943002">https://zenodo.org/record/7943002</a> <a href="https://zenodo.org/record/7943079">https://zenodo.org/record/7943079</a> <a href="https://zenodo.org/record/7943323">https://zenodo.org/record/7943323</a>	Gao et al. 2023m_part1 Gao et al. 2023m_part2 Gao et al. 2023m_part3 Gao et al. 2023n_part4
WRF-CHIMERE_NO	<a href="https://doi.org/10.5281/zenodo.7951775">https://doi.org/10.5281/zenodo.7951775</a> <a href="https://doi.org/10.5281/zenodo.7951779">https://doi.org/10.5281/zenodo.7951779</a> <a href="https://doi.org/10.5281/zenodo.7951791">https://doi.org/10.5281/zenodo.7951791</a> <a href="https://doi.org/10.5281/zenodo.7951793">https://doi.org/10.5281/zenodo.7951793</a>	<a href="https://zenodo.org/record/7951775">https://zenodo.org/record/7951775</a> <a href="https://zenodo.org/record/7951779">https://zenodo.org/record/7951779</a> <a href="https://zenodo.org/record/7951791">https://zenodo.org/record/7951791</a> <a href="https://zenodo.org/record/7951793">https://zenodo.org/record/7951793</a>	Gao et al. 2023n_part1 Gao et al. 2023n_part2 Gao et al. 2023n_part3 Gao et al. 2023n_part4
WRF-CHIMERE_ARI	<a href="https://doi.org/10.5281/zenodo.7952838">https://doi.org/10.5281/zenodo.7952838</a> <a href="https://doi.org/10.5281/zenodo.7952840">https://doi.org/10.5281/zenodo.7952840</a> <a href="https://doi.org/10.5281/zenodo.7952842">https://doi.org/10.5281/zenodo.7952842</a> <a href="https://doi.org/10.5281/zenodo.7952844">https://doi.org/10.5281/zenodo.7952844</a>	<a href="https://zenodo.org/record/7952838">https://zenodo.org/record/7952838</a> <a href="https://zenodo.org/record/7952840">https://zenodo.org/record/7952840</a> <a href="https://zenodo.org/record/7952842">https://zenodo.org/record/7952842</a> <a href="https://zenodo.org/record/7952844">https://zenodo.org/record/7952844</a>	Gao et al. 2023o_part1 Gao et al. 2023o_part2 Gao et al. 2023o_part3 Gao et al. 2023o_part4
WRF-CHIMERE_BOTH	<a href="https://doi.org/10.5281/zenodo.7952859">https://doi.org/10.5281/zenodo.7952859</a> <a href="https://doi.org/10.5281/zenodo.7952863">https://doi.org/10.5281/zenodo.7952863</a> <a href="https://doi.org/10.5281/zenodo.7952865">https://doi.org/10.5281/zenodo.7952865</a> <a href="https://doi.org/10.5281/zenodo.7952867">https://doi.org/10.5281/zenodo.7952867</a>	<a href="https://zenodo.org/record/7952859">https://zenodo.org/record/7952859</a> <a href="https://zenodo.org/record/7952863">https://zenodo.org/record/7952863</a> <a href="https://zenodo.org/record/7952865">https://zenodo.org/record/7952865</a> <a href="https://zenodo.org/record/7952867">https://zenodo.org/record/7952867</a>	Gao et al. 2023p_part1 Gao et al. 2023p_part2 Gao et al. 2023p_part3 Gao et al. 2023p_part4



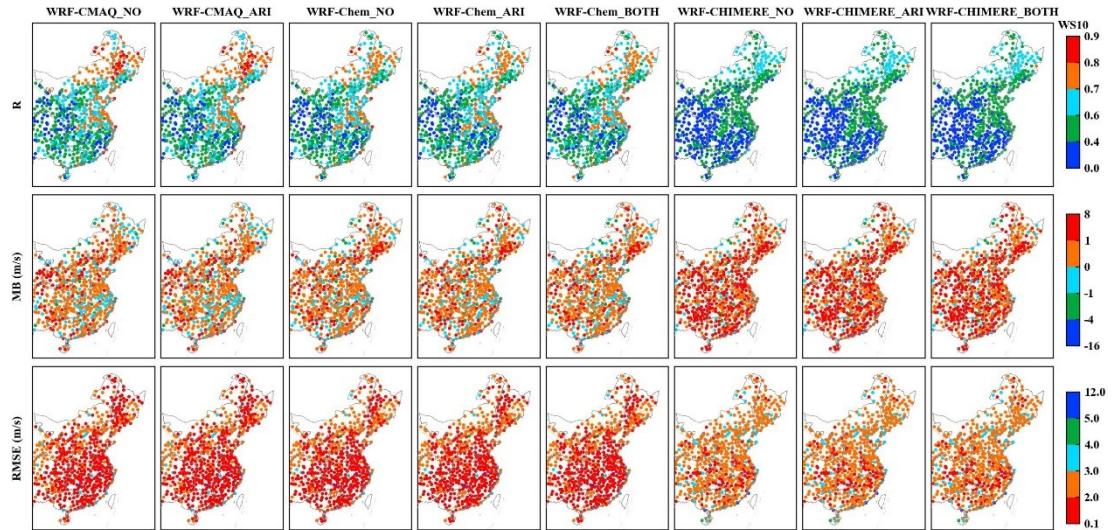
1  
2 Figure S1. Statistical metrics (R, MB and RMSE) between simulated and observed  
3 annual T2 in eastern China.  
4



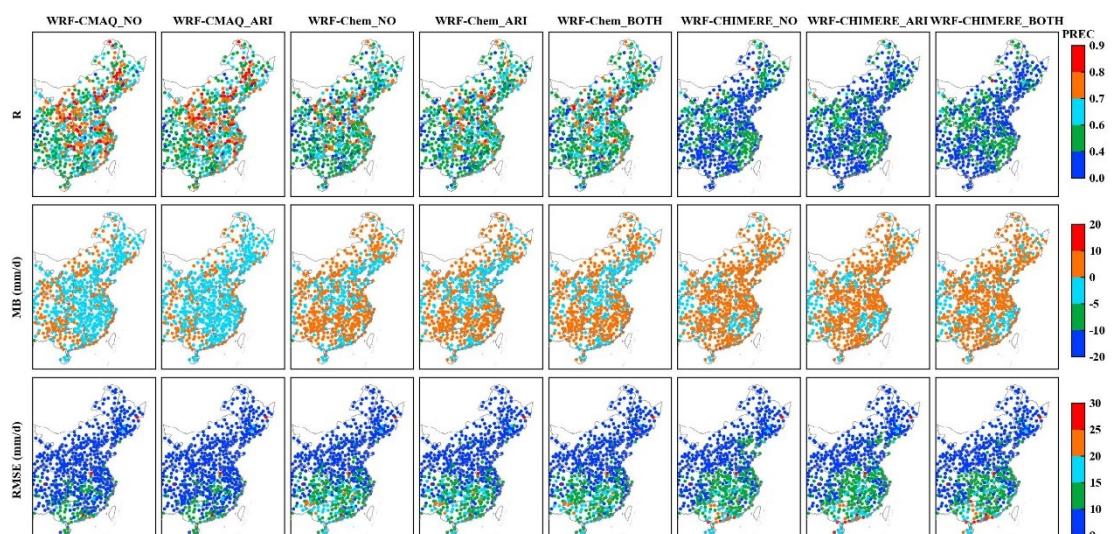
5  
6 Figure S2. The same as Fig. S1 but for Q2.  
7



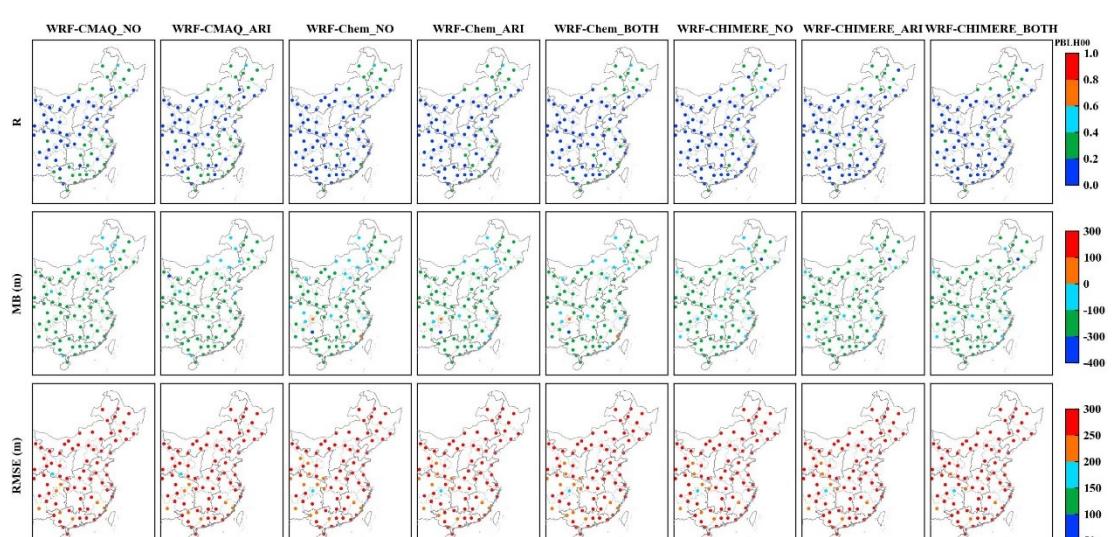
8  
9 Figure S3. The same as Fig. S1 but for RH2.  
10



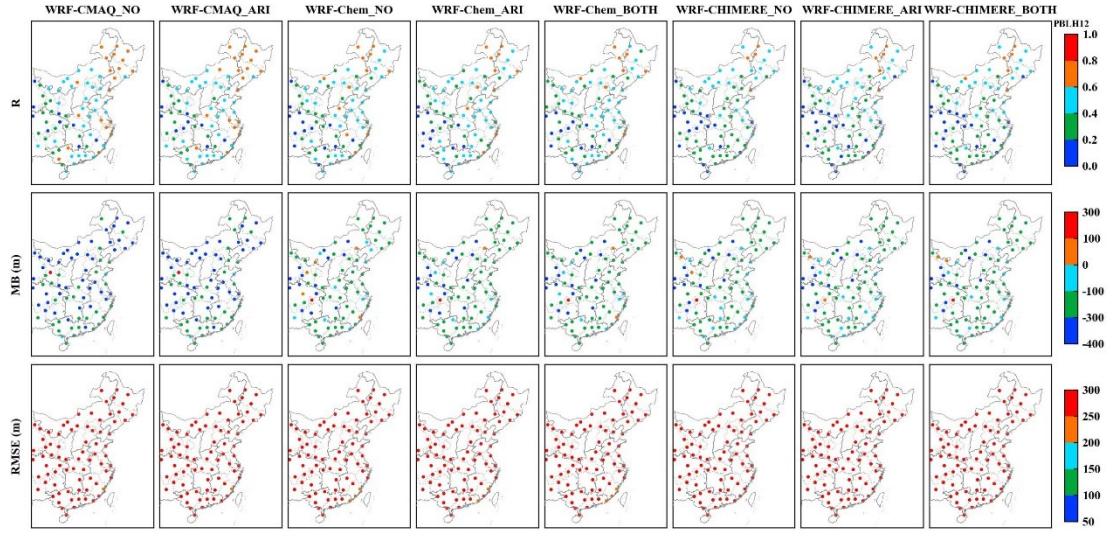
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2 Figure S4. The same as Fig. S1 but for WS10.  
3



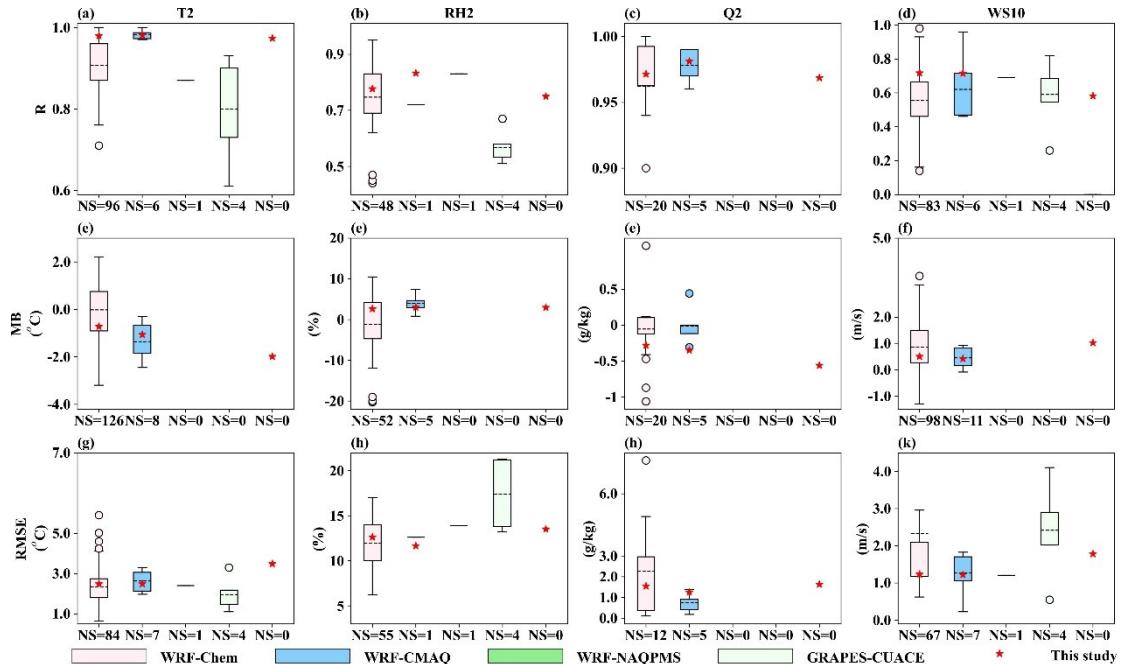
4  
5 Figure S5. The same as Fig. S1 but for PREC.  
6



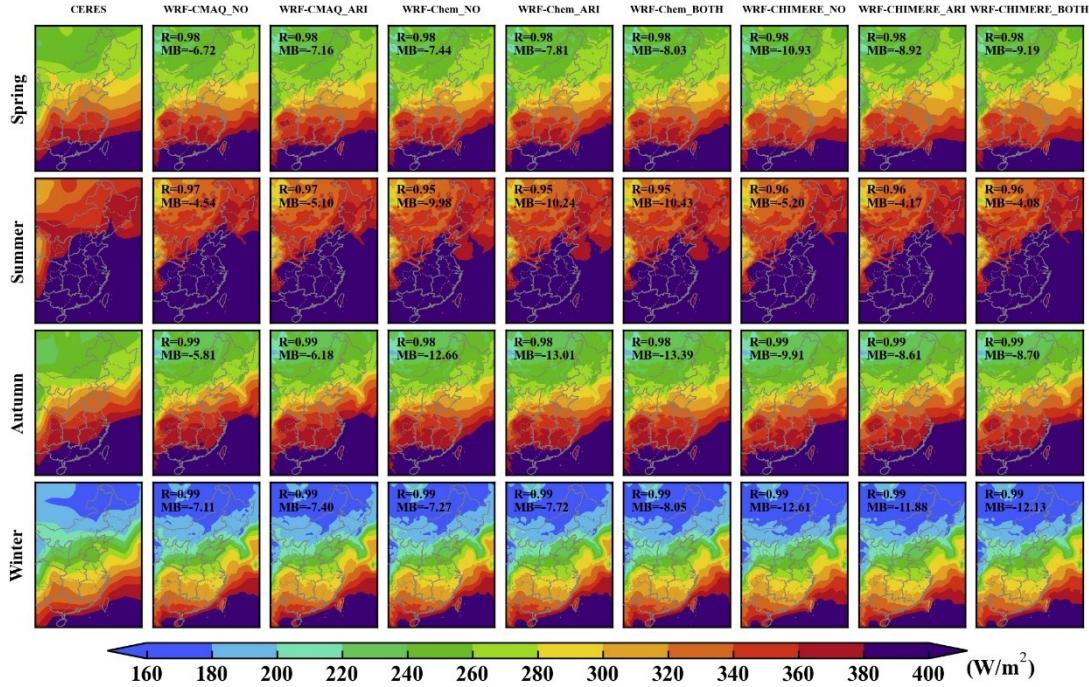
7  
8 Figure S6. The same as Fig. S1 but for PBLH at 08:00 LT.  
9



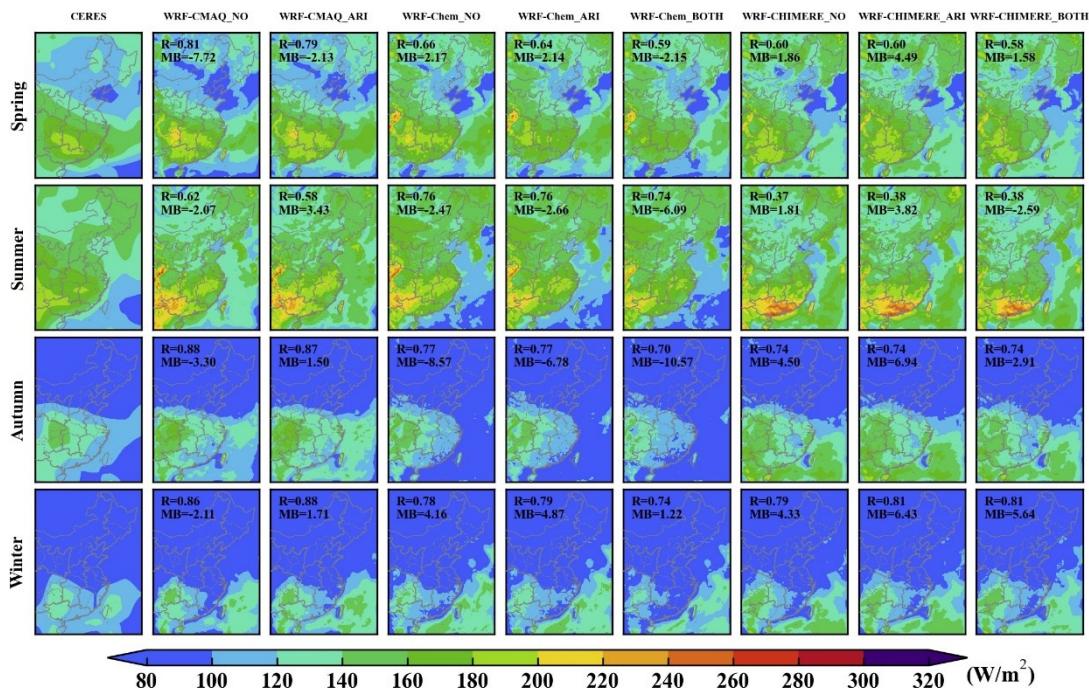
1  
2 Figure S7. The same as Fig. S1 but for PBLH at 20:00 LT.  
3



4  
5 Figure S8. Comparisons of model capacities between our study (red stars)  
6 and previous literature (box plots) in terms of the surface T2, RH2, Q2, and WS10 in eastern China.  
7 Note that red stars in the fifth column of each subgraph represent the statistical metrics  
8 of WRF-CHIMERE in this study.  
9



1  
2 Figure S9. Spatial distributions of seasonal SLR between CERES observations and  
3 simulations from WRF-CMAQ, WRF-Chem, and WRF-CHIMERE with and without  
4 aerosol feedbacks in eastern China.  
5



6  
7 Figure S10. The same as Fig. S9 but for SRTOA.  
8

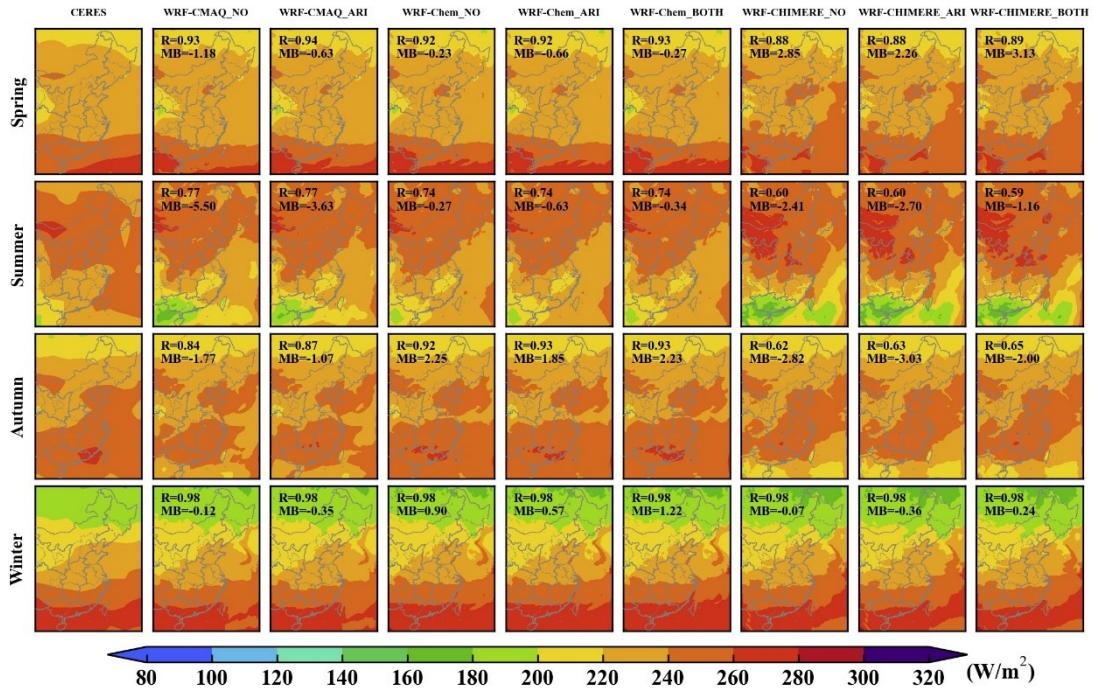


Figure S11. The same as Fig. S9 but for LRTOA.

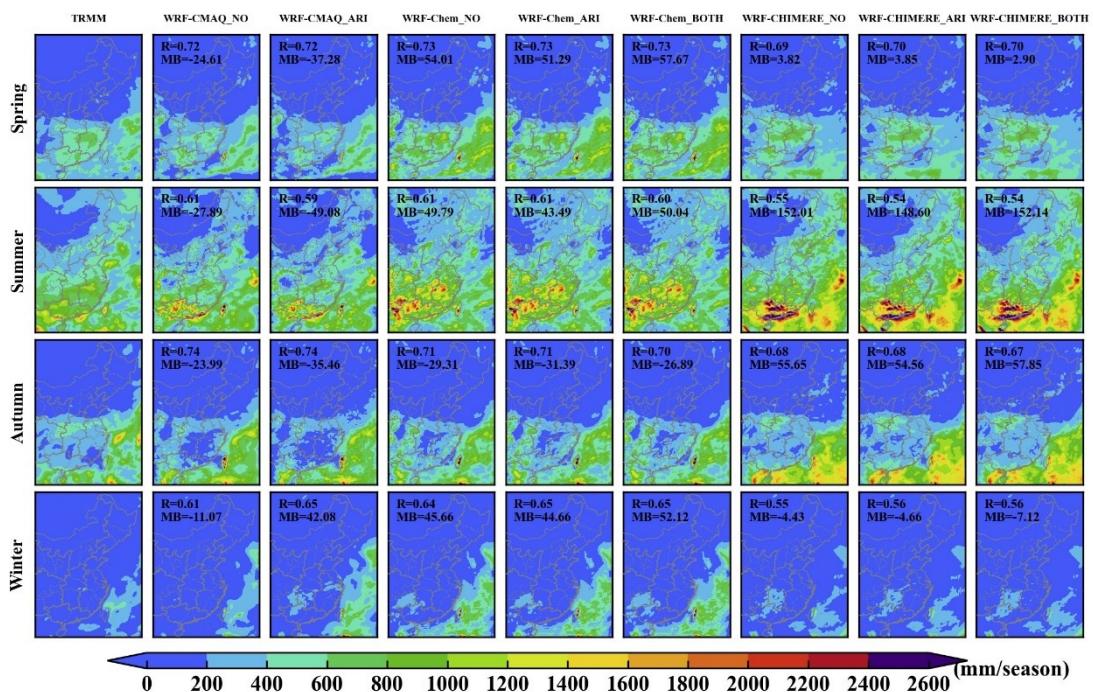
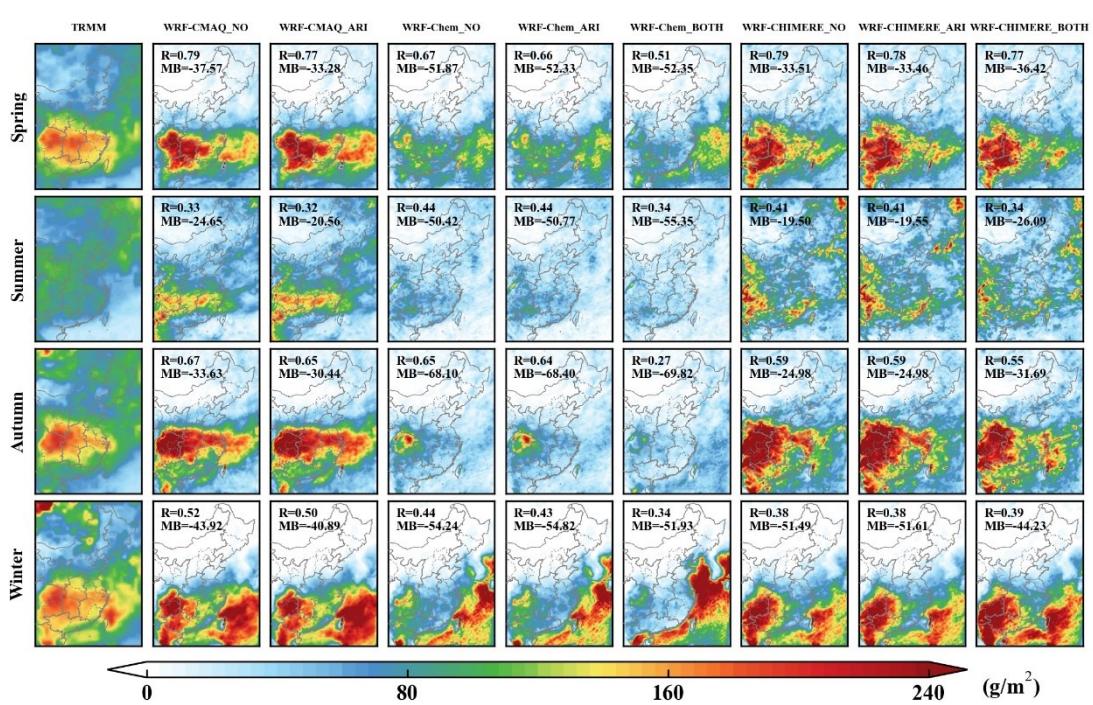
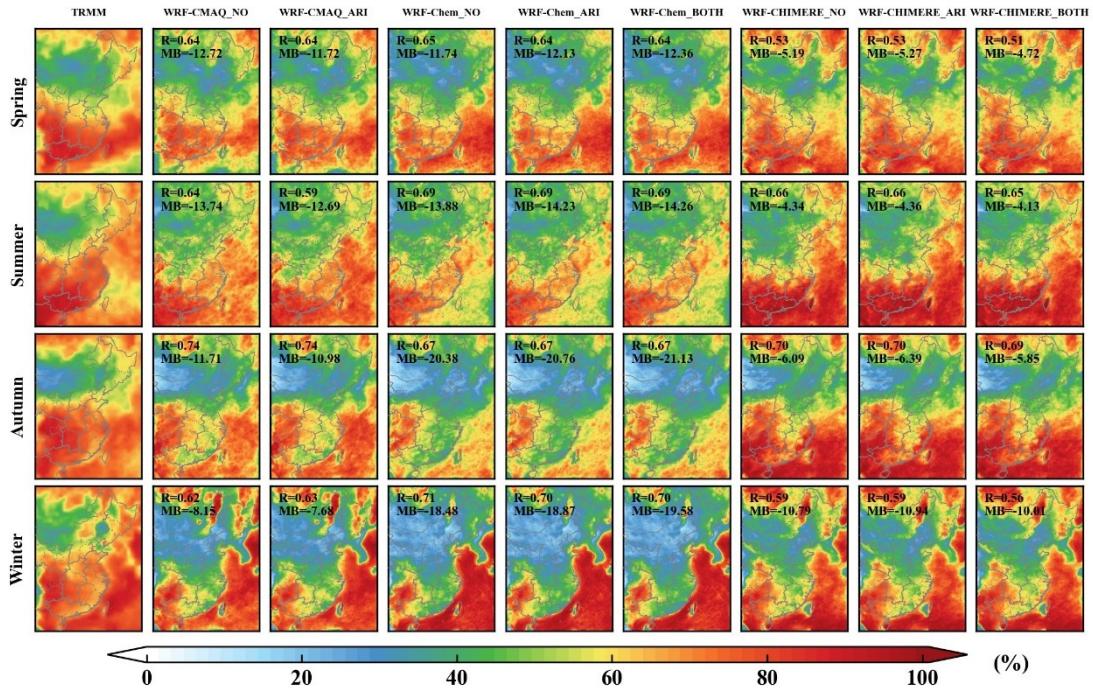


Figure S12. The same as Fig. S9 but for precipitation.



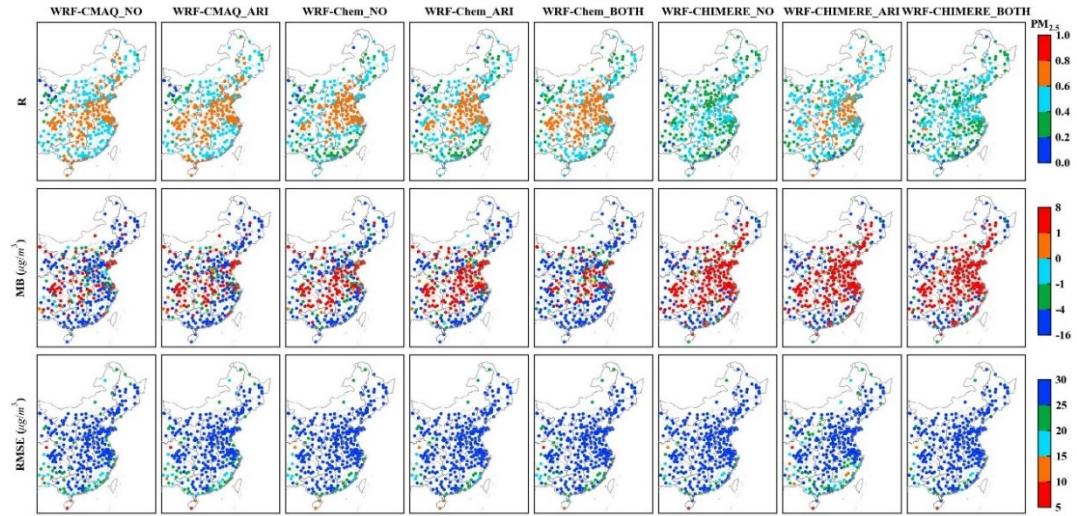


Figure S15. Statistical metrics (R, MB and RMSE) between simulated and observed annual  $\text{PM}_{2.5}$  concentrations in eastern China.

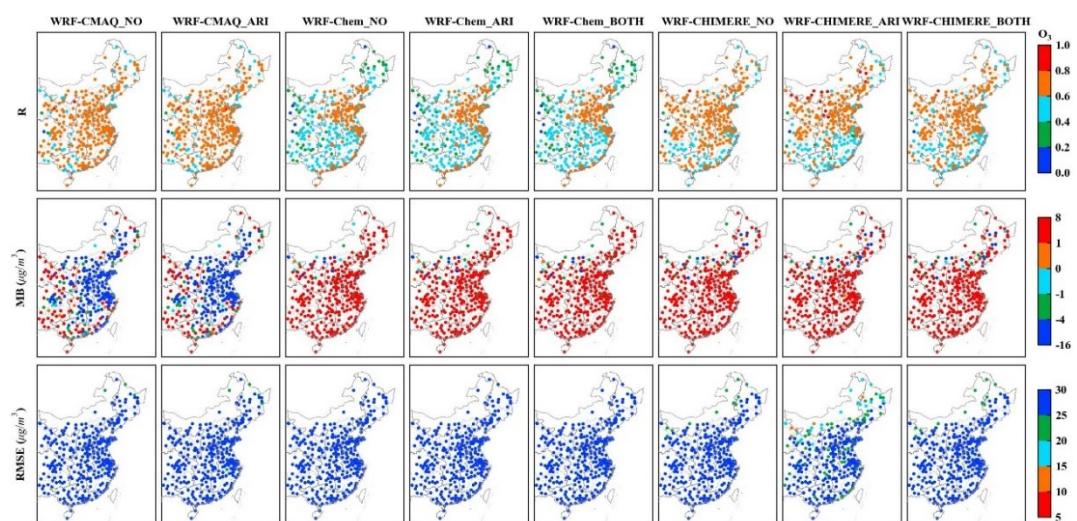


Figure S16. The same as Fig. S15 but for  $\text{O}_3$ .

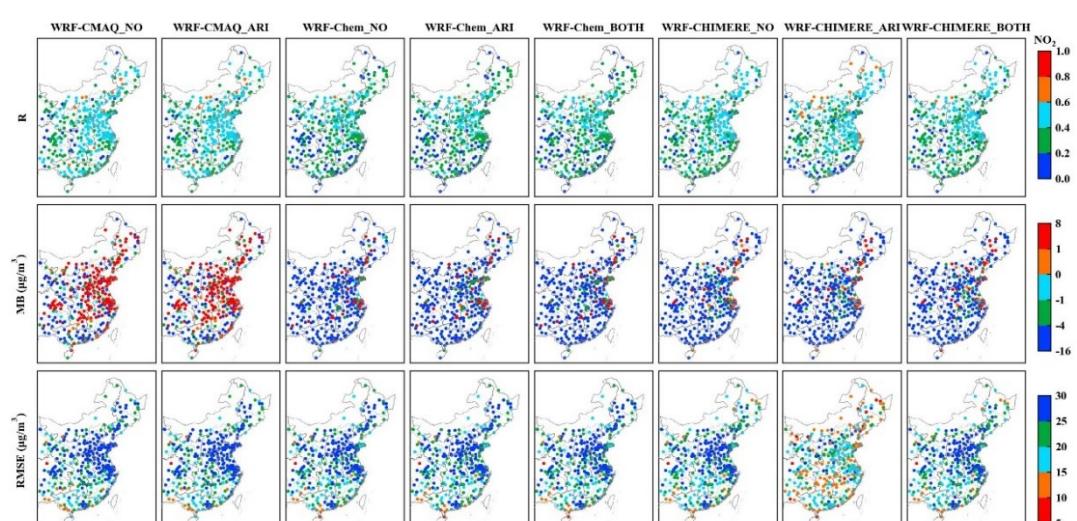


Figure S17. The same as Fig. S15 but for  $\text{NO}_2$ .

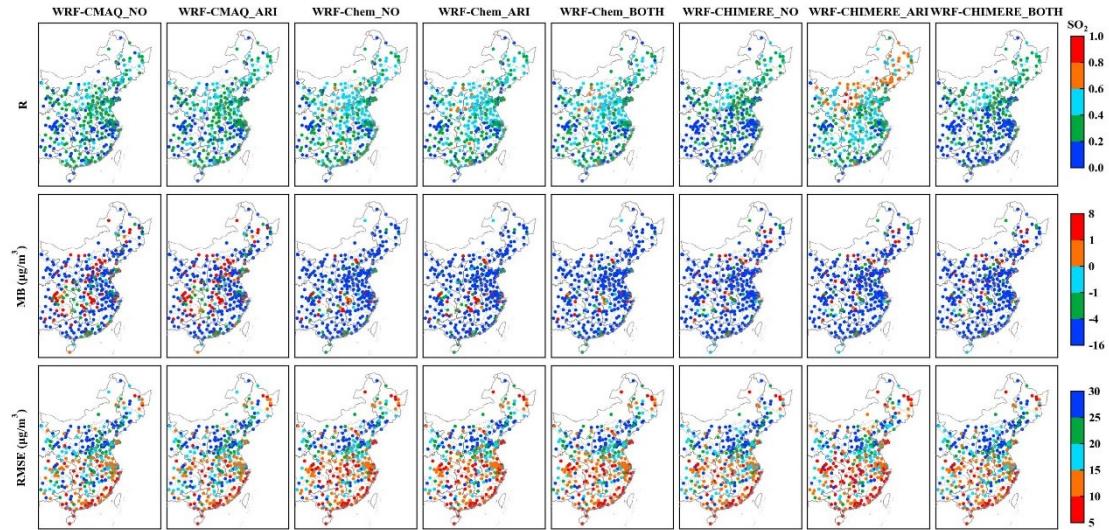


Figure S18. The same as Fig. S15 but for  $\text{SO}_2$ .

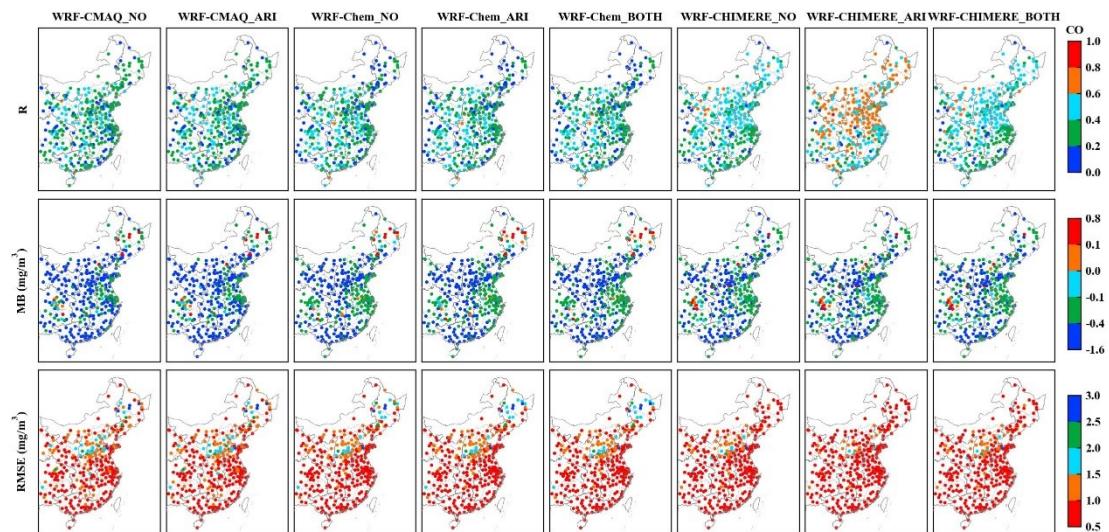


Figure S19. The same as Fig. S15 but for CO.

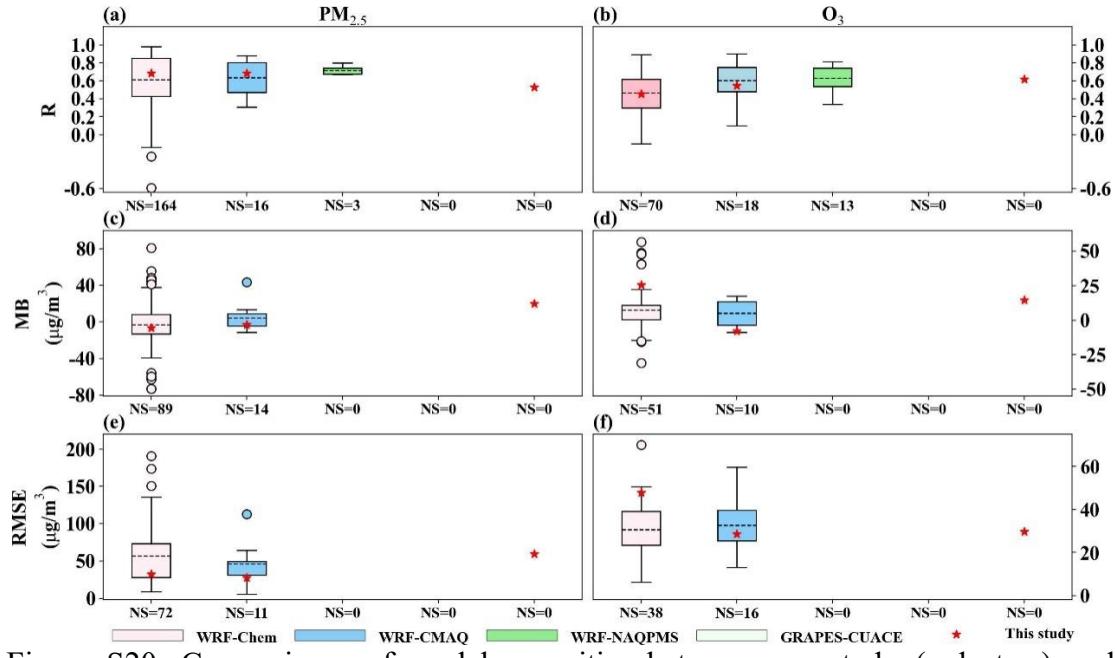


Figure S20. Comparisons of model capacities between our study (red stars) and previous literature (box plots) in terms of surface PM<sub>2.5</sub> and O<sub>3</sub> concentrations in eastern China. Note that red stars in the fifth column of each subgraph represent the statistical metrics of WRF-CHIMERE in this study.

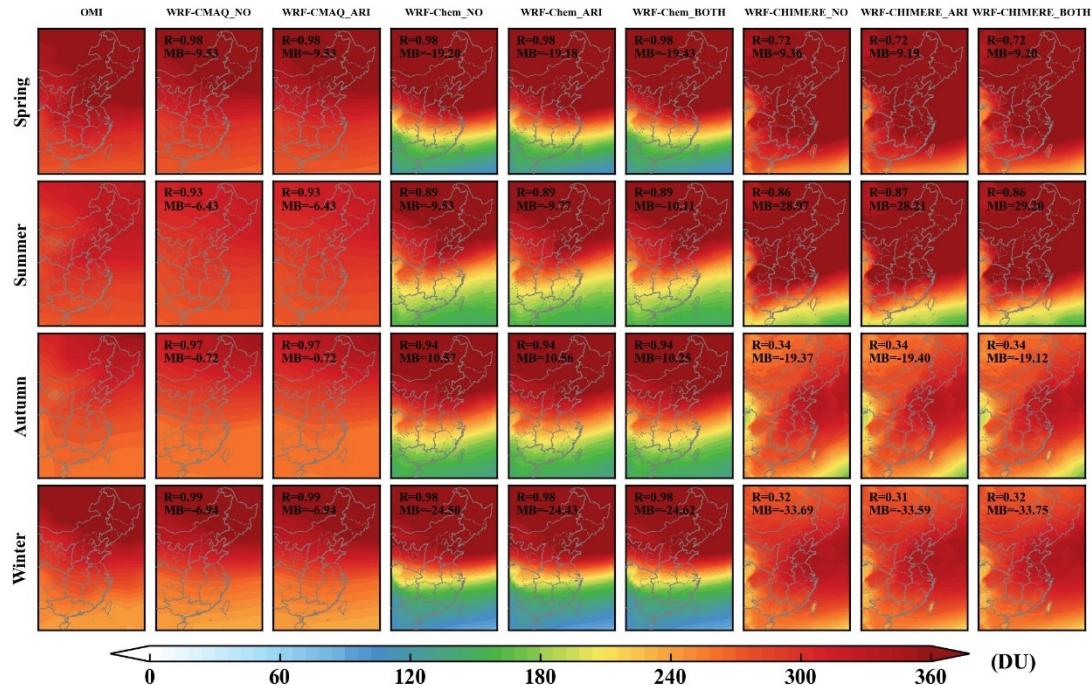


Figure S21. Spatial distributions of seasonal total column ozone between OMI observations and simulations from WRF-CMAQ, WRF-Chem and WRF-CHIMERE with and without aerosol feedbacks in eastern China.

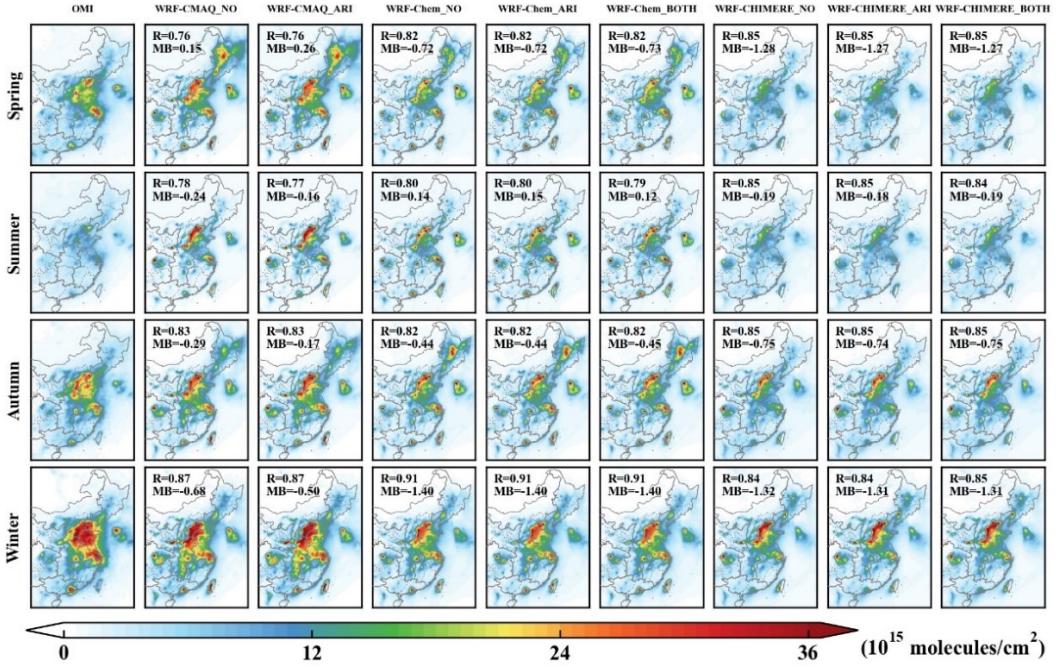


Figure S22. The same as Fig. S21 but for tropospheric NO<sub>2</sub> column.

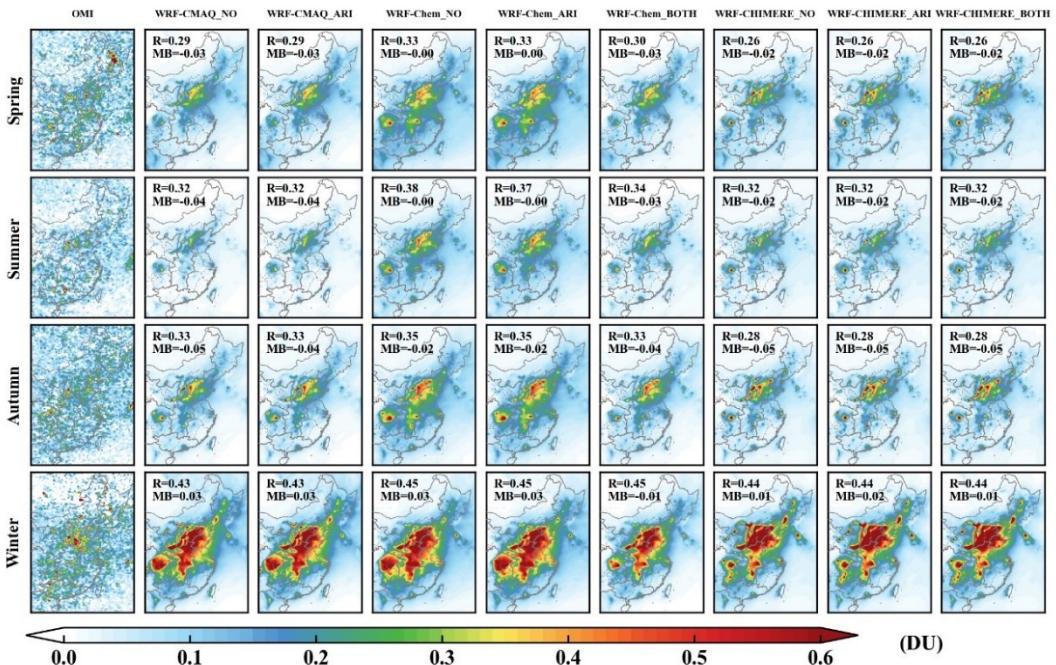


Figure S23. The same as Fig. S21 but for PBL SO<sub>2</sub> column.

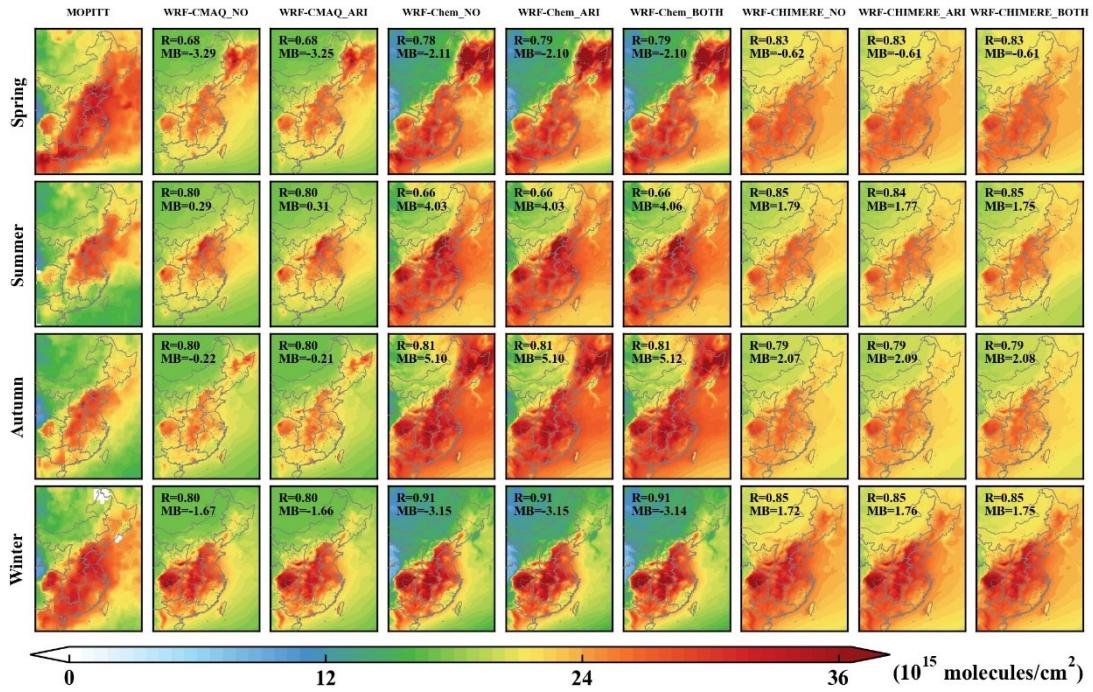


Figure S24. The same as Fig. S21 but for total CO column concentrations.

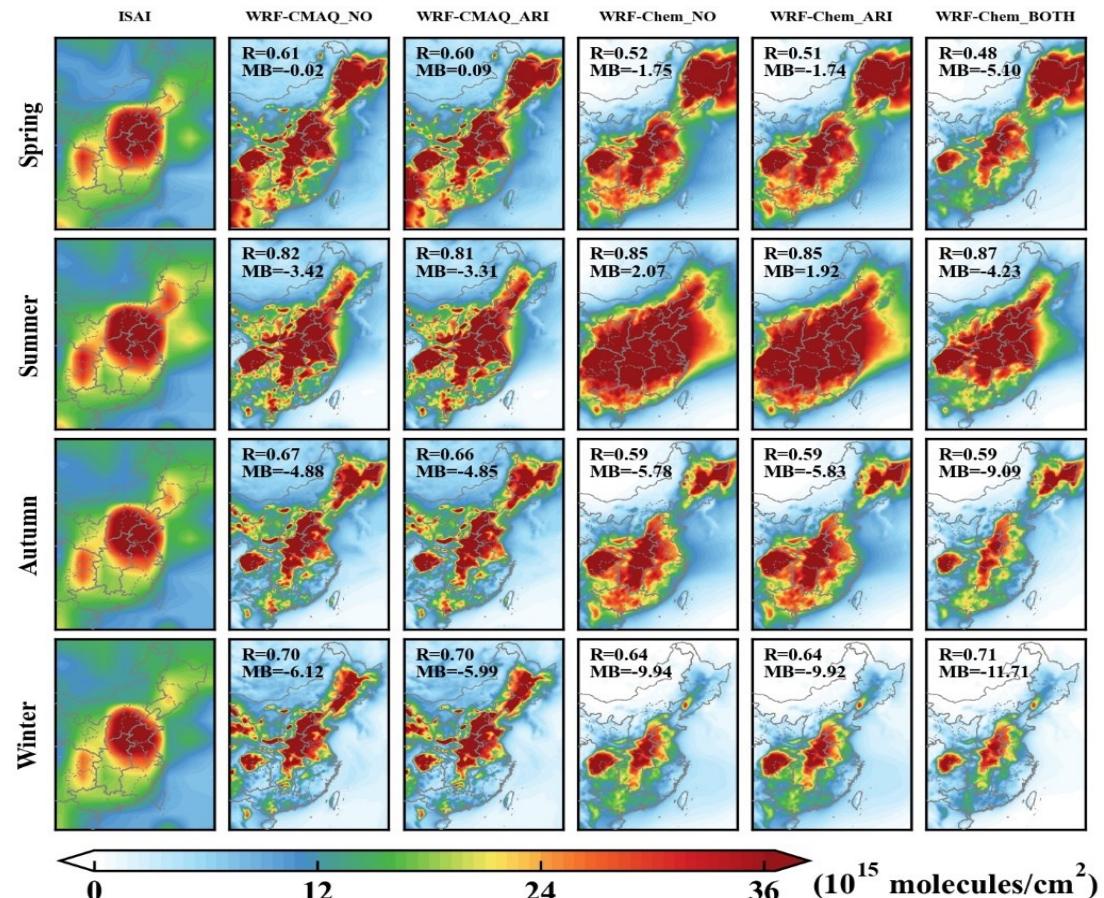


Figure S25. Spatial distributions of seasonal total NH<sub>3</sub> column between MOPITT observations and simulations from WRF-CMAQ and WRF-Chem with and without aerosol feedbacks in eastern China.

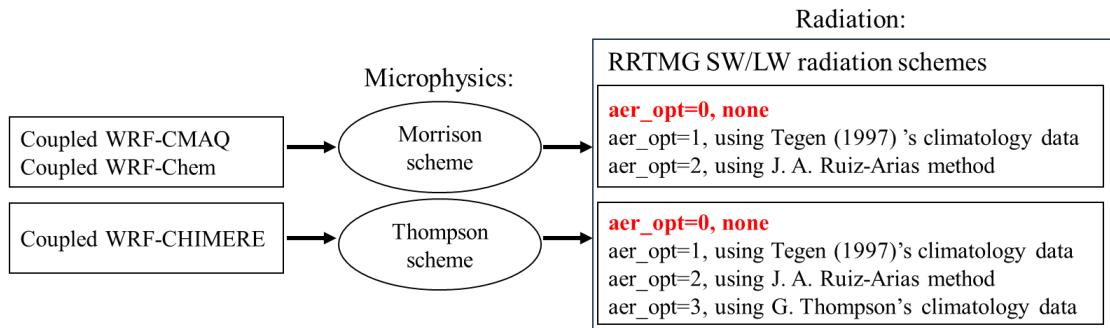


Figure S26. Summary of the selected options of radiation and microphysics schemes in coupled WRF-CMAQ, WRF-Chem and WRF-CHIMERE in this study.

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37 Guangyi Yang, Mengduo Zhang, Shengjin Xie: Simulation results from WRF-  
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