

1 **Air Quality and Climate Change, Topic 3 of the Model Inter-Comparison**
2 **Study for Asia Phase III (MICS-Asia III), Part II: aerosol radiative effects**
3 **and aerosol feedbacks**

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38 **Abstract**

39 Topic 3 of the Model Inter-Comparison Study for Asia (MICS-Asia) Phase III examines how
40 online coupled air quality models perform in simulating wintertime haze events in the North
41 China Plain region and evaluates the importance of aerosol radiative feedbacks. This paper
42 discusses the estimates of aerosol radiative forcing, aerosol feedbacks, and possible causes for
43 the differences among the participating models. Over the Beijing-Tianjin-Hebei (BTH) region,
44 the ensemble mean of estimated aerosol direct radiative forcing (ADRF) at the top of
45 atmosphere, inside the atmosphere and at the surface are -1.1, 7.7 and -8.8 W/m² during January
46 2010, respectively. Subdivisions of direct and indirect aerosol radiative forcing confirm the
47 dominant role of direct forcing. During severe haze days (January 17-19, 2010), the averaged
48 reduction in near surface temperature for the BTH region can reach 0.3-1.6 °C. The responses
49 of wind speeds at 10 m (WS10) inferred from different models show consistent declines in
50 eastern China. For the BTH region, aerosol-radiation feedback induced daytime changes in
51 PM_{2.5} concentrations during severe haze days range from 6.0 to 12.9 μg/m³ (< 6%). Sensitivity
52 simulations indicate the important effect of aerosol mixing states on the estimates of ADRF
53 and aerosol feedbacks. Besides, BC exhibits large contribution to atmospheric heating and
54 feedbacks although it accounts for a small share of mass concentration of PM_{2.5}.

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56 **1 Introduction**

57 Aerosols change weather and climate via the following pathways: they absorb and scatter solar
58 and thermal radiation to alter the radiative balance of the earth-atmosphere system (*Gao et al.,*
59 *2019b; Liu et al., 2011; Jia et al., 2018*), which is referred to as direct effects; and, they serve
60 as cloud condensation nuclei (CCN) and/or ice nuclei (IN) to modify cloud properties, which
61 is referred to as indirect effects (*Haywood and Boucher, 2000*). The suppression of cloud
62 convection induced by direct effects of absorbing aerosols is known as the semi-direct effect
63 (*Huang et al., 2006; Lohmann and Feichter, 2005*). Increases in cloud droplet number can
64 increase cloud albedo for a constant liquid water path (LWP), which is further classified as the
65 first indirect effect or Twomey effect (*Twomey, 1991*). More but smaller cloud droplets reduce
66 precipitation intensity but increase cloud lifetime, which is known as the cloud lifetime or

67 second indirect aerosol effect (*Albrecht, 1989*). In turn, changes in the radiative balance can
68 alter meteorological variables (e.g. temperature, relative humidity, photolysis rate, etc.) and
69 further the transport, diffusion and chemical conversion of trace gases and aerosols, while
70 changes in clouds can affect in-cloud aqueous-phase chemistry and wet deposition of gases and
71 aerosols.

72 The impacts of meteorology on chemistry have been explicitly treated in chemical transport
73 models (CTMs). For example, temperature modulates chemical reaction and photolysis rates,
74 affects volatility of chemical species, and biogenic emissions, wind speed and direction
75 determine transport and mixing, and precipitation influences wet deposition (*Baklanov et al.,*
76 *2014*). However, due to the complexity of these processes and lack of computational resources,
77 the influences of atmospheric compositions on weather and climate have been generally
78 ignored in previous CTMs (*Baklanov et al., 2014*). Studies examining how aerosols interact
79 with weather/climate remain uncertain and limited. Recently, with the rapid development of
80 coupled meteorology and chemistry models, many new studies have been conducted to
81 investigate the aerosol direct and indirect effects and feedbacks (*Baklanov et al., 2017; Forkel*
82 *et al., 2015; Gao et al., 2016, 2017; Grell et al., 2005; Han et al., 2010; Huang et al., 2016;*
83 *Jacobson et al., 2007; Saide et al., 2012; Wang et al., 2014; Yang et al., 2011; Zhang et al.,*
84 *2010*). In highly polluted regions like Asia, aerosol feedbacks can be particularly important
85 (*Gao et al., 2016, 2017*). High concentrations of aerosols would enhance the stability of
86 boundary layer due to reductions in radiation that reach the surface, which in turn can cause
87 further increases in PM_{2.5} concentrations (*Ding et al., 2016; Gao et al., 2016*).

88 Aerosol feedbacks during haze events in China have been explored using multiple online
89 coupled meteorology-chemistry models, including WRF-Chem (the Weather Research
90 Forecasting model coupled with Chemistry, *Chen et al., 2013, 2018; Gao et al., 2016, 2017,*
91 *2019a; Liu et al., 2015*), WRF-CMAQ (Community Multiscale Air Quality, *Wang et al., 2014*).
92 Nevertheless, large uncertainties remain in the modelling of these processes, due to the lack of
93 direct observational constraints and challenges in predicting properties of aerosols. Thus, the
94 inter-comparison of coupled meteorology-chemistry models is of great significance to better
95 understand the differences, causes, and uncertainties within these processes.

96 Topic 3: air quality and climate change within the Model Inter-Comparison Study for Asia

97 Phase III (MICS-Asia phase III) was initialized to address these issues (*Gao et al., 2018a*).
98 Results from seven applications of fully online coupled meteorology-chemistry models using
99 harmonized emission and chemical boundary conditions were submitted to this topic (*Gao et*
100 *al., 2018a*). These model applications include two applications of WRF-Chem by different
101 institutions, two applications of the National Aeronautics and Space Administration (NASA)
102 Unified WRF (NU-WRF) model with different model resolutions, one application of the
103 Regional Integrated Environment Modeling System with Chemistry (RIEMS-Chem, *Han et al.,*
104 *2010*), one application of the coupled Regional Climate Chemistry Modeling System
105 (RegCCMS), and one application of the coupled WRF-CMAQ model (*Gao et al., 2018a*). More
106 detailed information of the participating models, and information about how the experiments
107 were designed and how models perform have been archived in *Gao et al. (2018a)*.

108 In this paper, we analyze the results from the participating models to address the following
109 questions: (1) how large is the aerosol radiative forcing during winter haze episodes in China
110 and how differently are models estimating it? (2) how do aerosol feedbacks change
111 meteorological variables? and how do current models differ in estimating these changes? (3)
112 how do aerosol feedbacks contribute to the evolution of high aerosol concentrations during
113 winter haze episodes? and what are the best estimates from different models? And (4) what are
114 the major causes of the differences among the models? Sect. 2 describes briefly how the
115 experiments were designed and how models perform. Sect. 3 presents the estimates of aerosol
116 direct radiative forcing inferred from multiple models, including the separation of direct and
117 indirect effects. In Sect. 4, we discuss the impacts of aerosol-radiation feedbacks on
118 meteorological variables and PM_{2.5} concentrations. Sect. 5 illustrates the sensitivity of aerosol
119 forcing and feedbacks to different processes in the model, and the summary is presented in Sect.
120 6.

121

122 **2 Overview of MICS-Asia III Topic 3**

123 The participants were requested to use common emissions to simulate air quality during
124 January 2010 and submit requested model variables. The participating models include one
125 application of the Weather Research Forecasting model coupled with Chemistry (WRF-Chem;

126 Fast et al., 2006; *Grell et al., 2005*) by Pusan National University (PNU) (M1); one application
127 of the WRF-Chem model by the University of Iowa (UIOWA) (M2); two applications (two
128 domains: 45 and 15 km horizontal resolutions) of the National Aeronautics and Space
129 Administration (NASA) Unified WRF (NU-WRF; *Peters-Lidard et al., 2015*) model by the
130 Universities Space Research Association (USRA) and NASA's Goddard Space Flight Center
131 (M3 and M4); one application of the Regional Integrated Environment Modeling System with
132 Chemistry (RIEMS-Chem; *Han et al., 2010*) by the Institute of Atmospheric Physics (IAP),
133 Chinese Academy of Sciences (M5); one application of the coupled Regional Climate
134 Chemistry Modeling System (RegCCMS; *Wang et al., 2010*) from Nanjing University (M6);
135 and one application of the coupled WRF-CMAQ (Community Multiscale Air Quality) model
136 by the University of Tennessee at Knoxville (UTK) (M7) (Table 1). A new Asian emission
137 inventory was developed for MICS-Asia III by integrating state-of-the-art national or regional
138 inventories (*Li et al., 2017*), which was provided to all modeling groups, along with biogenic
139 emissions, biomass burning emissions, etc. Simulations from two global chemical transport
140 models (e.g., GEOS-Chem (The Goddard Earth Observing System Model-Chemistry) and
141 MOZART (Model for OZone And Related chemical Tracers)) were provided as boundary
142 conditions for MICS-Asia III. The entire month of January 2010 was simulated and covered
143 by one single simulation for each participating model. Comprehensive model evaluations
144 indicate that all models could capture the observed near-surface temperature and water vapor
145 mixing ratio, but overestimated near-surface wind speeds. These models were able to represent
146 the observed daily maximum downward shortwave radiation, particularly low values during
147 haze days. The observed variations of air pollutants, including SO₂, NO_x, CO, O₃, PM_{2.5}, and
148 PM₁₀, were reproduced by these models. However, large differences in the models were found
149 in the predicted PM_{2.5} chemical compositions.

150

151 **3 Aerosol Direct and Indirect Forcing**

152 **Fig. 1** shows the monthly mean all-sky aerosol direct radiative forcing (ADRF) over China.
153 The spatial distributions of ADRF at the surface and inside the atmosphere inferred from
154 multiple models are generally consistent, with the largest values in eastern and southwestern

155 China. Over the Beijing-Tianjin-Hebei (BTH) region (areas marked in Figure S1), M7 reports
156 the highest ADRF at the surface (-17.0 W/m^2), and the largest ADRF inside the atmosphere
157 (14.6 W/m^2) (**Table 2**). M6 shows the lowest ADRF both at the surface and inside the
158 atmosphere (-3.6 and 3.6 W/m^2) (**Table 2**). It is noticed that M6 predicts lower aerosol optical
159 depth (AOD) than M7 (*Gao et al., 2018a*), which could partly explain the weaker ADRF
160 estimated by M6. M6 uses an external assumption of aerosol mixing states, which is likely to
161 cause weaker absorption and ADRF in the atmosphere (*Conant et al., 2003*). However, the
162 reported ADRF at the top of the atmosphere (TOA) vary widely, and no consensus is reached
163 on whether the forcing is positive or negative. The spatial pattern of ADRF at the TOA inferred
164 from M5 are consistently negative across the modeling domain, while the results inferred from
165 other models are patchy with positive values to the north or to the southwest (**Fig. 1**). Consistent
166 negative ADRF at the TOA estimated by M5 is related to the strong negative forcing at the
167 surface and the predicted high concentrations of sulfate by M5 (*Gao et al., 2018a*). Over the
168 BTH region, simulated ADRF at the TOA range from -2.6 to 0.2 W/m^2 (**Table 2**). *Li et al.*
169 (*2010*) reported observation-based estimates of aerosol radiative forcing across China to be
170 0.3 ± 1.6 at the TOA. *Chung et al. (2005)* and *Chung et al. (2010)* estimated the forcing over
171 south Asia to be -2.9 W/m^2 and -3.6 W/m^2 at the TOA, respectively. The magnitudes of the
172 model estimated aerosol radiative forcing values are generally in line with these estimates
173 inferred from observations, while discrepancies among models could be due to assumptions of
174 aerosol mixing states and other model treatments (parameterization of hygroscopicity, soil dust,
175 etc.). The discussions on how different model treatments affect the results of ADRF is provided
176 in Sect. 5.

177 **Fig. 2** exhibits the ensemble mean of monthly averaged ADRF at the TOA, inside the
178 atmosphere and at the surface. Elevated forcing inside the atmosphere and at the surface are
179 mainly located in east China. However, the ensemble mean of forcing at the TOA over the
180 ocean is slightly higher than that over the land. Over the BTH region, the ensemble mean of
181 ADRF at the TOA, inside the atmosphere and at the surface are -1.1 , 7.7 and -8.8 W/m^2 ,
182 respectively. In winter, the aerosol radiative forcing in China is largely contributed by the
183 power sector and residential sector, but with different signs of the contribution (*Gao et al.,*
184 *2018b*).

185 M4 and M5 further provide subdivision of direct and indirect aerosol radiative forcing. As
186 listed in **Table 3**, although the magnitudes of forcing estimated by M4 and M5 differ from each
187 other, the dominant roles of direct forcing are consistent. Over North China and during
188 wintertime, aerosol indirect forcing is negligible due to the lack of water vapor and the stable
189 weather conditions.

190

191 **4 Impact of aerosol feedbacks on meteorological variables and PM_{2.5}** 192 **concentrations**

193 When extreme haze events happen, high aerosol loadings can reduce significantly the
194 shortwave radiation reaching the surface, modifying near-surface temperature (*Gao et al.,*
195 *2017*). **Fig. 3** displays the aerosol-radiation feedback induced changes in temperature at 2 m
196 (T2) from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (Table 1: M1: WRF-Chem, Pusan
197 National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF, NASA; M5:
198 RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7:
199 WRF-CMAQ, University of Tennessee; *Gao et al., 2018a*). The participating models show
200 different degrees of reductions in T2. M5 exhibits the most widespread areas with reductions,
201 which include Northeastern China. However, significant reductions in T2 inferred from other
202 models are mainly concentrated in southern and eastern China (**Fig. 3**). In Beijing (areas
203 marked in Figure S1), the monthly averaged reductions in T2 from multiple models range from
204 0 to 0.7 °C, with the greatest changes calculated from M4 (**Table 2**). In the Beijing-Tianjin-
205 Hebei (BTH) region, similar magnitudes (0-0.8 °C) are found. When only severe haze days
206 (January 17-19) are considered, the averaged reductions in T2 for Beijing (0.1-1.7 °C) and the
207 BTH region (0.3-1.6 °C) are further enhanced (**Table 4**). In terms of aerosol-radiation feedback
208 induced temperature reduction, M1 and M2 generally report similar magnitudes, which are
209 lower than M4, M5 and M7. Model evaluations of PM_{2.5} composition in *Gao et al. (2018a)*
210 reveal that M4 overpredicts the concentrations of organic carbon, which could be one of the
211 reasons for the higher estimated reductions in T2 due to aerosols.

212 Pronounced decreases in water vapor at 2 m (Q2) are mostly located in southern China (**Fig.**
213 **4**), where water vapor is more abundant due to the proximity to the sea. During extreme haze

214 days, the aerosol-radiation feedback induced decreases in Q2 in the BTH region from multiple
215 models range from 0.07 to 0.29 g/kg, with the lowest estimate from M1 and the highest from
216 M4 (**Table 4**).

217 The responses of wind speeds at 10 m (WS10) inferred from different models are generally
218 consistent, displaying decreases in eastern China except M6 (**Fig. 5**). In the BTH region, the
219 monthly mean aerosol-radiation feedback induced decreases in WS10 range from 0.02 to 0.09
220 m/s (**Table 2**), and more pronounced reductions are suggested by M4 and M7 (**Fig. 5**).

221 Because of aerosol-radiation feedback, most models report that surface PM_{2.5} concentrations
222 are enhanced in China, with the exception of M6 (**Fig. 6**). It is also noteworthy that PM_{2.5}
223 concentrations decrease in the Gobi desert and Taklimakan desert of western China in M5 and
224 M2, which is caused by the decreased wind speed near the surface due to the weakened
225 downward transport of momentum from upper layer above boundary layer to the surface (*Han*
226 *et al.*, 2013). The changes of PM_{2.5} concentrations suggested by M6 are patchy over east China,
227 with decreases to the north and to the southwest. The monthly mean PM_{2.5} are enhanced by 0.1-
228 1.4 $\mu\text{g}/\text{m}^3$ for Beijing, and by 0.8-2.2 $\mu\text{g}/\text{m}^3$ for the BTH region. The enhancement fractions
229 are generally below 2% for Beijing, and below 4% for the BTH region (**Table 2**).

230 To further understand how aerosol-radiation feedback contributes to the formation of haze
231 event, we calculate the mean increase during extreme haze days (January 17-19). For the BTH
232 region, the contribution of aerosol-radiation feedback to PM_{2.5} concentrations are lower than
233 4%, and the enhancement are below 8.5 $\mu\text{g}/\text{m}^3$. *Gao et al.* (2017) demonstrates that the aerosol-
234 radiation feedback induced changes in PM_{2.5} are negligible during nighttime, so we further
235 calculate daytime mean changes, as listed in **Table 4**. For the BTH region, M2 reports the
236 largest enhancement (12.9 $\mu\text{g}/\text{m}^3$) of PM_{2.5} concentrations during daytime. Other models,
237 except M6, report similar magnitudes of enhancement, ranging from 5.3 to 6.6 $\mu\text{g}/\text{m}^3$. The
238 enhancement fraction remains less than 6% for the BTH region, and below 8.3% for Beijing.
239 **Table 4** also displays the maximum enhancement of PM_{2.5} during haze days over the BTH
240 region. M7 suggests the largest PM_{2.5} enhancement (up to 60.9 $\mu\text{g}/\text{m}^3$), followed by M2 (up to
241 55.4 $\mu\text{g}/\text{m}^3$). Other three models, M1, M4, M5, and M6 indicate the aerosol-radiation induced
242 increase in PM_{2.5} can reach up to more than 20 $\mu\text{g}/\text{m}^3$ in the BTH region (**Table 4**).

243 The contributions of aerosol-radiation feedback to haze formation in China have been

244 investigated in many previous studies (*Ding et al., 2016; Gao et al., 2015; Gao et al., 2016;*
245 *Liu et al., 2018; Wang et al., 2014a; Wang et al., 2014b; Wang et al., 2015; Wu et al., 2019;*
246 *Zhang et al., 2015; Zhang et al., 2018; Zhong et al., 2018*), but the reported values diverge.
247 *Ding et al. (2016), Wang et al. (2014a)* and *Zhong et al. (2018)* indicate that the aerosol
248 radiative effects can increase PM_{2.5} by more than 100 µg/m³ (maximum hourly changes) or
249 +70%. *Gao et al. (2015), Wang et al. (2014b), Wang et al. (2015), and Zhang et al. (2018)*
250 suggest that the contributions are generally within the range of 10-30%. These reports are
251 different from this study in terms of study periods, region, and pollution levels. Most of
252 previous reports focused on the January 2013 haze episodes (*Wang et al., 2014a*), while the
253 monthly mean concentrations of PM_{2.5} in January 2010 are nearly 50% lower than that of
254 January 2013. According to the findings in this study, the contribution of aerosol-radiation
255 feedback to haze formation during January 2010 are generally below 10%. Uncertainties still
256 remain as suggested by the errors in the simulated chemical compositions (*Gao et al., 2018a*).
257 Concentrations of sulfate and organic aerosol are generally underestimated by most of the
258 participating models, and M4 overestimates the concentrations of organic aerosols (*Gao et al.,*
259 *2018a*). These model errors were attributed to the incomplete multiphase oxidation
260 mechanisms of sulfate, and different treatments of secondary organic aerosol (SOA) formation
261 in these models (*Gao et al., 2018a*).

262

263 **5 Sensitivity to Different Processes**

264 To explore the potential causes for the differences among models, and the major factors that
265 influence aerosol-radiation feedback, several sensitivity simulations were conducted with the
266 RIEMS-Chem model (M5) (*Han et al., 2010*). These simulations aim to examine the effects of
267 mixing states of aerosols, hygroscopic growth, black carbon and soil dust.

268 5.1 Aerosol mixing states

269 In the control simulation, inorganic aerosols and BC are assumed to be internally mixed to form
270 a homogeneous mixture. The refractive index of this mixture is estimated using the volume-
271 weighted average of the refractive index of individual component. The size of the mixture is
272 prescribed to be the maximum size of the mixed aerosol components. For example, the size of

273 the mixture of sulfate and BC is set to be equal to the size of sulfate, assuming a small BC
274 particle sticking to a larger sulfate particle.

275 An additional simulation was conducted with the aerosols were treated as externally mixed,
276 and the corresponding results are displayed in **Fig. 7-9**. For external mixing assumption, each
277 aerosol component is considered individually, and the total AOD is calculated as the sum of
278 extinction by each aerosol component. Compared with the results with internal mixing
279 assumption, results with external mixing assumption generally exhibit a weaker (negative)
280 ADRF at the surface (~15%), a stronger (negative) ADRF at TOA (~50%) and a decreased
281 (positive) ADRF in the atmosphere (~30%) (**Fig. 9a, 9f, 9k**). These responses of ADRF to the
282 assumption of aerosol mixing states are consistent with *Conant et al. (2003)*. However, *Curci*
283 *et al. (2015)* reported lower AOD with internal mixing assumption than with external mixing
284 assumption. In *Curci et al. (2015)*, aerosol mass was distributed in less numerous particles with
285 larger sizes. As a result, fewer scattering agents and lower AOD were estimated.

286 Aerosol feedbacks estimated by M5 also tend to be weaker with external mixing assumption
287 than with internal mixing assumption (changes in surface meteorological variables and PM_{2.5}
288 concentrations, **Fig. 8a, 8d, 8g, and 8j**). The monthly averaged changes in T2, WS10 and PM_{2.5}
289 are -0.6 °C, -0.04 m/s and 2.2 µg/m³ for the BTH region with internal mixing assumption, while
290 the corresponding values change to -0.6 °C, -0.03 m/s and 1.8 µg/m³ with external mixing
291 assumption. These differences emphasize the important influences of aerosol mixing states on
292 the estimates of ADRF and aerosol feedbacks. However, aerosol mixing states are also varying
293 with time and location. Measurements in North China suggest that aerosols are partially
294 internally mixed, and the fraction of internal mixing increased from clean to haze periods (*Li*
295 *et al., 2014*).

296

297 5.2 Hygroscopic growth

298 Given the appreciable effect of aerosol hygroscopic growth on ADRF (*Li et al., 2014*), another
299 simulation was conducted with decreased relative humidity (RH). In this simulation, FNL
300 nudging was applied above boundary layer to reduce RH This perturbation of RH was based
301 on the fact that M5 overestimates relative humidity (water vapor mixing ratio) (*Gao et al.,*
302 *2017*). With FNL nudging, RH was reduced by 5-10% in the BTH and by ~25% in the middle

303 and lower reaches of the Yangtze River, leading to lower values of AOD (**Fig. 7f**) and weaker
304 ADRF at the surface and TOA (**Fig. 9e, 9j, and 9o**, about 10% lower in the BTH region).

305

306 5.3 Soil dust and sea salt

307 M5 (RIEMS-Chem) includes naturally emitted soil dust and sea salt, while the other models
308 except M2 (WRF-Chem, University of Iowa) do not consider soil dust in their model settings.

309 In an additional sensitivity simulation, soil dust and sea salt emissions were turned off in M5
310 to examine the influence on ADRF and aerosol feedbacks (**Fig. 9d, 9l and 9n**). In January 2010,

311 significant amounts of soil dust were emitted from the Taklimakan desert, influencing wide
312 areas of China. M5 estimates that the monthly mean ADRF at the surface due to dust and sea

313 salt is about -12 W/m^2 over the Taklimakan desert, $-4\sim-6 \text{ W/m}^2$ in the middle reaches of the
314 Yellow River and the Yangtze River Delta, and about $-2\sim-4 \text{ W/m}^2$ over the BTH region. Over

315 the BTH region, the contribution of dust and sea salt aerosols to total ADRF can reach 5~10%.
316 Table 2 illustrates that M5 predicts the largest (negative) radiative forcing at TOA over the BTH

317 region. The above analyses with reduced relative humidity and perturbations in dust and sea
318 salt suggest that the inclusion of dust and overprediction of relative humidity by M5 are

319 important reasons.

320

321 5.4 The effect of BC

322 Two sets of simulations, namely without BC and with doubled BC concentrations, were
323 conducted to examine the influences of BC on aerosol radiative forcing and feedbacks. In the

324 control simulation, the aerosol induced changes in monthly T2, WS10 and PM_{2.5} are $-0.6 \text{ }^\circ\text{C}$, $-$
325 0.04 m/s and $2.2 \text{ } \mu\text{g/m}^3$ for the BTH region, respectively. When BC is not included (only

326 scattering aerosols and dust), the corresponding aerosol induced changes are $-0.5 \text{ }^\circ\text{C}$, -0.02 m/s
327 and $1.0 \text{ } \mu\text{g/m}^3$, respectively. When BC concentrations are doubled, these values change to -0.7

328 $^\circ\text{C}$, -0.05 m/s and $3.2 \text{ } \mu\text{g/m}^3$, respectively. The comparison between the control case and two
329 additional sensitivity cases indicates that the changes caused by BC are comparable to those by

330 scattering aerosols. The contribution of BC to aerosol feedbacks can reach up to 40~50%. It is
331 also found that the influence of BC on aerosol feedbacks with internal mixing assumption is

332 larger than that with external mixing assumption (Figure not shown).

333 Large uncertainties still remain in the estimates of the role of BC in aerosol feedbacks relative
334 to scattering aerosols. *Gao et al. (2016)* suggested that the impacts of BC on boundary layer
335 height and PM_{2.5} concentrations can account for as high as 60% of the total aerosol feedbacks
336 in the North China Plain at 2 p.m., although it only accounts for a small share of PM in terms
337 of mass concentration. *Qiu et al. (2017)* indicated that PM_{2.5} concentrations averaged over the
338 North China Plain increased by 16.8% and 1.0% due to scattering aerosols and BC, respectively.
339 It should be noted that most participating models, including RIEMS-Chem, tend to
340 underpredict the total mass concentrations of scattering aerosols (inorganic and organic
341 aerosols) by up to a factor of two over the study period, leading to overestimation of the
342 contribution of BC.

343

344 **6 Summary**

345 Topic 3 of MICS-Asia III (*Gao et al., 2018a*) focuses on understanding how current online
346 coupled air quality models perform in capturing extreme aerosol pollution event in North China
347 and how aerosols interact with radiation and weather. Seven applications of different online
348 coupled meteorology-chemistry models were involved in this activity. *Gao et al. (2018a)* has
349 demonstrated that main features of the accumulation of air pollutants are generally well
350 represented, while large differences in the models were found in the predicted PM_{2.5} chemical
351 compositions. These inconsistencies would lead to differences in estimated ADRF and aerosol
352 feedbacks.

353 The spatial distributions of ADRF at the surface and inside the atmosphere inferred from
354 multiple models are generally consistent, but the spatial distributions of ADRF at the TOA
355 estimated by these models greatly differ. Over the BTH region, the ensemble mean of ADRF
356 at the TOA, inside the atmosphere and at the surface are -1.1, 7.7 and -8.8 W/m², respectively.
357 Subdivisions of direct and indirect aerosol radiative forcing confirm the dominant roles of
358 direct forcing.

359 During severe haze days (January 17-19), the averaged reduction in T2 for the BTH region can
360 reach 0.3-1.6 °C. The responses of wind speeds at 10 m (WS10) inferred from different models
361 show consistent declines in eastern China. For the BTH region, aerosol-radiation feedback

362 induced changes in daytime PM_{2.5} range from 5.3 to 12.9 µg/m³ (< 6%). Our findings differ
363 from previous studies (*Ding et al., 2016; Gao et al., 2015; Gao et al., 2016; Liu et al., 2018;*
364 *Wang et al., 2014a; Wang et al., 2014b; Wang et al., 2015; Wu et al., 2019; Zhang et al., 2015;*
365 *Zhang et al., 2018; Zhong et al., 2018*) in terms of study period, region and pollution levels.
366 The monthly mean concentrations of PM_{2.5} in January 2010 (current study period) are about
367 50% lower than those in January 2013.

368 Sensitivity simulations were conducted with the RIEMS-Chem model (M5) to understand the
369 influences of aerosols mixing states, hygroscopic growth, black carbon and soil dust. The
370 results indicate the important effect of aerosol mixing states on the estimates of ADRF and
371 aerosol feedbacks. It was also found that BC exhibits large contribution to atmospheric heating
372 and feedbacks, but uncertainties remain in estimating its contribution given the fact that the
373 observed aerosol chemical components were not perfectly simulated. *Huang et al. (2015)*
374 separated the contributions of different aerosol components to aerosol direct radiative forcing,
375 highlighting the roles of BC and sulfate. Future studies are also needed to improve predicitions
376 of aerosol chemical components and to separate the effects of individual aerosol component on
377 aerosol feedbacks.

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379 **Author Contributions**

380 M.G., Z.H., and G.R.C. designed the study, and M.G. processed and analyzed the data. M.G.,
381 Z.H., and G.R.C. wrote the paper with inputs from all other authors.

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383 **Data availability**

384 The measurements and model simulations data can be accessed through contacting the
385 corresponding authors.

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387 **Competing interests**

388 The authors declare that they have no conflict of interests.

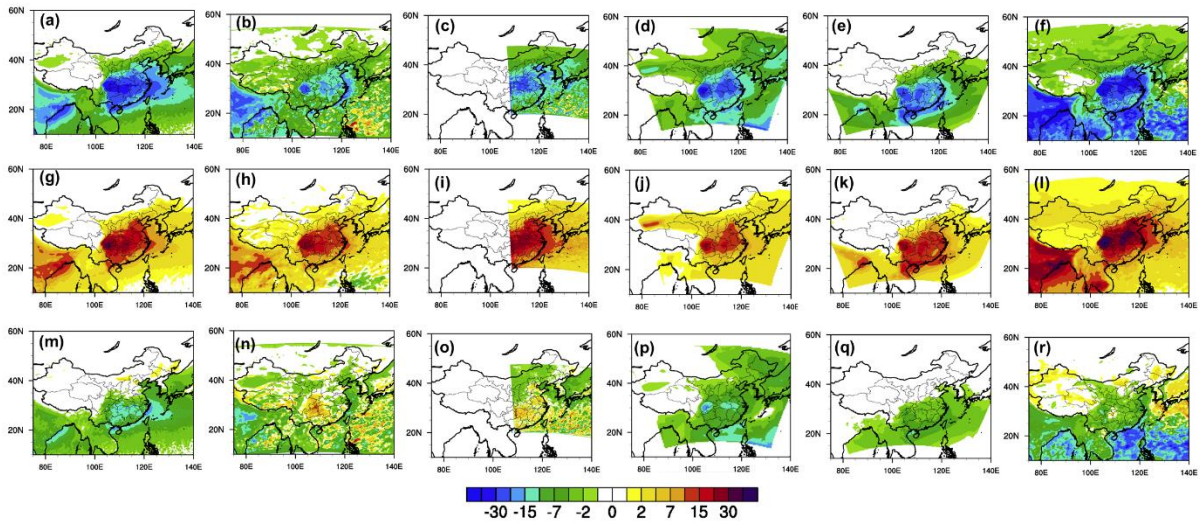
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390 **Acknowledgement**

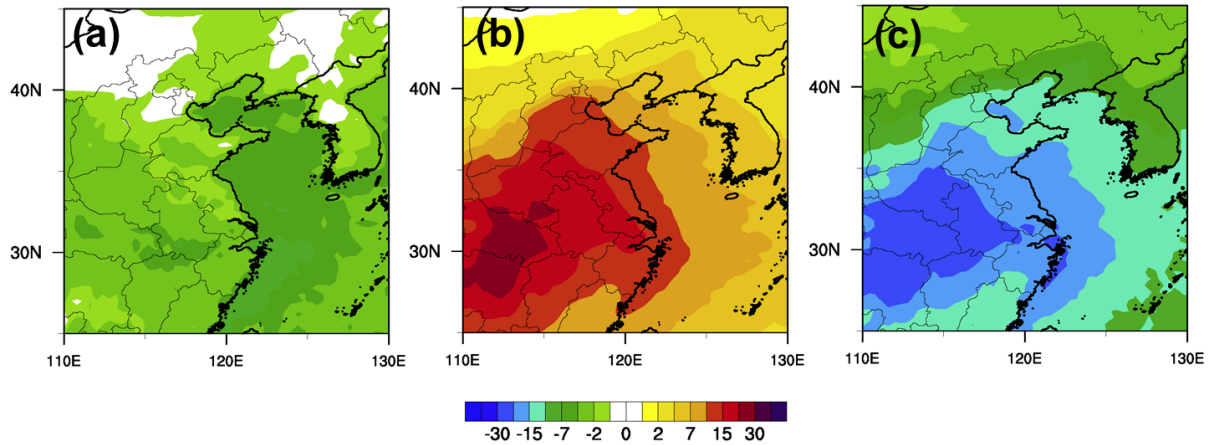
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 393 the special fund of State Key Joint Laboratory of Environment Simulation and Pollution
 394 Control (19K03ESPCT), Natural Science Foundation of Guangdong Province
 395 (2019A1515011633), and National Natural Science Foundation of China (NSFC91543202).

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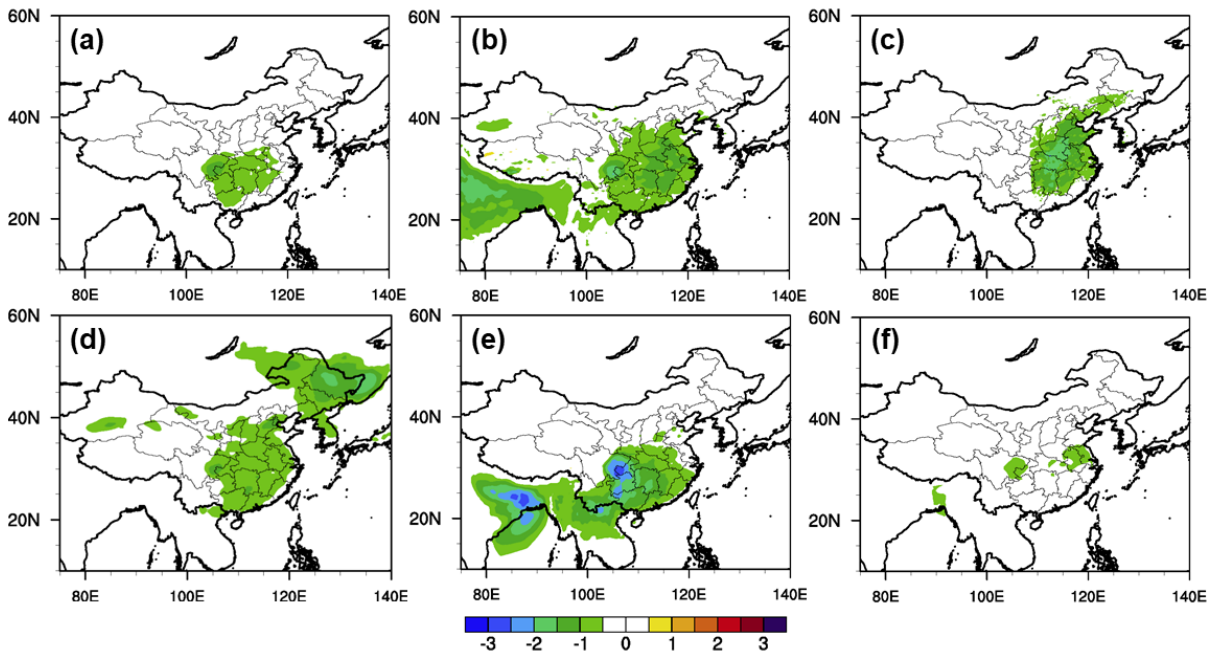


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 403 Figure 1. Monthly (January 2010) mean aerosol direct radiative forcing at the surface, inside the atmosphere and
 404 at the top of the atmosphere inferred from M1 (a, g, m), M2 (b, h, n), M4 (c, i, o), M5 (d, j, p), M6 (e, k, q), M7
 405 (f, l, r) (M1: WRF-Chem, Pusan National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF,
 406 NASA; M5: RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7: WRF-
 407 CMAQ, University of Tennessee; *Gao et al., 2018a*)
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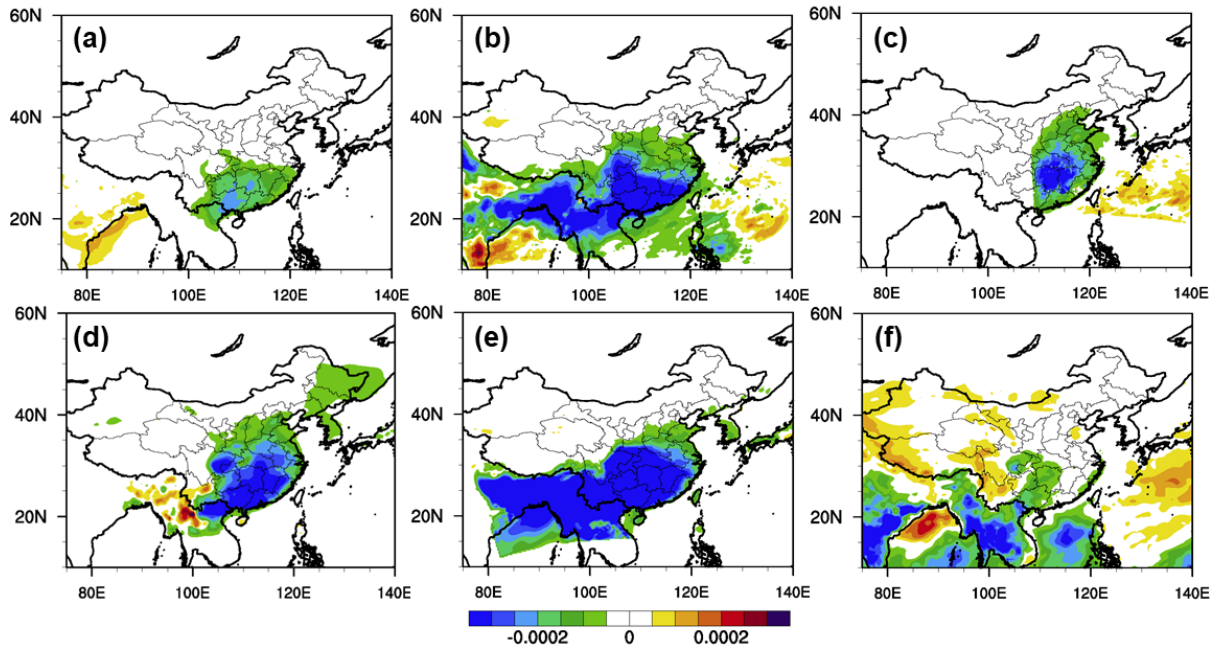
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Figure 2. Ensemble mean of monthly (January 2010) mean aerosol direct radiative forcing at the top of the atmosphere (a), inside the atmosphere (b) and at the surface (c)

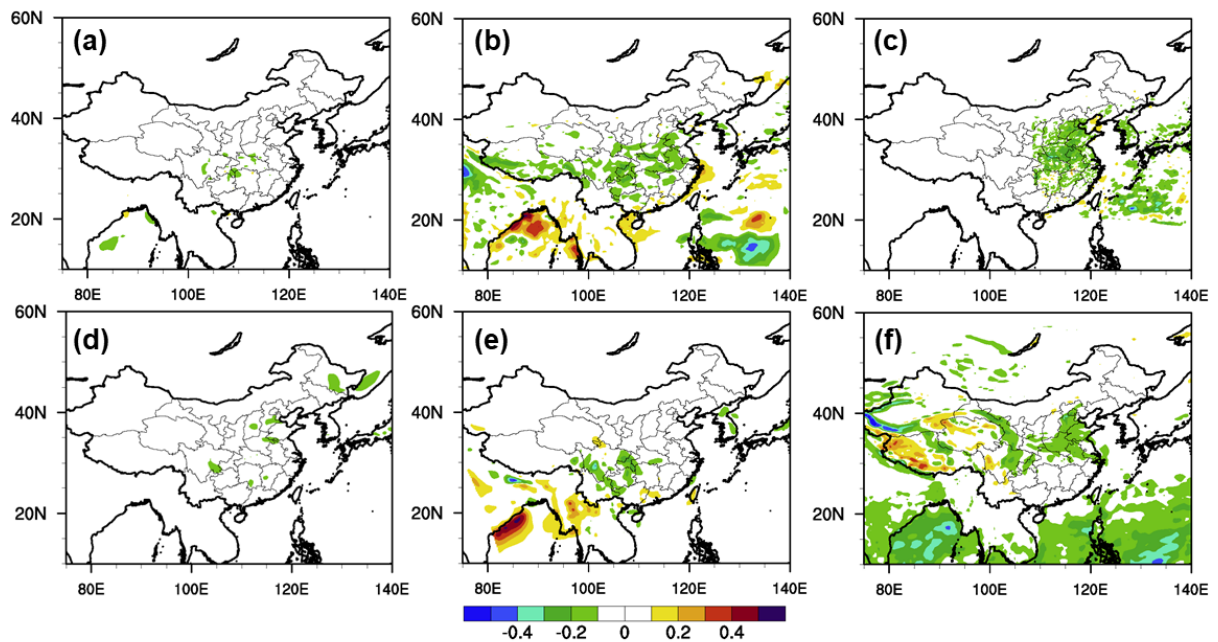


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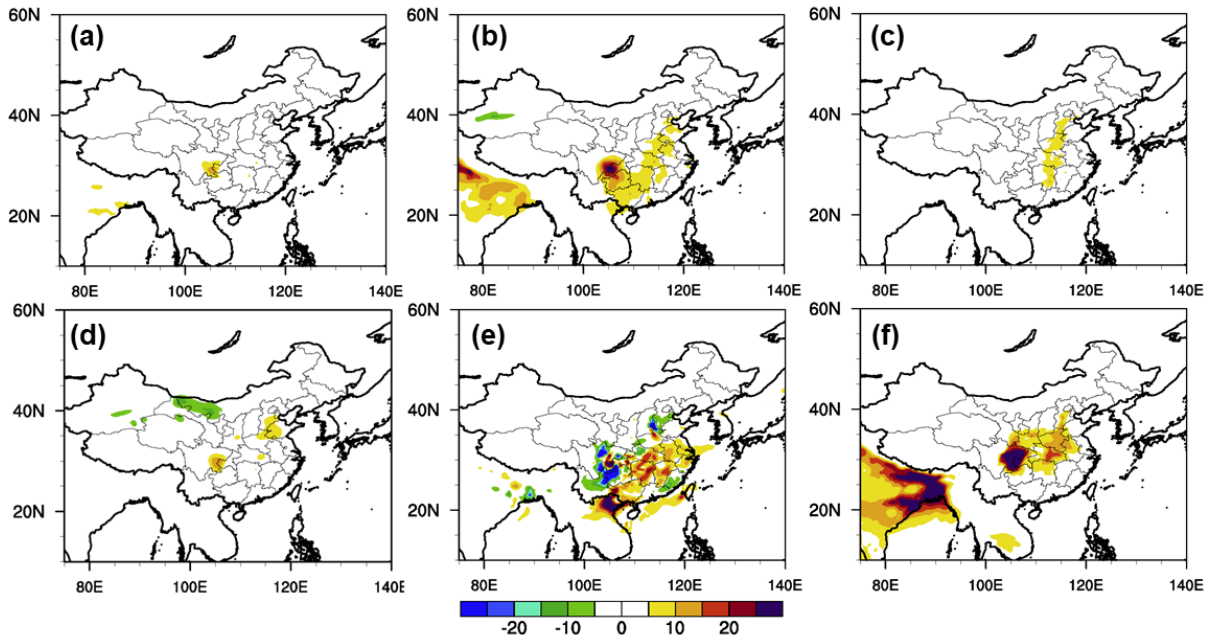
Figure 3. Monthly (January 2010) mean changes in temperature at 2 m (T_2 , °C) due to aerosol radiative effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et al., 2018a*)



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 423 Figure 4. Monthly (January 2010) mean changes in water vapor at 2 m (Q_2 , kg/kg) due to aerosol radiative
 424 effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of
 425 Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of
 426 Tennessee; *Gao et al., 2018a*)
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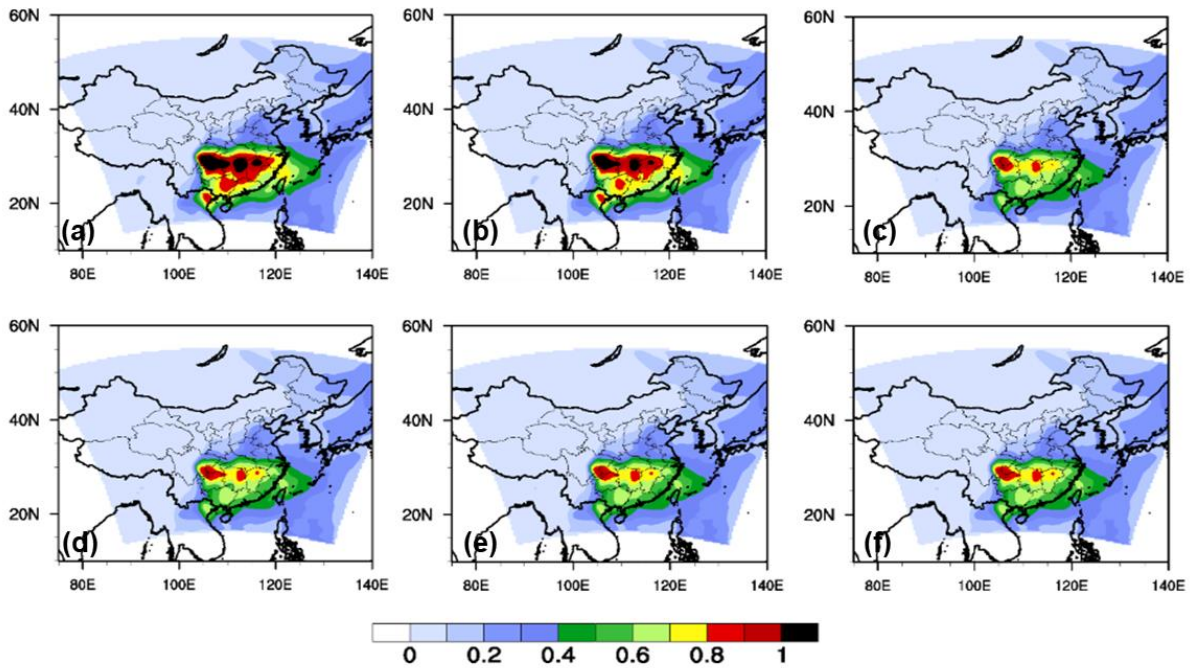


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 429 Figure 5. Monthly (January 2010) mean changes in wind speeds at 10 m (WS_{10} , m/s) due to aerosol radiative
 430 effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of
 431 Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of
 432 Tennessee; *Gao et al., 2018a*)
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 435 Figure 6. Monthly (January 2010) mean changes in surface $PM_{2.5}$ ($\mu\text{g}/\text{m}^3$) due to aerosol radiative effects from
 436 M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4:
 437 NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et*
 438 *al.*, 2018a)

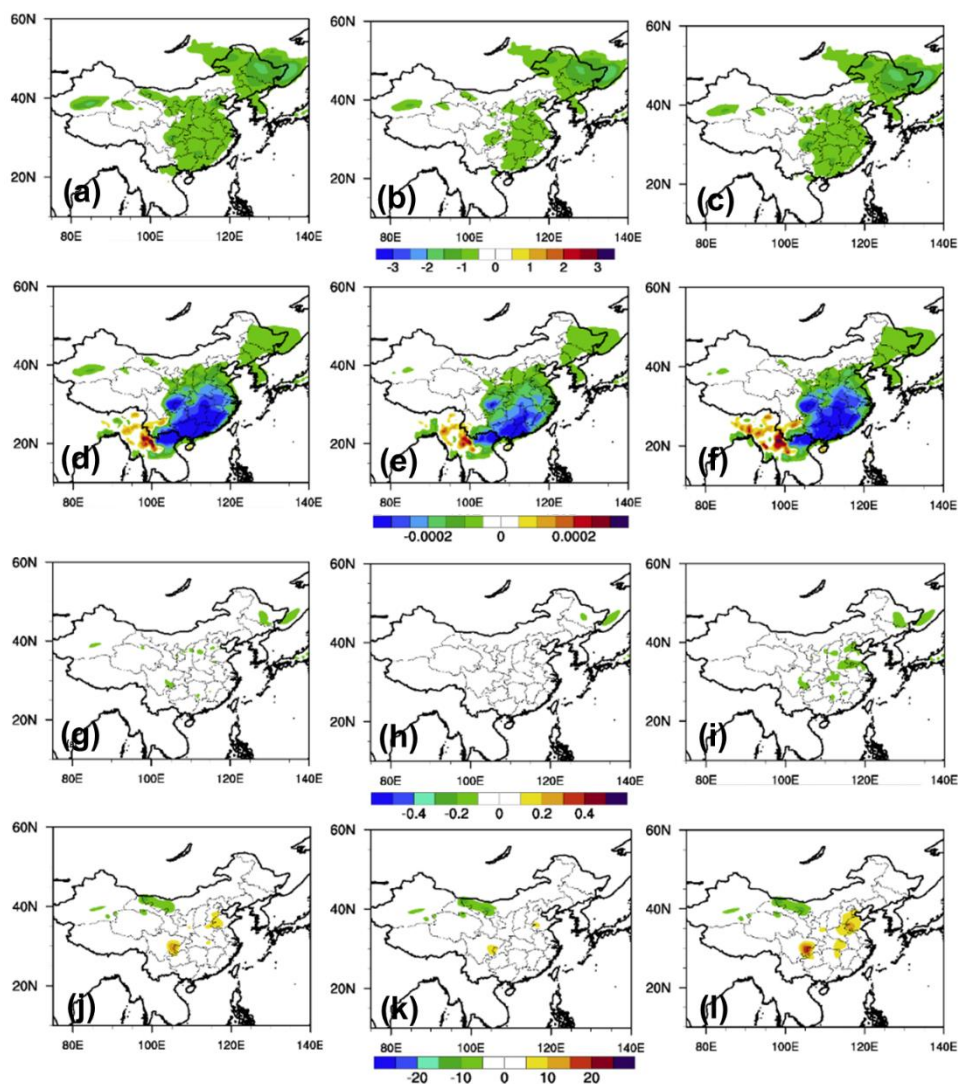
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 443 Figure 7. Monthly (January 2010) mean RIEMS-Chem modeled AOD from different simulations: control run
 444 (default simulation with internal mixing assumption) (a), external mixing assumption (b), internal mixing
 445 assumption but without BC (c), internal mixing assumption but with doubled BC (d), without dust and sea-salt

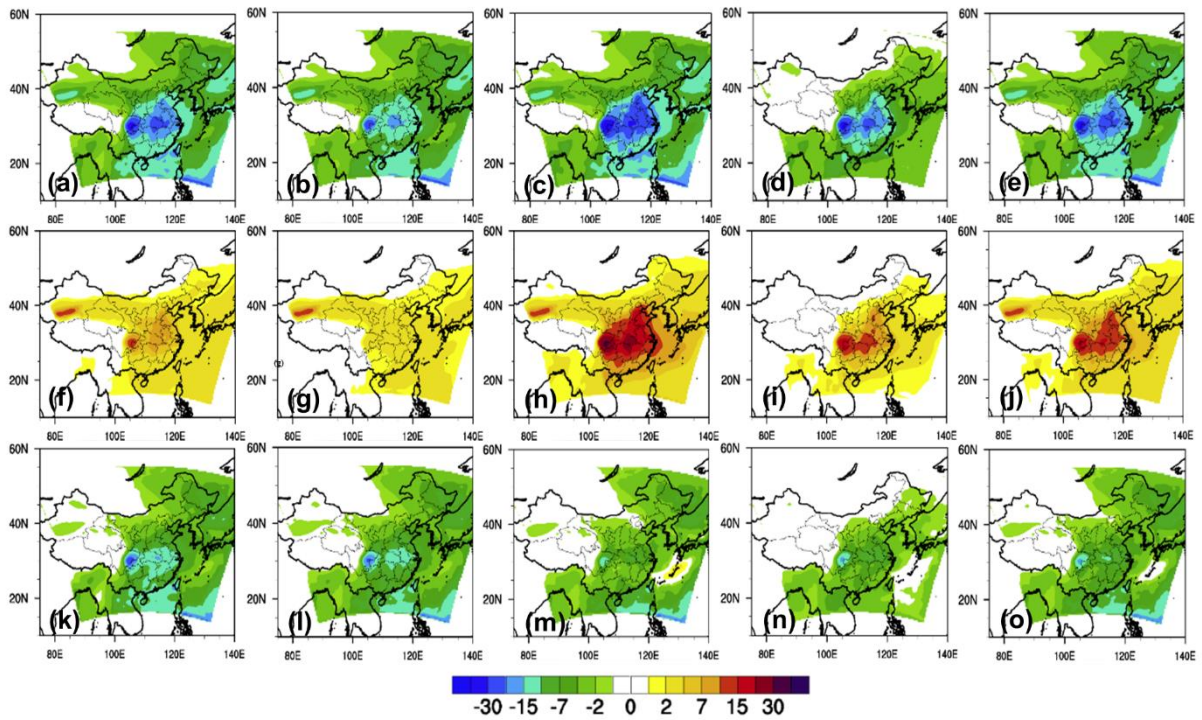
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(e), and reduced RH (f)



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Figure 8. Monthly (January 2010) mean RIEMS-Chem modeled changes in T2 (°C), Q2 (kg/kg), WS10 (m/s) and PM_{2.5} (µg/m³) from different simulations: external mixing assumption (first column), internal mixing assumption but without BC (second column) and internal mixing assumption but with doubled BC (third column)



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Figure 9. Monthly (January 2010) mean RIEMS-Chem modeled aerosol direct radiative forcing at the surface (a-e), inside the atmosphere (f-j) and at the top of the atmosphere (k-o) from different simulations: external mixing assumption (first column), internal mixing assumption but without BC (second column), internal mixing assumption but with doubled BC (third column), without dust and sea-salt (fourth column), and reduced RH (fifth column)

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Table 1 Participating models in Topic 3

Models	M1: WRF- Chem1	M2: WRF- Chem2	M3: NU- WRF1	M4: NU- WRF2	M5: RIEMS- Chem	M6: RegCCMS	M7: WRF- CMAQ
Modelling Group	Pusan National University	University of Iowa	USRA/NAS A	USRA/NASA	Institute of Atmospheric Physics	Nanjing University	University of Tennessee
Grid Resolution	45km	50km	45km	15km	60km	50km	45km
Vertical Layers	40 layers to 50mb	27 layers to 50mb	60 layers to 20mb	60 layers to 20mb	16 layers to 100mb	18 layers to 50mb	
Gas phase chemistry	RACM	CBMZ	RADM2	RADM2	CBM4	CBM4	SAPRC99
Aerosols	MADE	MOSAIC- 8bin	GOCART	GOCART	Sulfate, nitrate, ammonium, BC, OC, SOA, 5 bins of soil dust, and 5 bins of sea salt	Sulfate, nitrate, ammonium, BC and POC	AE06
Chemical Boundary Conditions	Climatologica l data from NALROM	MOZART	MOZART GOCART	MOZART GOCART	GEOS-Chem	Climatological data	GEOS- Chem

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505 Table 2 Monthly Mean (January 2010) Aerosol Direct Radiative Forcing (W/m^2) and
 506 Changes in T2 ($^{\circ}C$), Q2 (g/kg), WS10 (0.1 m/s), and PM_{2.5} ($\mu g/m^3$) for Beijing and Beijing-
 507 Tianjin-Hebei region (areas marked in Fig. S1)

Beijing	M1 PNU	M2 UIOWA	M4 NASA	M5 IAP	M6 NJU	M7 UTK
ADRF TOA	-0.6	-2.2	-0.8	-1.4	-0.1	-2.5
ADRF ATM	5.8	4.3	9.3	5.1	2.4	11.6
ADRF SFC	-6.4	-6.5	-10.1	-6.5	-2.5	-14.1
T2	-0.1	-0.3	-0.7	-0.5	-0.1	0.0
Q2	-1.2E-2	-2.3E-2	-6.4E-2	-5.8E-2	-5.8E-3	2.1E-2
WS10	-0.2	-0.2	-0.6	-0.2	0.0	-1.2
PM _{2.5}	0.1 (0.2%)	1.4 (1.6%)	1.1 (1.7%)	0.6 (1.4%)	-1.2 (- 2.2%)	1.0 (1.4%)
BTH						
ADRF TOA	0.2	-1.4	-0.3	-2.6	0.0	-2.4
ADRF ATM	7.3	5.4	10.1	6.3	3.6	14.6
ADRF SFC	-7.1	-6.8	-10.4	-8.9	-3.6	-17.0
T2	-0.2	-0.4	-0.8	-0.6	-0.2	0.0
Q2	-1.0E-2	-2.5E-2	-8.1E-2	-7.6E-2	-2.9E-2	2.5E-2
WS10	-0.2	-0.2	-0.9	-0.4	0.1	-0.9
PM _{2.5}	0.8 (1.4%)	1.8 (1.8%)	2.2 (3.2 %)	2.2 (3.9%)	-4.2 (- 5.7%)	2.2 (2.4%)

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524 Table 3 Monthly Mean (January 2010) Aerosol Direct Radiative Forcing and indirect
 525 Radiative Forcing (W/m^2) at the top of the atmosphere inferred from M4 and M5 (areas
 526 marked in Fig. S1)

Beijing	direct	Indirect
M4	-0.77	-0.15
M5	-1.43	-0.01
BTH		
M4	-0.28	0.1
M5	-2.63	-0.04

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562 Table 4 Mean Aerosol (January 2010) Direct Radiative Forcing (W/m^2) and Changes in T2
 563 ($^{\circ}C$), Q2 (g/kg), WS10 (0.1 m/s), and $PM_{2.5}$ ($\mu g/m^3$) for Beijing and Beijing-Tianjin-Hebei
 564 (BTH) region averaged over January 17-19 2010 (areas marked in Fig. S1)

Beijing	M1 PNU	M2 UIOWA	M4 NASA	M5 IAP	M6 NJU	M7 UTK
ADRF TOA	2.6	-1.4	1.8	-3.0	-0.6	-3.3
ADRF ATM	18.6	9.8	21.5	13.3	7.3	32.3
ADRF SFC	-16.0	-11.2	-19.7	-16.3	-7.9	-35.6
T2	-0.5	-0.5	-1.7	-1.3	-0.1	-1.5
Q2	-7.4E-2	-6.2E-2	-2.6E-1	-1.8E-1	-1.3E-2	-9.2E-2
WS10	-0.1	0.2	-2.3	0.4	0.5	-0.8
$PM_{2.5}$	-1.1 (- 0.9%)	3.8 (1.7%)	6.3 (3.8%)	1.0 (0.8%)	-7.9 (- 4.7%)	1.3 (1.1%)
BTH						
ADRF TOA	1.4	0.1	4.9	-4.6	-0.7	-3.8
ADRF ATM	18.3	12.0	19.1	13.2	10.0	36.1
ADRF SFC	-16.9	-11.9	-14.2	-17.8	-10.7	-39.9
T2	-0.6	-0.7	-1.6	-1.2	-0.3	-1.5
Q2	-7.1E-2	-8.2E-2	-2.9E-1	-2.0E-1	-1.2E-1	-8.9E-2
WS10	-0.3	-0.4	-2.5	0.0	0.3	-0.9
$PM_{2.5}$	2.9 (2.3%)	8.5 (3.7%)	5.3 (3.9%)	5.3 (3.9%)	-10.5 (- 6.2%)	5.1 (2.7%)
Daytime $PM_{2.5}$						
Beijing	2.4 (2.0%)	8.5 (3.9%)	8.4 (5.5%)	-0.7 (- 0.6%)	-4.2 (- 3.2%)	10.7 (8.3%)
BTH	6.0 (4.9%)	12.9 (5.9%)	6.6 (5.2%)	5.3 (4.0%)	-6.2 (- 3.8%)	6.4 (3.8%)
	Up to 26.4	Up to 55.4	Up to 26.5	Up to 21.1	Up to 22.8	Up to 60.9

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