

1 **Two-way coupled meteorology and air quality models in Asia: a systematic review**
2 **and meta-analysis of impacts of aerosol feedbacks on meteorology and air quality**
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10 **Abstract**

11 Atmospheric aerosols can exert influence on meteorology and air quality through aerosol-
12 radiation interactions (ARI) and aerosol-cloud interactions (ACI) and this two-way feedback has
13 been studied by applying two-way coupled meteorology and air quality models. As one of regions
14 with high aerosol loading in the world, Asia has attracted many researchers to investigate the aerosol
15 effects with several two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE,
16 WRF-NAQPMS and GATOR-GCMOM) over the last decade. This paper attempts to offer
17 bibliographic analysis regarding the current status of applications of two-way coupled models in
18 Asia, related research focuses, model performances and the effects of ARI or/and ACI on
19 meteorology and air quality. There are total 160 peer-reviewed articles published between 2010 and
20 2019 in Asia meeting the inclusion criteria, with more than 79 % of papers involving the WRF-
21 Chem model. The number of relevant publications has an upward trend annually and East Asia,
22 India, China, as well as the North China Plain are the most studied areas. The effects of ARI and
23 both ARI and ACI induced by natural aerosols (particularly mineral dust) and anthropogenic
24 aerosols (bulk aerosols, different chemical compositions and aerosols from different sources) are
25 widely investigated in Asia. Through the meta-analysis of surface meteorological and air quality
26 variables simulated by two-way coupled models, the model performance affected by aerosol
27 feedbacks depends on different variables, simulation time lengths, selection of two-way coupled
28 models, and study areas. Future research perspectives with respect to the development, improvement,
29 application, and evaluation of two-way coupled meteorology and air quality models are proposed.

30 **1 Introduction**

31 Atmospheric pollutants can affect local weather and global climate via many mechanisms as
32 extensively summarized in the Intergovernmental Panel on Climate Change (IPCC) reports (IPCC,
33 2007, 2014, 2021), and also exhibit impacts on human health and ecosystems (Lelieveld et al., 2015;
34 Wu and Zhang, 2018). Atmospheric pollutants can modify the radiation energy balance, thus
35 influence meteorological conditions (Gray et al., 2010; Yiğit et al., 2016). Compared to other climate
36 agents, the short-lived and localized aerosols could induce changes in meteorology and climate
37 through aerosol-radiation interactions (ARI, Sathesh and Moorthy, 2005; Tremback et al., 1986)
38 and aerosol-cloud interactions (ACI, Lohmann and Feichter, 2005; Martin and Leight, 1949) or both
39 (Haywood and Boucher, 2000; Sud and Walker, 1990). ARI (previously known as direct effect and
40 semi-direct effect) are based on scattering and absorbing solar radiation by aerosols as well as cloud
41 dissipation by heating (Ackerman et al., 2000; Koch and Genio, 2010; McCormick and Ludwig,
42 1967; Wilcox, 2012), and ACI (known as indirect effect) are concerned with aerosols altering albedo
43 and lifetime of clouds (Albrecht, 1989; Lohmann and Feichter, 2005; Twomey, 1977). As our
44 knowledge base of aerosol-radiation-cloud interactions that involve extremely complex physical
45 and chemical processes has been expanding, accurately assessing the effects of these interactions
46 still remains a big challenge (Chung, 2012; Fan et al., 2016; Kuniyal and Guleria, 2019; Rosenfeld
47 et al., 2019, 2008).

48 The interactions between air pollutants and meteorology can be investigated by observational
49 analyses and/or air quality models. So far, many observational studies using measurement data from
50 a variety of sources have been conducted to analyze these interactions (Bellouin et al., 2008; Groß
51 et al., 2013; Rosenfeld et al., 2019; Wendisch et al., 2002). Yu et al. (2006) reviewed research work
52 that adopted satellite and ground-based measurements to estimate the ARI-induced changes of
53 radiative forcing and the associated uncertainties in the analysis. Yoon et al. (2019) analyzed the
54 effects of aerosols on the radiative forcing based on the Aerosol Robotic Network observations and

57 demonstrated that these effects depended on aerosol types. On the other hand, since the uncertainties
58 in ARI estimations were associated with ACI (Kuniyal and Guleria, 2019), the simultaneous
59 assessments of both ARI and ACI effects were needed and had gradually been conducted via satellite
60 observations (Illingworth et al., 2015; Kant et al., 2019; Quaas et al., 2008; Sekiguchi et al., 2003).
61 In the early stages, observational studies of ACI effects were based on several cloud parameters
62 mainly derived from surface-based microwave radiometer (Kim et al., 2003; Liu et al., 2003) and
63 cloud radar (Feingold et al., 2003; Penner et al., 2004). Later on, with the further development of
64 satellite observation technology and enhanced spatial resolution of satellite measurement comparing
65 against traditional ground observations, the satellite-retrieved cloud parameters (effective cloud
66 droplet radius, liquid water path (LWP) and cloud cover) were utilized to identify the ACI effects
67 studies on cloud scale. (Goren and Rosenfeld, 2014; Rosenfeld et al., 2014). Moreover, in order to
68 clarify whether aerosols affect precipitation positively or negatively, the effects of ACI on cloud
69 properties and precipitation were widely investigated but with various answers (Andreae and
70 Rosenfeld, 2008; Casazza et al., 2018; Fan et al., 2018; Rosenfeld et al., 2014). Analyses of satellite
71 and/or ground observations revealed that increased aerosols could suppress (enhance) precipitation
72 in drier (wetter) environments (Donat et al., 2016; Li et al., 2011; Rosenfeld, 2000; Rosenfeld et al.,
73 2008). Most recently, Rosenfeld et al. (2019) further used satellite-derived cloud information
74 (droplet concentration and updraft velocity at cloud base, LWP at cloud cores, cloud geometrical
75 thickness and cloud fraction) to single out ACI under a certain meteorological condition, and found
76 that the cloudiness change caused by aerosol in marine low-level clouds was much greater than
77 previous analyses (Sato and Suzuki, 2019). Despite the fact that aforementioned studies had
78 significantly improved our understanding of aerosol effects, many limitations still exist, such as low
79 temporal resolution of satellite data, low spatial resolution of ground monitoring sites and lack of
80 vertical distribution information of aerosol and cloud (Rosenfeld et al., 2014; Sato and Suzuki, 2019;
81 Yu et al., 2006).

82 Numerical models can also be used to study the interactions between air pollutants and
83 meteorology. Air quality models simulate physical and chemical processes in the atmosphere (ATM)
84 and are classified as offline and online models (El-Harbawi, 2013). Offline models (also known as
85 traditional air quality models) require outputs from meteorological models to subsequently drive
86 chemical models (Byun and Schere, 2006; ENVIRON, 2008; Seaman, 2000). Comparing to online
87 models, offline models usually are computationally efficient but incapable of capturing two-way
88 feedbacks between chemistry and meteorology (North et al., 2014). Online models or coupled
89 models are designed and developed to consider the two-way feedbacks and attempted to accurately
90 simulate both meteorology and air quality (Briant et al., 2017; Grell et al., 2005; Wong et al., 2012).
91 Two-way coupled models can be generally categorized as integrated and access models based on
92 whether using a coupler to exchange variables between meteorological and chemical modules
93 (Baklanov et al., 2014). As Zhang (2008) pointed out, Jacobson (1994, 1997) and Jacobson et al.
94 (1996) pioneered the development of a fully-coupled model named Gas, Aerosol, Transport,
95 Radiation, General Circulation, Mesoscale, and Ocean Model (GATOR-GCMOM) in order to
96 investigate all the processes related to ARI and ACI. Currently, there are three representative two-
97 way coupled meteorology and air quality models, namely the Weather Research and Forecasting-
98 Chemistry (WRF-Chem) (Grell et al., 2005), WRF coupled with Community Multiscale Air Quality
99 (CMAQ) (Wong et al., 2012) and WRF coupled with a multi-scale chemistry-transport model for
100 atmospheric composition analysis and forecast (WRF-CHIMERE) (Briant et al., 2017). The WRF-
101 Chem is an integrated model that includes various chemical modules in the meteorological model
102 (i.e., WRF) without using a coupler. For the remaining two models, which belong to access model,
103 the WRF-CMAQ uses a subroutine called *aqprep* (Wong et al., 2012) as its coupler while the WRF-
104 CHEMERE a general coupling software named Ocean Atmosphere Sea Ice Soil-Model Coupling
105 Toolkit (Craig et al. 2017). With more growing interest in coupled models and their developments,
106 applications and evaluations, two review papers thoroughly summarized the related works published
107 before 2008 (Zhang, 2008) and 2014 (Baklanov et al., 2014). Zhang (2008) overviewed the
108 developments and applications of five coupled models in the United States (US) and the treatments
109 of chemical and physical processes in these coupled models with emphasis on the ACI related
110 processes. Another paper presented a systematic review on the similarities and differences of
111 eighteen integrated or access models in Europe and discussed the descriptions of interactions
112 between meteorological and chemical processes in these models as well as the model evaluation
113 methodologies involved (Baklanov et al., 2014). Some of these coupled models can not only be used

114 to investigate the interactions between air quality and meteorology at regional scales but also at
115 global and hemispheric scales (Grell et al., 2011; Jacobson, 2001; Mailler et al., 2017; Xing et al.,
116 2015a), but large scale studies were not included in the two review papers by Zhang (2008) and
117 Baklanov et al. (2014). These reviews only focused on application and evaluation of coupled models
118 in US and Europe but there is still no systematic review targeting two-way coupled model
119 applications in Asia.

120 Compared to US and Europe, Asia has been suffering more severe air pollution in the past three
121 decades (Bollasina et al., 2011; Gurjar et al., 2016; Rohde and Muller, 2015) due to the rapid
122 industrialization, urbanization and population growth together with unfavorable meteorological
123 conditions (Jeong and Park, 2017; Lelieveld et al., 2018; Li M. et al., 2017). Then, the interactions
124 between atmospheric pollution and meteorology in Asia, which have received a lot of attention from
125 scientific community, are investigated using extensive observations and a certain number of
126 numerical simulations (Li et al., 2016; Nguyen et al., 2019a; Wang et al., 2010). Based on airborne,
127 ground-based, and satellite-based observations, multiple important experiments have been carried
128 out to analyze properties of radiation, cloud and aerosols in Asia, as briefly reviewed by Lin N. et
129 al. (2014). Recent observational studies confirmed that increasing aerosol loadings play important
130 roles in radiation budget (Banas et al., 2020; Eck et al., 2018), cloud properties (Dahutia et al., 2019;
131 Yang et al., 2019), precipitation intensity along with vertical distributions of precipitation types
132 (Guo et al., 2018, 2014). According to previous observational studies in Southeast Asia (SEA), Tsay
133 et al. (2013) and Lin N. et al. (2014) comprehensively summarized the spatiotemporal characteristics
134 of biomass burning (BB) aerosols and clouds as well as their interactions. Li et al. (2016) analyzed
135 how ARI or ACI influenced climate/meteorology in Asia utilizing observations and climate models.
136 With regard to the impacts of aerosols on cloud, precipitation and climate in East Asia (EA), a
137 detailed review of observations and modeling simulations has also been presented by Li Z. et al.
138 (2019). Since the 2000s, substantial progresses have been made in the climate-air pollution
139 interactions in Asia based on regional climate models simulations, which have been summarized by
140 Li et al. (2016). Moreover, starting from year of 2010, with the development and availability of two-
141 way coupled meteorology and air quality models, more and more modeling studies have been
142 conducted to explore the ARI or/and ACI effects in Asia (Nguyen et al., 2019a; Sekiguchi et al.,
143 2018; Wang et al., 2010; Wang J. et al., 2014). In recent studies, a series of WRF-Chem and WRF-
144 CMAQ simulations were performed to assess the consequences of ARI on radiative forcing,
145 planetary boundary layer height (PBLH), precipitation, and fine particulate matter (PM_{2.5}) and ozone
146 concentrations (Huang et al., 2016; Nguyen et al., 2019a; Sekiguchi et al., 2018; J. Wang et al.,
147 2014). Different from current released version of WRF-CMAQ model (based on WRF version 4.3
148 and CMAQ version 5.3.3) that only includes ARI, WRF-Chem with ACI (starting from WRF-Chem
149 version 3.0, Chapman et al., 2009) has been implemented for analyzing the complicated aerosol
150 effects that lead to variations of cloud properties, precipitations and PM_{2.5} concentrations (Bai et al.,
151 2020; Liu Z. et al., 2018; Park et al., 2018; Zhao et al., 2017). To quantify the individual or joint
152 effects of ARI or/and ACI on meteorological variables and pollutants concentrations, several
153 modeling studies have been performed in Asia (Chen et al., 2019a; Ma et al., 2016; Zhang B. et al.,
154 2015; Zhang et al., 2018). In addition, model comparisons (including offline and online models)
155 targeting EA have been carried out recently under the Model Inter-Comparison Study for Asia
156 (MICS-Asia) phase III (Chen et al., 2019b; Gao M. et al., 2018a; Li J. et al., 2019). As mentioned
157 above, even though there are already several reviews regarding the observational studies of ARI
158 or/and ACI (Li et al., 2016; Li Z. et al., 2019S; Lin N. et al., 2014; Tsay et al., 2013) it is necessary
159 to conduct a systematic review in Asia focusing on applications of two-way coupled meteorology
160 and air quality models as well as simulated variations of meteorology and air quality induced by
161 aerosol effects.

162 This paper is constructed as follows: Section 2 describes the methodology for literature
163 searching, paper inclusion, and analysis; Section 3 summarizes the basic information about
164 publications as well as developments and applications of coupled models in Asia and Section 4
165 provides the recent overviews of their research points. Sections 5 to 6 present systematic review and
166 meta-analysis of the effects of aerosol feedbacks on model performance, meteorology and air quality
167 in Asia. The summary and perspective are provided in Section 7.

169 **2 Methodology**

170 **2.1 Criteria and synthesis**

171 Since 2010, in Asia, regional studies of aerosol effects on meteorology and air quality based
172 on coupled models have been increasing gradually, therefore in this study we performed a systematic
173 search of literatures to identify relevant studies from January 1, 2010 to December 31, 2019. In
174 order to find all the relevant papers in English, Chinese, Japanese and Korean, we deployed several
175 science-based search engines, including Google Scholar, the Web of Science, the China National
176 Knowledge Infrastructure, the Japan Information Platform for S&T Innovation, the Korean Studies
177 Information Service System. The different keywords and their combinations for paper searching are
178 as follows: (1) model-related keywords including “coupled model”, “two-way”, “WRF”, “NU-
179 WRF”, “WRF-Chem”, “CMAQ”, “WRF-CMAQ”, “CAMx”, “CHIMERE”, “WRF-CHIMERE”
180 and “GATOR-GCMOM”; (2) effect-related keywords including “aerosol radiation interaction”,
181 “ARI”, “aerosol cloud interaction”, “ACI”, “aerosol effect” and “aerosol feedback”; (3) air
182 pollution-related keywords including “air quality”, “aerosol”, “PM2.5”, “O3”, “CO”, “SO2”,
183 “NO2”, “dust”, “BC”, “black carbon”, “blown carbon”, “carbonaceous”, “primary pollutants”; (4)
184 meteorology-related keywords including “meteorology”, “radiation”, “wind”, “temperature”,
185 “specific humidity”, “relative humidity”, “planetary boundary layer”, “cloud” and “precipitation”;
186 (5) region-related keywords including “Asia”, “East Asia”, “Northeast Asia”, “South Asia”,
187 “Southeast Asia”, “Far East”, “China”, “India”, “Japan”, “Korea”, “Singapore”, “Thailand”,
188 “Malaysia”, “Nepal”, “North China Plain”, “Yangtze River Delta”, “Pearl River Delta”, “middle
189 reaches of the Yangtze River”, “Sichuan Basin”, “Guanzhong Plain”, “Northeast China”,
190 “Northwest China” “East China”, “Tibet Plateau”, “Taiwan”, “northern Indian”, “southern Indian”,
191 “Gangetic Basin”, “Kathmandu Valley”.

192 After applying the search engines and the keywords combinations mentioned above, we found
193 946 relevant papers. In order to identify which paper should be included or excluded in this paper,
194 following criteria were applied: (1) duplicate literatures were deleted; (2) studies of using coupled
195 models in Asia with aerosol feedbacks turned on were included, and observational studies of aerosol
196 effects were excluded; (3) publications involving coupled climate model were excluded. According
197 to these criteria, not only regional studies, but also studies using the coupled models at global or
198 hemispheric scales involving Asia or its subregions were included. Then, we carefully examined all
199 the included papers and further checked the listed reference in each paper to make sure that no
200 related paper was neglected. A flowchart that illustrated the detailed procedures applied for article
201 identification is presented in Appendix Figure A1 (Note: Although the deadline for literature
202 searching is 2019, any literature published in 2020 is also included.). There was a total of 160
203 publications included in our study.

204 **2.2 Analysis method**

205 To summarize the current status of coupled models applied in Asia and quantitatively analyze
206 the effects of aerosol feedbacks on model performance as well as meteorology and air quality, we
207 carried out a series of analyses based on data extracted from the selected papers. We firstly compiled
208 the publication information of the included papers as well as the information regarding model name,
209 simulated time period, study region, simulation design, and aerosol effects. Secondly, we
210 summarized the important findings of two-way coupled model applications in Asia according to
211 different aerosol sources and components to clearly acquire what are the major research focuses in
212 past studies. Finally, we gathered all the simulated results of meteorological and air quality variables
213 with/out aerosol effects and their statistical indices (SI). For questionable results, the quality
214 assurance was conducted after personal communications with original authors to decide whether
215 they were deleted and/or corrected. All the extracted publication and statistical information were
216 exported into an Excel file, which was provided in Table S1. Moreover, we performed quantitative
217 analyses of the effects of aerosol feedbacks through following steps. (1) We discussed whether
218 meteorological and air quality variables were overestimated or underestimated based on their SI.
219 Then, variations of the SI of these variables were further analyzed in detail with/out turning on ARI
220 or/and ACI in two-way coupled models. (2) We investigated the SI of simulation results at different
221 simulation time lengths and spatial resolutions in coupled models. (3) More detailed inter-model
222 comparisons of model performance based on the compiled SI among different coupled models are
223 conducted. (4) Differences in simulation results with/out aerosol feedbacks were grouped by study

225 regions and time scales (yearly, seasonal, monthly, daily and hourly). Toward a better understanding
226 of the complicated interactions between air quality and meteorology in Asia, the results sections in
227 this paper are organized following above analysis methods (1) - (3) and represented in Section 5,
228 and the results following method (4) were represented in Section 6. In addition, Excel and Python
229 were used to conduct data processing and plotting in this study.

230 3 Basic overview

231 3.1 Summary of applications of coupled models in Asia

233 A total of 160 articles were selected according to the inclusion criteria, and their basic
234 information was compiled in Table 1. In Asia, five two-way coupled models are applied to study the
235 ARI and ACI effects. These include GATOR-GCMOM, two commonly used models, i.e., WRF-
236 Chem and WRF-CMAQ, and two locally developed models, i.e., the global-regional assimilation
237 and prediction system coupled with the Chinese Unified Atmospheric Chemistry Environment
238 forecasting system (GRAPES-CUACE) and WRF coupled with nested air-quality prediction
239 modeling system (WRF-NAQPMS). 127 out of total 160 papers involved the applications of WRF-
240 Chem in Asia since its two-way coupled version was publicly available in 2006 (Fast et al., 2006).
241 WRF-CMAQ was applied in only 16 studies due to its later initial release in 2012 (Wong et al.,
242 2012). GRAPES-CUACE was developed by the China Meteorological Administration and
243 introduced in details in Zhou et al. (2008, 2012, 2016), then firstly utilized in Wang et al. (2010) to
244 estimate impacts of aerosol feedbacks on meteorology and dust cycle in EA. The coupled version
245 of WRF-NAQPMS was developed by the Institute of Atmospheric Physics, Chinese Academy of
246 Sciences and could improve the prediction accuracy of haze pollution in the North China Plain (NCP)
247 (Wang Z.F. et al., 2014). Note that GRAPES-CUACE and WRF-NAQPMS were only applied in
248 China. There were only three published papers about the applications of GATOR-GCMOM in
249 Northeast Asia (NEA), NCP and India. In the included papers, 93, 33, 31 studies targeted various
250 areas in China, EA and India, respectively. There were 79 papers regarding effects of ARI (7 health),
251 63 both ARI and ACI (1 health) and 18 ACI. ACI studies were much less than ARI related ones,
252 which indicated that ACI related studies need to be paid with more attention in the future.
253 Considering that the choices of cloud microphysics and radiation schemes can affect coupled models'
254 results (Baró et al., 2015; Jimenez et al., 2016), these schemes used in the selected studies were also
255 summarized in Table 1. This table presents a concise overview of coupled models' applications in
256 Asia with the purpose of providing basic information regarding models, study periods and areas,
257 aerosol effects, scheme selections, and reference. More complete information is summarized Table
258 S1 including model version, horizontal resolution, vertical layer, aerosol and gas phase chemical
259 mechanisms, photolysis rate, PBL, land surface, surface layer, cumulus, urban canopy schemes,
260 meteorological initial and boundary conditions (ICs and BCs), chemical ICs and BCs, spin-up time,
261 and anthropogenic natural emissions.

262 It should be noted that in Table 1 there were four model inter-comparison studies that aimed at
263 evaluating model performance, identifying error sources and uncertainties, and providing optimal
264 model setups. By comparing simulations from two coupled models (WRF-Chem and Spectral
265 Radiation-Transport Model for Aerosol Species) (Takemura et al., 2003) in India (Govardhan et al.,
266 2016), it was found that the spatial distributions of various aerosol species (black carbon (BC),
267 mineral dust and sea salt) were similar with the two models. Based on the intercomparisons of WRF-
268 Chem simulations in different areas, Yang et al. (2017) revealed that aerosol feedbacks could
269 enhance PM_{2.5} concentrations in the Indo-Gangetic Plain but suppress the concentrations in the
270 Tibetan Plateau (TP). Targeting China and India, Gao M. et al. (2018b) also applied the WRF-Chem
271 model to quantify the contributions of different emission sectors to aerosol radiative forcings,
272 suggesting that reducing the uncertainties in emission inventories were critical, especially for India.
273 Moreover, for the NCP region, Gao M. et al. (2018a) presented a comparison study with multiple
274 online models under the MICS-Asia Phase III and pointed out noticeable discrepancies in the
275 simulated secondary inorganic aerosols under heavy haze conditions and the importance of accurate
276 wind speed at 10 meters above surface (WS10) predictions by these models. Comprehensive
277 comparative studies for Asia have been emerging lately but are still limited, comparing to those for
278 North America and Europe, such as the Air Quality Model Evaluation International Initiative Phase
279 II (Brunner et al., 2015; Campbell et al., 2015; Forkel et al., 2016; Im et al., 2015a, 2015b; Kong et
280 al., 2015; Makar et al., 2015a, 2015b; Wang K. et al., 2015).

Table 1. Basic information of coupled model applications in Asia during 2010-2019.

No.	Model	Study period	Region	Aerosol effect	Short/long-wave radiation scheme	Microphysics scheme	Reference
1	WRF-Chem	2013	India	ARI	Dudhia/RRTM	Thompson	Singh et al. (2020)*
2	WRF-Chem	12/2015	India	ARI	Goddard/RRTM	Lin	Bharali et al. (2019)
3	WRF-Chem	10/13/2016 to 11/20/2016	India	ARI	RRTMG	†	Shahid et al. (2019)
4	WRF-Chem	12/27/2017 to 12/30/2017	NCP	ARI	RRTMG	Lin	Wang D. et al. (2019)
5	WRF-Chem	12/05/2015 to 01/04/2016	NCP	ARI	Goddard	WSM 6-class graupel	Wu et al. (2019a)
6	WRF-Chem	12/05/2015 to 01/04/2016	NCP	ARI	Goddard	WSM 6-class graupel	Wu et al. (2019b)
7	WRF-Chem	06/01/2006 to 12/31/2011	NWC	ARI	RRTMG	Morrison	Yuan et al. (2019)
8	WRF-Chem	07/2016, 10/2016, 01/2017, 04/2017 02/17/2014 to 02/26/2014, 10/21/2014 to 10/25/2014, 11/05/2014 to 11/11/2014, 12/18/2015 to 12/24/2015	NCP	ARI	Goddard/RRTM	Lin	Zhang et al. (2019)
9	WRF-Chem	03/15/2012 to 03/25/2012	WA	ARI	RRTMG	Morrison	Zhou et al. (2019)
10	WRF-Chem	03/15/2012 to 03/25/2012	China & India	ARI	RRTMG	Morrison	Bran et al. (2018)
11	WRF-Chem	2013	CA	ARI	RRTMG	Lin	Gao M. et al. (2018b)
12	WRF-Chem	05/01/2007 to 05/07/2007	YRD	ARI	RRTMG	Lin	Li and Sokolik (2018)
13	WRF-Chem	06/02/2012 to 06/15/2012	YRD	ARI	RRTMG	Lin	Li M. et al. (2018)
14	WRF-Chem	12/15/2016 to 12/21/2016	NCP	ARI	RRTMG	Morrison	Liu Q. et al. (2018)
15	WRF-Chem	11/30/2016 to 12/04/2016	NCP	ARI	RRTMG	Lin	Miao et al. (2018)
16	WRF-Chem	2010	India	ARI	RRTMG	Morrison	Soni et al. (2018)
17	WRF-Chem	01/01/2013 to 01/31/2013	NCP	ARI	Goddard/RRTM	Lin	Wang L. et al. (2018)
18	WRF-Chem	12/2013	EC	ARI	RRTMG	Lin	Wang Z. et al. (2018)
19	WRF-Chem	2013	TP	ARI	RRTMG	Morrison	Yang et al. (2018)
20	WRF-Chem	03/11/2015 to 03/26/2015	EA	ARI	RRTMG	Lin	Zhou et al. (2018)
21	WRF-Chem	01/2013	EC	ARI	RRTMG	Lin	Gao et al. (2017a)
22	WRF-Chem	03/16/2014 to 03/18/2014	YRD	ARI	RRTMG	Lin	Li M. M. et al. (2017a)
23	WRF-Chem	10/15/2015 to 10/17/2015	YRD	ARI	Goddard/RRTM	Lin	Li M. M. et al. (2017b)
24	WRF-Chem	02/21/2014 to 02/27/2014	NCP	ARI	RRTMG	Lin	Qiu et al. (2017)
25	WRF-Chem	07/21/2012	NCP	ARI	RRTMG	Lin	Yang and Liu (2017a)
26	WRF-Chem	07/21/2012	NCP	ARI	RRTMG	Lin	Yang and Liu (2017b)
27	WRF-Chem	05/30/2013 to 06/27/2013	EC	ARI	RRTMG	Lin	Yao et al. (2017)
28	WRF-Chem	11/15/2013 to 12/30/2013	SEC	ARI	RRTMG	Lin	Zhan et al. (2017)
29	WRF-Chem	03/2012	India	ARI	RRTMG	Thompson	Feng et al. (2016)
30	WRF-Chem	1960-2010	NCP	ARI	Goddard/RRTM	Lin	Gao M. et al. (2016a)
31	WRF-Chem	01/2008, 04/2008, 07/2008, 10/2008	EA	ARI	Goddard/RRTM	Lin	Liu X. et al. (2016)
32	WRF-Chem	04/2011	NCP	ARI	RRTMG	Single-Moment 5-class	Liu L. et al. (2016)
33	WRF-Chem	09/21/2011 to 09/23/2011	NCP	ARI	RRTMG	Lin	Miao et al. (2016)
34	WRF-Chem	03/2005	EA	ARI	Goddard/RRTM	Morrison	Wang et al. (2016)
35	WRF-Chem	06/23/2008 to 07/20/2008	NWC	ARI	RRTMG	Morrison	Yang et al. (2016)
36	WRF-Chem	01/2007, 04/2007, 07/2007, 10/2007	EA	ARI	RRTM	Lin	Zhong et al. (2016)
37	WRF-Chem	05/2011, 10/2011	India	ARI	RRTMG	Thompson	Govardhan et al. (2015)
38	WRF-Chem	2006	China	ARI	RRTMG	Lin	Huang et al. (2015)
39	WRF-Chem	2007 to 2011	EA	ARI	Goddard/RRTM	Lin	Chen et al. (2014)
40	WRF-Chem	11/2007 to 12/2008	EA	ARI	RRTMG	Lin	Gao et al. (2014)
41	WRF-Chem	10/2006	SEA	ARI	RRTM	Lin	Ge et al. (2014)

42	WRF-Chem	04/17/2010 to 04/22/2010	India	ARI	RRTM	Thompson	Kumar et al. (2014)
43	WRF-Chem	01/11/2013 to 01/14/2013	NCP	ARI	Goddard/RRTM	Lin	Li and Liao (2014)
44	WRF-Chem	03/15/2008 to 03/18/2008	EA	ARI	RRTMG	Morrison	Lin C. et al. (2014)
45	WRF-Chem	07/21/2006 to 07/30/2006	NWC	ARI	RRTMG	Morrison	Chen et al. (2013)
46	WRF-Chem	05/12/2009 to 05/22/2009	India	ARI	Goddard/RRTM	Milbrandt-Yau	Dipu et al. (2013)
47	WRF-Chem	2008	India	ARI	Goddard/RRTM	Thompson	Kumar et al. (2012a)
48	WRF-Chem	2008	India	ARI	Goddard/RRTM	Thompson	Kumar et al. (2012b)
49	WRF-Chem	1999	India	ARI	Goddard/*	Lin	Seethala et al. (2011)
50	WRF-Chem	2006	China	ARI	†	†	Zhuang et al. (2011)
51	WRF-Chem	12/14/2013 to 12/16/2013	PRD	ARI & ACI	RRTMG	Morrison	Liu et al. (2020)*
52	WRF-Chem	11/30/2009 to 12/01/2009	NCP	ARI & ACI	Goddard/RRTM	Morrison	Jia et al. (2019)
53	WRF-Chem	11/25/2013 to 12/26/2013	EC	ARI & ACI	RRTMG	Lin	Wang Z. et al. (2019)
54	WRF-Chem	01/2014	China	ARI & ACI	RRTMG	Morrison	Archer-Nicholls et al. (2019)
55	WRF-Chem	12/01/2016 to 12/09/2016, 12/19/2016 to 12/24/2016	YRD	ARI & ACI	RRTMG	Lin	Li M. et al. (2019)
56	WRF-Chem	05/06/2013 to 20/06/2013 & 24/08/2014 to 08/09/2014	India	ARI & ACI	RRTM	Lin	Kedia et al. (2019a)
57	WRF-Chem	06/2010 to 09/2010	India	ARI & ACI	RRTM	Lin, Morrison, Thompson	Kedia et al. (2019b)
58	WRF-Chem	04/2013	PRD	ARI & ACI	RRTMG	Lin	Huang et al. (2019)
59	WRF-Chem	11/30/2013 to 12/10/2013	EC	ARI & ACI	RRTMG	Morrison	Ding et al. (2019)
60	WRF-Chem	12/01/2015	NCP	ARI & ACI	RRTMG	Lin	Chen et al. (2019a)
61	WRF-Chem	04/12/2015 to 27/12/2015	EA	ARI & ACI	Goddard	WSM 6-class graupel	An et al. (2019)
62	WRF-Chem	06/2015 to 02/2016	MRYR	ARI & ACI	Goddard/RRTM	WSM 6-class graupel	Liu L. et al. (2018)
63	WRF-Chem	06/2008, 06/2009, 06/2010, 06/2011, 06/2012	PRD	ARI & ACI	RRTMG	Morrison	Liu Z. et al. (2018)
64	WRF-Chem	01/2014, 04/2014, 07/2014, 10/2014	China	ARI & ACI	RRTMG	Lin	Zhang et al. (2018)
65	WRF-Chem	10/01/2015 to 10/26/2015	YRD	ARI & ACI	RRTMG	Lin	Gao J. et al. (2018)
66	WRF-Chem	2001, 2006, 2011	EA	ARI & ACI	RRTMG	Morrison	Zhang et al. (2017)
67	WRF-Chem	06/01/2011 to 06/06/2011	EC	ARI & ACI	Goddard/RRTM	Lin	Wu et al. (2017)
68	WRF-Chem	11/27/2013 to 12/12/2013	YRD	ARI & ACI	Goddard/RRTM	Single-Moment 5-class	Sun et al. (2017)
69	WRF-Chem	2005 & 2009	YRD	ARI & ACI	RRTMG	Morrison	Zhong et al. (2017)
70	WRF-Chem	11/05/2014 to 11/11/2014	NCP	ARI & ACI	Goddard/RRTM	Lin	Gao et al. (2017b)
71	WRF-Chem	01/2013	NCP	ARI & ACI	Goddard/RRTM	Lin	Gao et al. (2017c)
72	WRF-Chem	01/2010, 07/2010	China	ARI & ACI	†	†	Ma and Wen (2017)
73	WRF-Chem	06/01/2008 to 07/05/2008	India	ARI & ACI	†	†	Lau et al. (2017)
74	WRF-Chem	01/2013	NCP	ARI & ACI	Goddard/RRTM	Morrison	Kajino et al. (2017)
75	WRF-Chem	03/01/2009 to 03/31/2009	TP & India	ARI & ACI	RRTMG	Morrison	Yang et al. (2017)
76	WRF-Chem	2001, 2006, 2011	EA	ARI & ACI	RRTMG	Morrison	He et al. (2017)
77	WRF-Chem	05/2008 to 08/2008	YRD	ARI & ACI	†	†	Campbell et al. (2017)
78	WRF-Chem	12/07/2013 to 12/09/2013	EC	ARI & ACI	Goddard/RRTM	Morrison	Zhang Yue et al. (2016)
79	WRF-Chem	01/2006, 04/2006, 07/2006, 10/2006	China	ARI & ACI	Goddard/RRTM	Lin	Ma et al. (2016)
80	WRF-Chem	01/2005, 04/2005, 07/2005, 10/2005	EC	ARI & ACI	Goddard/RRTM	Lin	Zhang Yang et al. (2016a)
81	WRF-Chem	01/2005, 04/2005, 07/2005, 10/2005	EC	ARI & ACI	Goddard/RRTM	Lin	Zhang Yang et al. (2016b)
82	WRF-Chem	06/2012	EC	ARI & ACI	RRTMG	Lin	Huang et al. (2016)
83	WRF-Chem	01/2010, 07/2010	YRD	ARI & ACI	Goddard/RRTM	Lin	Xie et al. (2016)
84	WRF-Chem	11/12/2012 to 11/16/2012, 11/02/2013 to 11/06/2013	India	ARI & ACI	Goddard/RRTM	Lin	Srinivas et al. (2016)
85	WRF-Chem	07/2010	India	ARI & ACI	RRTMG	Lin	Kedia et al. (2016)

86	WRF-Chem	05/20/2008 to 08/31/2015	India	ARI & ACI	Goddard/RRTM	Lin	Jin et al. (2016a)
87	WRF-Chem	05/20/2008 to 08/31/2015	India	ARI & ACI	Goddard/RRTM	Lin	Jin et al. (2016b)
88	WRF-Chem	01/2010	NCP	ARI & ACI	Goddard/RRTM	Lin	Gao M. et al. (2016b)
89	WRF-Chem	01/05/2008 to 01/09/2008	NCP	ARI & ACI	RRTMG	Lin	Gao Y. et al. (2016)
90	WRF-Chem	12/2013	EC	ARI & ACI	RRTMG	Lin	Ding et al. (2016)
91	WRF-Chem	02/15/2013 to 02/17/2013	NCP	ARI & ACI	Goddard/RRTM	†	Yang et al. (2015)
92	WRF-Chem	01/2010, 04/2010, 07/2010, 10/2010	NCP	ARI & ACI	Goddard/RRTM	Lin	Shen et al. (2015)
93	WRF-Chem	2006 & 2011	EA	ARI & ACI	RRTMG	Morrison	Zhang Y. et al. (2015a)
94	WRF-Chem	2006 & 2011	EA	ARI & ACI	RRTMG	Morrison	Chen Y. et al. (2015)
95	WRF-Chem	06/27/2008 to 06/28/2008	NCP	ARI & ACI	RRTM	Lin	Zhong et al. (2015)
96	WRF-Chem	05/20/2008 to 08/31/2015	India	ARI & ACI	Goddard/RRTM	Lin	Jin et al. (2015)
97	WRF-Chem	03/2005, 04/2005, 05/2005	India	ARI & ACI	Goddard/RRTM	Thompson	Jena et al. (2015)
98	WRF-Chem	01/02/2013 to 01/26/2013	NCP	ARI & ACI	RRTMG	Morrison	Gao Y. et al. (2015)
99	WRF-Chem	07/08/2013 to 07/09/2013	SWC	ARI & ACI	RRTMG	†	Fan et al. (2015)
100	WRF-Chem	01/2010, 04/2010, 07/2010, 10/2010	NCP	ARI & ACI	Goddard/RRTM	Lin	Chen D. et al. (2015)
101	WRF-Chem	01/2013	EC	ARI & ACI	Goddard/RRTM	Lin	Zhang B. et al. (2015)
102	WRF-Chem	2006 & 2007	EA	ARI & ACI	Goddard/†	Lin	Wu et al. (2013)
103	WRF-Chem	09/27/2010 to 10/22/2010	India	ARI & ACI	Goddard/RRTM	Lin	Beig et al. (2013)
104	WRF-Chem	12/1/2009	NCP	ARI & ACI	Goddard/RRTM	Lin	Jia and Guo, (2012)
105	WRF-Chem	01/2001, 07/2001	EA	ARI & ACI	Goddard/RRTM	Lin	Zhang et al. (2012)
106	WRF-Chem	11/10/2007 to 01/01/2008	China	ARI & ACI	RRTMG	Lin	Gao et al. (2012)
107	WRF-Chem	06/18/2018 to 06/19/2018	MRYR	ACI	Goddard/RRTM	†	Bai et al. (2020)*
108	WRF-Chem	06/07/2017 to 06/12/2017	YRD	ACI	RRTMG	Morrison	Liu et al. (2019)
109	WRF-Chem	03/2010 to 05/2010	EA	ACI	RRTMG	Morrison	Wang K. et al. (2018)
110	WRF-Chem	03/09/2012 to 04/30/2012	EA	ACI	RRTMG	Thompson	Su and Fung (2018a)
111	WRF-Chem	03/09/2012 to 04/30/2012	EA	ACI	RRTMG	Thompson	Su and Fung (2018b)
112	WRF-Chem	05/18/2015 to 06/13/2015	NEA	ACI	RRTMG	Morrison	Park et al. (2018)
113	WRF-Chem	08/2008	EC	ACI	RRTMG	Lin	Gao and Zhang (2018)
114	WRF-Chem	10/03/2013 to 10/07/2013	SEC	ACI	RRTMG	Morrison	Shen et al. (2017)
115	WRF-Chem	01/2013, 07/2013	China	ACI	Fu-Liou-Gu	Morrison	Zhao et al. (2017)
116	WRF-Chem	06/04/2004 to 07/10/2004	India	ACI	Goddard	Lin	Bhattacharya et al. (2017)
117	WRF-Chem	09/20/2013 to 09/23/2013	PRD	ACI	RRTMG	Lin	Jiang et al. (2016)
118	WRF-Chem	2005 & 2010	EA	ACI	RRTMG	Morrison	Y. Zhang et al. (2015b)
119	WRF-Chem	08/20/2009 to 08/29/2008	India	ACI	Goddard/RRTM	Morrison	Sarangi et al. (2015)
		01/2001, 04/2001, 07/2001, 10/2001,					
120	WRF-Chem	01/2005, 04/2005, 07/2005, 10/2005, 01/2008, 04/2008, 07/2008, 10/2008	EA	ACI	†	†	Zhang et al. (2014)
121	WRF-Chem	07/2008	EC	ACI	RRTMG	Morrison	Lin et al. (2014)
122	WRF-Chem	1980 to 2010	SEC	ACI	†	†	Bennartz et al. (2011)
123	WRF-Chem	2008 & 2050	China	ARI (Health)	†	†	Zhong et al. (2019)
124	WRF-Chem	2015 & 2050	India	ARI (Health)	RRTM	Thompson	Conibear et al. (2018a)
125	WRF-Chem	2014	India	ARI (Health)	RRTM	Thompson	Conibear et al. (2018b)
126	WRF-Chem	2011	India	ARI (Health)	Goddard/RRTM	Thompson	Ghude et al. (2016)
127	WRF-Chem	2013	NCP	ARI (Health)	RRTMG	†	Gao M. et al. (2015)
128	WRF-CMAQ	03/2006 & 04/2006 to 03/2010 & 04/2010	EA	ARI	†	†	Dong et al. (2019)
129	WRF-CMAQ	04/10/2016 to 06/19/2016	NEA	ARI	RRTMG	Single-Moment 3-class	Jung et al. (2019)

130	WRF-CMAQ	2014	EA	ARI	RRTMG	Morrison	Nguyen et al. (2019a)
131	WRF-CMAQ	2014	SEA	ARI	RRTMG	Morrison	Nguyen et al. (2019b)
132	WRF-CMAQ	02/2015	NEA	ARI	RRTMG	Single-Moment 5-class	Yoo et al. (2019)
133	WRF-CMAQ	01/2014, 02/2014, 03/2014	EA	ARI	RRTMG	Morrison	Sekiguchi et al. (2018)
134	WRF-CMAQ	2006 to 2010, 2013	EA	ARI	RRTMG	Morrison	Hong et al. (2017)
135	WRF-CMAQ	01/2013, 07/2013	China	ARI	RRTMG	Morrison	Xing et al. (2017)
136	WRF-CMAQ	1990 to 2010	EA	ARI	RRTMG	Morrison	Xing et al. (2016)
137	WRF-CMAQ	1990 to 2010	EC	ARI	RRTMG	Morrison	Xing et al. (2015a)
138	WRF-CMAQ	1990 to 2010	EC	ARI	RRTMG	Morrison	Xing et al. (2015b)
139	WRF-CMAQ	1990 to 2010	EC	ARI	RRTMG	Morrison	Xing et al. (2015c)
140	WRF-CMAQ	01/2013	China	ARI	RRTMG	Morrison	J. Wang et al. (2014)
141	WRF-CMAQ	01/2013, 04/2013, 07/2013, 10/2013	China	ACI	RRTMG	Morrison	Chang (2018)
142	WRF-CMAQ	2050	China	ARI (Health)	RRTMG	Morrison	Hong et al. (2019)
143	WRF-CMAQ	1990 to 2010	EA & India	ARI (Health)	RRTMG	Morrison	Wang et al. (2017)
144	GRAPES-CUACE	12/15/2016 to 12/24/2016	NCP	ARI	Goddard	†	H. Wang et al. (2018)
145	GRAPES-CUACE	07/07/2008 to 07/11/2008	EC	ARI	CLIRAD	†	H. Wang et al. (2015)
146	GRAPES-CUACE	04/26/2006	EA	ARI	Goddard/†	†	Wang and Niu (2013)
147	GRAPES-CUACE	04/26/2006	EA	ARI	Goddard/†	†	Wang et al. (2013)
148	GRAPES-CUACE	07/13/2008 to 07/31/2008	NCP	ARI	†	†	Zhou et al. (2012)
149	GRAPES-CUACE	04/26/2006	EA	ARI	Goddard/†	†	Wang et al. (2010)
150	GRAPES-CUACE	01/2013	EC	ACI	†	Single-Moment 6-class	Zhou et al. (2016)
151	WRF-NAQPMS	2013	EA	ARI	†	†	Li J. et al. (2018)
152	WRF-NAQPMS	09/27/2013 to 10/01/2013	NCP	ARI	Goddard/RRTM	Lin	Wang Z. et al. (2014)
153	WRF-NAQPMS	01/01/2013	EC	ARI	Goddard/RRTM	Lin	Wang Z. F. et al. (2014)
154	GATOR-GCMOM	2000 & 2009	NEA	ARI & ACI	†	†	Ten Hoeve and Jacobson, 2012
155	GATOR-GCMOM	2002 & 2009	India	ARI & ACI	†	†	Jacobson et al. (2019)
156	GATOR-GCMOM	2000 & 2009	NCP	ARI & ACI	†	†	Jacobson et al. (2015)
157	Multi-model comparison	†	EA	ARI & ACI	†	†	Chen et al. (2019b)
158	Multi-model comparison	2010	EA	ARI & ACI	†	†	Li J. et al., (2019)
159	Multi-model comparison	01/2010	NCP	ARI & ACI	†	†	Gao et al. (2018a)
160	Multi-model comparison	05/2011	India	ARI & ACI	†	†	Govardhan et al. (2016)

283 †: Unclear; *: A preprint version of this study was available online on October 31, 2019, and was formally published on January 1, 284 2020. (EA: East Asia, NEA: Northeast Asia, SEA: Southeast Asia, EC: East China, NCP: North China Plain, YRD: Yangtze River Delta, 285 SEC: Southeast China, NWC: Northwest China, TP: Tibetan Plateau, MRYR: middle reaches of the Yangtze River, SWC: Southwest China; 286 PRD: Pearl River Delta).

288 3.2 Spatiotemporal distribution of publications

289 To gain an overall understanding of applications of coupled models in Asia, the spatial 290 distributions of study areas of the selected literatures and the temporal variations of the annual 291 publication numbers were extracted from Table 1 and summarized. Figure 1 illustrates the spatial 292 distributions of study regions as well as the number of papers involving coupled models in Asia (Fig. 293 1a) and China (Fig. 1b). In this figure, the color and number in the pie charts represent individual 294 (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS, and GATOR-GCMOM) or 295 multiple coupled models and the quantity of corresponding articles, respectively. At subregional 296 scales, most studies targeted EA where high anthropogenic aerosol loading occurred in recent 297 decades, mainly using WRF-Chem and WRF-CMAQ (Fig. 1a). For other subregions, such as NEA, 298 SEA, Central Asia (CA), and West Asia (WA), there were rather limited research activities taking 299 into account aerosol feedbacks with two-way coupled models. National scale applications of two-

way coupled models targeted mostly modeling domains covering India and China but much less work were carried out in other countries, such as Japan and Korea, where air pollution levels are much lower. With respect to various areas in China (Fig. 1b), the research activities concentrated mostly in NCP and secondly in the East China (EC), then in the Yangtze River Delta (YRD) and Pearl River Delta (PRD) areas. WRF-Chem was the most popular model applied in all areas, but there were a few applications of GRAPES-CUACE and WRF-NAQPMS in EC and NCP.

Figure 2 depicts the temporal variations of research activities with two-way coupled models in Asia over the period of 2010 to 2019. The total number of papers related to two-way coupled models had an obvious upward trend in the past decade. Prior to 2014, applications of two-way coupled models in Asia were scarce, with about 1 to 6 publications per year. A noticeable increase of research activities emerged starting from 2014 and the growth was rapid from 2014 to 2016, at a rate of 7-9 more papers per year, especially in China. It could be related to the Action Plan on Prevention and Control of Atmospheric Pollution (2013-2017) implemented by the Chinese government. The growth was rather flat during 2016-2018 before reaching a peak of 31 articles in 2019. In addition, the pie charts in Fig. 2 indicates that modeling activities had been picking up with a diversified pattern in study domain from 2010 to 2019. The modeling domains extended from EA to China and India and then several subregions in Asia and various areas in China. For EA and India, investigations of aerosol feedbacks based on two-way coupled models rose from 1-2 papers per year during 2010-2013 to 4-8 during 2014-2019. Since 2014, most model simulations were carried out towards areas with severe air pollution in China, especially the NCP area where attracted 5-7 publications per year.

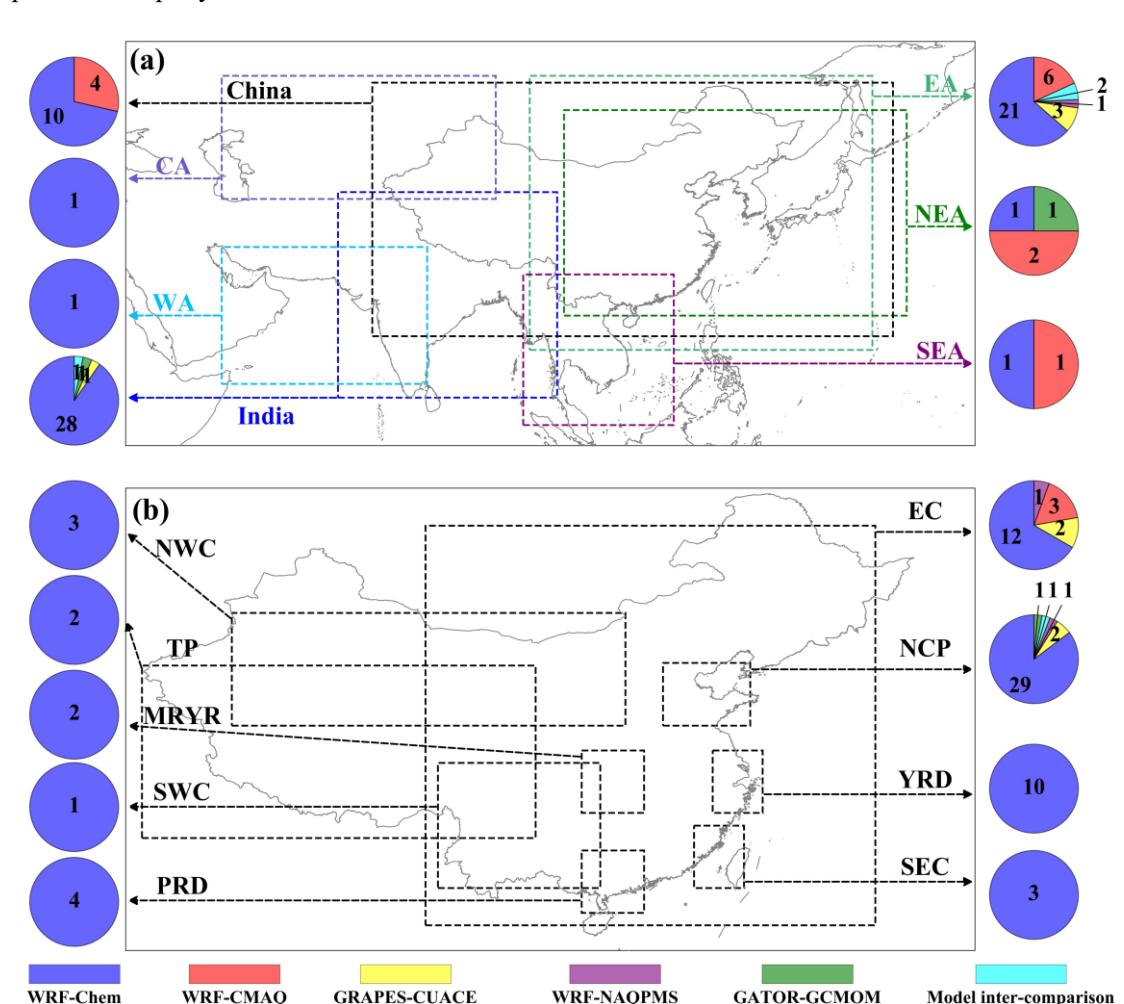
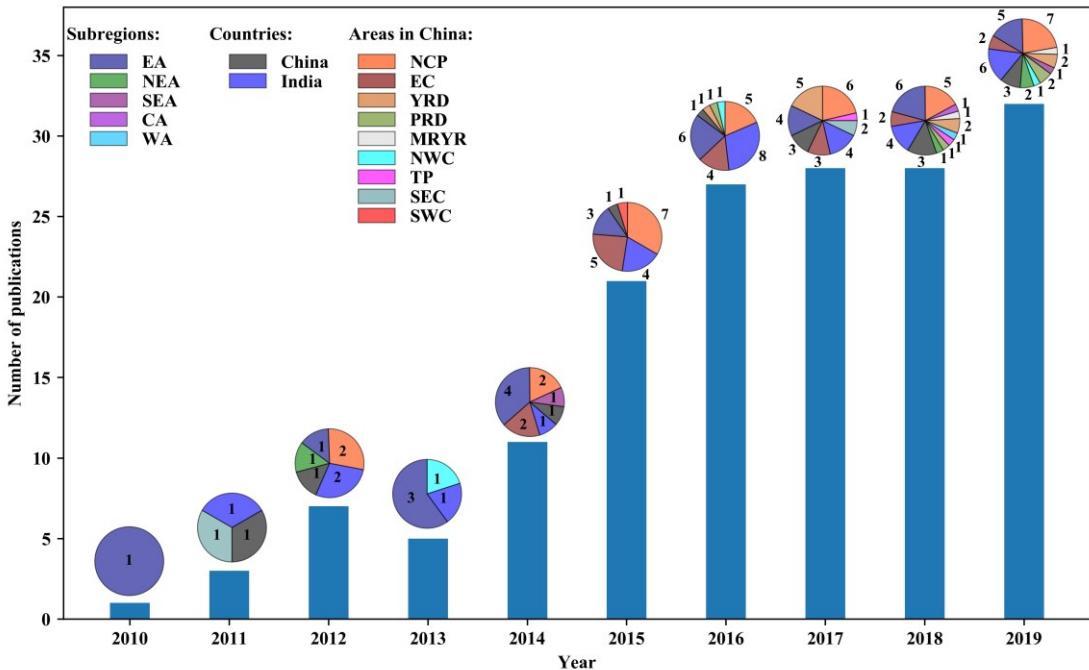


Figure 1. The spatial distributions of study domains as well as the two-way coupled modeling publication numbers in different subregions or countries of Asia (a) and areas of China (b). (EA: East Asia, NEA: Northeast Asia, SEA: Southeast Asia, EC: East China, NCP: North China Plain, YRD: Yangtze River Delta, SEC: Southeast China, NWC: Northwest China, TP: Tibetan Plateau, MRYR: middle reaches of the Yangtze River, SWC: Southwest China; PRD: Pearl River Delta).



329
330 Figure 2. The temporal variations of study activities adopting two-way coupled models in Asia during 2010-2019.
331 (EA: East Asia, NEA: Northeast Asia, SEA: Southeast Asia, EC: East China, NCP: North China Plain, YRD: Yangtze
332 River Delta, SEC: Southeast China, NWC: Northwest China, TP: Tibetan Plateau, MRYR: middle reaches of the
333 Yangtze River, SWC: Southwest China; PRD: Pearl River Delta).

3.3 Summary of modeling methodologies

336 The physiochemical processes involved with ARI and ACI are sophisticated in actual
337 conditions of atmospheric environment but their representations in two-way coupled models can be
338 rather different. Also, simulation results depend on how these models are configured and set up.
339 Therefore, the treatments of aerosol and cloud microphysics, and aerosol-radiation-cloud
340 interactions in WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-
341 GCMOM applied in Asia, as well as the various aspects of how the modeling studies being set up
342 in the selected papers are summarized in Tables 2-5, respectively, and outlined in this section.

343 Aerosol microphysics processes consist of particle nucleation, coagulation,
344 condensation/evaporation, gas/particle mass transfer, inorganic aerosol thermodynamic equilibrium,
345 aqueous chemistry and formation of secondary organic aerosol (SOA). Their representations in a
346 variety of aerosol mechanisms offered in the five two-way coupled models applied in Asia and
347 relevant references are compiled in Table 2. Note that the GOCART scheme in WRF-Chem is based
348 on a bulk aerosol mechanism that is not able to consider the details of these microphysics processes.
349 The binary homogeneous nucleation schemes with/out hydration developed by different authors are
350 applied in the five coupled models for simulating the new particle formation and GATOR-GCMOM
351 also adopts the ternary nucleation parameterization scheme for H_2SO_4 , NH_3 and H_2O vapors. All
352 the five coupled models calculate the aerosol-aerosol coagulation rate coefficients based the
353 Brownian coagulation theory, with certain enhancements in GATO-GCMOM as stated in details by
354 Jacobson (1999). The dynamic condensation/evaporation approaches of inorganic gases (e.g.,
355 H_2SO_4 , NH_3 , HNO_3 , and HCl) and organic gases (VOCs) based on the Fuchs-Sutugin expression
356 are implemented in various aerosol mechanisms offered by WRF-Chem, WRF-CMAQ, GRAPES-
357 CUACE, and WRF-NAQPMS, while GATOR-GCMOM deploys the condensation/evaporation
358 approach in which several terms of processes are factored in the 3-D equations of discrete size-
359 resolved aerosol growth (Jacobson, 2012). The mass transfer between gaseous and aerosol particles
360 are treated via two typical methods (i.e., bulk equilibrium and kinetic) in most coupled models, and
361 the hybrid and Henry's law equilibrium methods are also applied in the MADRID (WRF-Chem)
362 and the 6th/7th generation CMAQ aerosol modules (AERO6/AERO7) (WRF-CMAQ), respectively.
363 Different versions of the ISORROPIA module, the Model for an Aerosol Reacting System-version
364 A (MARS-A), the Multicomponent Equilibrium Solver for Aerosols with the Multicomponent
365 Taylor Expansion Method (MESA-MTEM), and the EQUISOLV version 2 (EQUISOLV

366 II) modules are implemented for computing the inorganic aerosol thermodynamic equilibrium in
 367 these two-way coupled models. For aqueous chemistry, the bulk aqueous chemistry scheme and
 368 variations of the CMAQ's standard aqueous chemistry module (AQCHEM) are the most applied,
 369 and the CBM-IV aqueous chemistry scheme, the Regional Acid Deposition Model (RADM)
 370 aqueous chemistry module, and the size-resolved aqueous chemistry module are utilized as well.
 371 Multiple approaches have been incorporated into the five coupled models for calculating the SOA
 372 formation and include the volatility basis set (VBS) approach, approaches considering reversible
 373 absorption or combined absorption and dissolution, fixed or bulk two-product yield approaches, and
 374 the approach of time-dependent organics condensation/evaporation with considering vapor pressure.
 375

376 Table 2. Treatments of aerosol microphysics processes in two-way coupled models (WRF-Chem, WRF-CMAQ,
 377 GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) applied in Asia.

WRF-Chem			WRF-CMAQ						GRAPES-CUACE		WRF-NAQPMS		GATOR-GCMOM
	GOCART	MADE/SORGAM	AEROS	MAM3/MAM7	MOSAIC	MADRID	AEROS	AERO6	AERO7	CUACE ³⁸	AEROS		
New particle formation/if with hydration	None	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Kulmala et al., 1998)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Kulmala et al., 1998)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Wexler, et al., 1994)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Wexler, et al., 1994)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (McMurtry and Friedlander, 1979)/Unclear	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Kulmala et al., 1998)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Vehkamäki et al., 2002)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Vehkamäki et al., 2002)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Kulmala et al., 1998)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Yu, 2006)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Vehkamäki et al., 2002)/Yes; H ₂ SO ₄ -NH ₃ -H ₂ O ternary homogeneous nucleation (Napari et al., 2002)/Yes	H ₂ SO ₄ -H ₂ O binary homogeneous nucleation (Vehkamäki et al., 2002)/Yes; H ₂ SO ₄ -NH ₃ -H ₂ O ternary homogeneous nucleation (Napari et al., 2002)/Yes
Coagulation	None	Brownian motion (Binkowski and Shankar, 1995)	Brownian motion (Binkowski and Roselle, 2003)	Brownian motion (Whitby, 1978)	Brownian motion (Jacobson et al., 1994)	Brownian motion (Jacobson et al., 1994)	Brownian motion (Binkowski and Roselle, 2003)	Brownian motion (Binkowski and Roselle, 2003)	Brownian motion (Binkowski and Roselle, 2003)	Brownian motion (Jacobson et al., 1994)	Brownian motion (Jacobson et al., 1994; Chen et al., 2017)	Brownian motion, Brownian diffusion enhancement, turbulent shear, turbulent inertial motion, gravitational setting, Van der Waals forces, viscous forces, fractal geometry (Jacobson, 2003)	Brownian motion, Brownian diffusion enhancement, turbulent shear, turbulent inertial motion, gravitational setting, Van der Waals forces, viscous forces, fractal geometry (Jacobson, 2003)
Condensation/ Evaporation	None	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor and VOCs based on Fuchs-Sutugin expression (Binkowski and Shankar, 1995); Condensation/ evaporation of volatile inorganic gases to/from the gas-phase concentrations of coarse particle surfaces using ISORROPIA in reverse mode (CMAQ User's Guide)	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor and VOCs based on Fuchs-Sutugin expression (Binkowski and Shankar, 1995); Condensation/ evaporation of volatile inorganic gases to/from the gas-phase concentrations of coarse particle surfaces using ISORROPIA in reverse mode (CMAQ User's Guide)	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor, NH ₃ (7 modes) and semi-volatile organics; Condensation/ evaporation of SOA gas (Liu et al., 2012)	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor, methanesulfonic acid, HNO ₃ , HCl and NH ₃ with adaptive step time-split Euler approach (Zaveri et al., 2008)	Dynamical condensation/ evaporation of semi-volatile species for analytical predictor of condensation with moving-center approach (Zhang et al., 2010)	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor and VOCs based on Fuchs-Sutugin expression (Binkowski and Shankar, 1995); Condensation/ evaporation of volatile inorganic gases to/from the gas-phase concentrations of coarse particle surfaces using ISORROPIA in reverse mode (CMAQ User's Guide)	Same as in AEROS	Same as in AEROS	Dynamical condensation/ evaporation of H ₂ SO ₄ vapor and gaseous precursors based on modified Fuchs-Sutugin expression (Jacobson, et al., 2019; Yu and Luo, 2009; Chen et al., 2019; Yu, 2006)	Condensation/ evaporation of H ₂ SO ₄ with advanced particle microphysics approach (Li et al., 2018; Yu and Luo, 2009; Chen et al., 2019; Yu, 2006)	Dynamical condensation of H ₂ O and involatile species with Analytical Predictor of Nucleation, Condensation, and Dissolution scheme (Jacobson, 2002); Evaporation of a volatile component over a single particle (Jacobson and Turco, 1995)	Dynamical condensation of H ₂ O and involatile species with Analytical Predictor of Nucleation, Condensation, and Dissolution scheme (Jacobson, 2002); Evaporation of a volatile component over a single particle (Jacobson and Turco, 1995)
Gas/particle mass transfer	None	1. Bulk equilibrium approach for HNO ₃ and NH ₃ (Zhang et al., 2005) 2. Kinetic approach for H ₂ SO ₄ (Zhang et al., 2016)	Kinetic approach for all species (Foley et al., 2010)	Bulk equilibrium approach for (NH ₄) ₂ SO ₄ (Hu and Zhang, 2014)	Kinetic approach for all species (Zaveri et al., 2008)	1. Bulk equilibrium approach for HNO ₃ and NH ₃ (Zhang et al., 2010) 2. Kinetic approach for all species (Zhang et al., 2010) 3. Hybrid approach (Zhang et al., 2010)	Kinetic approach for all species (Foley et al., 2010)	1. Henry's law equilibrium (Foley et al., 2017) 2. Kinetic approach for all species (Foley et al., 2017)	Same as in AEROS	Same as in AEROS	Kinetic approach for all species (Zhou et al., 2021)	Kinetic for all species (Chen et al., 2021)	Kinetic approach for all species (Jacobson, 1999)
Inorganic aerosol thermodynamic equilibrium	None	MARS-A (Binkowski and Shankar, 1995)	ISORROPIA (Byun and Kenneth, 2006)	ISORROPIA II (Hu and Zhang, 2014)	MESA-MTEM (Zaveri et al., 2008)	ISORROPIA (Zhang et al., 2010)	ISORROPIA (Byun and Kenneth, 2006)	ISORROPIA II (Appel et al., 2013)	ISORROPIA II (Appel et al., 2013)	ISSOROPIA (Zhou et al., 2012)	ISSOROPIA (Li et al., 2011)	EQUISOLV II (Jacobson, 1999)	
Aqueous chemistry	None	Bulk cloud-chemistry scheme (Fahey and Pandis, 2001; Zhang et al., 2015)	AQCHEM (Fahey et al., 2017)	Based on algorithm developed by Barth et al. (2001) (He and Zhang, 2014)	Same as in MADE/SORGAM (Fahey and Pandis, 2001; Chapman et al., 2009)	Same as in MADE/SORGAM (Fahey and Pandis, 2001; Chapman et al., 2009)	1. AQCHEM 2. AQCHEM-KMT (Fahey et al., 2017)	1. AQCHEM-KMT 2. AQCHEM-KMTI (Fahey et al., 2017)	1. AQCHEM-KMT 2. AQCHEM-KMTI (Fahey et al., 2017)	Based on aqueous chemistry in CBM-IV mechanism by Gery et al. (1989)	Based on the RADM mechanism used in CMAQ v4.6 (AEROS) (Li et al., 2011)	Bulk or size-resolved cloud-chemistry module (GATOR2012)	
SOA formation	None	1. Reversible absorption of 8 classes volatile organic compounds (VOCs) based on Caltech smog-chamber data (Odum et al., 2010)	Combined absorption and dissolution approaches for 9 parent VOCs and 32 SOA species (Carlton, et al., 2012)	Treatment of SOA from fixed mass yields for anthropogenic and biogenic precursor	1. Based on ambient ageing measurement of organic aerosols by Hodzic and Jimenez (2011) 2. Based on volatility basis set approach	1. Absorptive approach for 14 parent VOCs and 38 SOA species 2. Combined absorption and	Combined absorption and dissolution approaches for 9 parent VOCs and 32 SOA species	On the basis of SOA scheme in AEROS/6, updated parameterization of in-cloud SOA formation from biogenic VOCs (Foley et al., 2017)	On the basis of SOA scheme in AEROS/6, updated parameterization of monoterpane SOA yielded from photooxidation	Reversible absorption of 8 classes VOCs based on Caltech smog-chamber data (Zhou et al., 2012)	Bulk two-product yield parametrization (Fu et al., 2016; Odum et al., 1997)	Using Henry's Law to determine vapor pressure of organics and parametrize either time-dependent condensation or evaporation calculations.	

al., 1997; Griffin et al., 1999)	(Knott et al., 2014)	dissolution approaches for 42 hydrophilic and hydrophobic VOCs (Zhang et al., 2004)	(Foley et al., 2021)	(Jacobson, 2002)
2. Based on volatility basis set approach (Ahmadov et al., 2012)				

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*CUACE is the aerosol mechanism implemented in the GRAPES-CUACE model (Zhou et al., 2012).

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*GATOR2012 is the aerosol mechanism implemented in the GATOR-GCMOM model (Jacobson et al., 2012).

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In addition to aerosol microphysics processes, the cloud properties included in cloud microphysics schemes and the treatment of aerosol-cloud processes in the five two-way coupled models are different in terms of hydrometeor classes, cloud droplet size distribution, aerosol water uptake, in-/below-cloud scavenging, hydrometeor-aerosol coagulations, and sedimentation of aerosols and cloud droplets (Table 3). Among the microphysics schemes implemented in the five coupled models, mass concentrations of different hydrometeors (including cloud water, rain, ice, snow or graupel) are included but their number concentrations are only considered if the cloud microphysics schemes are two-moment or three-moment. The single modal approach with either lognormal or gamma distribution and the sectional approach with discrete size distributions for cloud droplets are applied in different microphysics schemes. Based on the Mie theory, WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM calculate cloud radiative properties (including extinction/scattering/absorption coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds) in their radiation schemes (e.g., RRTMG, GODDARD, GATOR2012). In atmosphere, the hygroscopic growth of aerosols due to water uptake is parameterized based on the Köhler or Zdanovskii-Stokes-Robinson theory and the hysteresis effects depending on the deliquescence and crystallization RH are taken into account in the five coupled models. The removal processes of aerosol particles include wet removal and sedimentation. Aerosol particles in accumulation and coarse modes can act as CCN or IN via activations in cloud, which can further develop to different types of hydrometeors (cloud water, rain, ice, snow and graupel), and then gradually form precipitations. These processes are named as in-cloud scavenging or rainout. The aerosol particles below cloud base also can be coagulated with the falling hydrometeors, which are known as below-cloud scavenging or wash out. Both representations of in- and below-cloud scavenging processes are based on scavenging rate approach in aerosol mechanisms of WRF-Chem, WRF-CMAQ, GRAPES-CUACE and WRF-NAQPMS except GATOR-GCMOM. Size-resolved sedimentation of aerosols are computed from one model layer to layers below down to the surface layer using settling velocity in most coupled models and the MOSAIC aerosol mechanism in WRF-Chem only considers the sedimentation in the lowest model level (Marelle et al., 2017).

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Table 3. Compilation of cloud properties and aerosol-cloud processes in two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) applied in Asia.

	WRF-Chem	WRF-CMAQ	GRAPES-CUACE	WRF-NAQPMS	GATOR-GCMOM
Hydrometeor (Cloud microphysics scheme)	Mass concentrations: Cloud water, rain, ice, snow and graupel (Morrison, Lin, Thompson, WSM 6 class and Milbrandt-Yau) Cloud water, rain, ice and snow (WSM 5 class) Number concentrations: Rain, ice, snow and graupel (Morrison and Milbrandt-Yau) Rain and ice (Thompson) None (Lin, WSM 5 class and WSM 6 class)	Mass concentrations: Cloud water, rain, ice, snow and graupel (Morrison) Cloud water, rain, ice and snow (WSM 5 class) Cloud water and rain (WSM 3 class) Number concentrations: Rain, ice, snow and graupel (Morrison) None (WSM 3 class and WSM 5 class)	Mass concentrations: Cloud water, rain, ice, snow and graupel (WSM 6 class) Number concentrations: None (WSM 6 class)	Mass concentrations Cloud water, rain, ice, snow and graupel (Lin) Number concentrations: None (Lin)	Mass concentrations: Cloud water, ice and graupel (GATOR2012) Number concentrations: Cloud water, ice and graupel (GATOR2012)
Cloud droplet size distribution (Cloud microphysics scheme)	1. Single, modal approach with lognormal distribution (Morrison and Lin) 2. Gamma distribution (Thompson, WSM 5 class and WSM 6 class)	1. Single, modal approach with lognormal distribution (Morrison) 2. Gamma distribution (WSM 3 class and WSM 5 class)	Gamma distribution (WSM 6 class)	Single, modal approach with lognormal distribution (Lin)	Sectional approach with multiple size distributions (GATOR2012*) (Jacobson, et al., 2007)
Cloud radiative properties (Radiation scheme)	Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds based on Mie scattering theory (RRTMG SW) Absorption coefficient of liquid and ice clouds using constant values (RRTMG LW) Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds from lookup tables (Goddard SW and LW)	Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds based on Mie scattering theory (RRTMG SW) Absorption coefficient of liquid and ice clouds using constant values (RRTMG LW)	Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds using lookup tables (Goddard SW) Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds from lookup tables (Goddard LW)	Extinction coefficient, single scattering albedo and asymmetry factor of liquid and ice clouds using lookup tables (Goddard SW) Clear sky optical depth from lookup table (RRTM LW)	Integrating spectral optical properties over each size bin of each hydrometeor particle size distribution (Toon SW and LW) (Jacobson and Jadhav, 2018)
Aerosol water uptake	Equilibrium with RH based on Köhler theory, and hysteresis is treated (Ghan and Zaveri, 2007)	The empirical equations of deliquescence and crystallization RH developed by Martin et al (2003), and hysteresis is treated (CMAQ source code)	Equilibrium with the mutual deliquescence and crystallization RH using the Zdanovskii-Stokes-Robinson equation, and hysteresis is treated (Personal communication)	Equilibrium with the mutual deliquescence and crystallization RH using the Zdanovskii-Stokes-Robinson equation, and hysteresis is treated (Nenes et al., 1998; Li et al., 2011)	Size-resolved equilibrium with the mutual deliquescence and crystallization RH using the Zdanovskii-Stokes-Robinson equation, and hysteresis is treated (Jacobson et al., 1996)
In-cloud scavenging (Aerosol mechanism)	Scavenging via nucleation, Brownian diffusion, collection and autoconversion in both grid-scale and sub-grid clouds with a first-order removal rate (MADE/SORGAM, MOSAIC, MAM3 and MAM7) (Easter et al., 2004)	Scavenging of interstitial aerosol in the Aitken mode and nucleation scavenging of aerosol in the accumulation and coarse modes by the cloud droplets in both grid-scale and sub-grid clouds (AERO5, AERO6 and AERO7) (Binkowski and Roselle, 2004; Fahey et al., 2017)	Algorithm of rainout removal tendency by Giorgi and Chameides (1986)	Employing a scavenging coefficient approach based on relationships described by Seinfeld and Pandis (1998), only hydrophilic particles can be scavenged (Chen et al., 2017)	Size-resolved aerosol activation; nucleation scavenging and autoconversion for size-resolved cloud droplets (GATOR2012) (Jacobson, 2003)

Below-cloud scavenging (Aerosol mechanism)	Scavenged aerosols are instantly removed by interception and impaction but not resuspended by evaporating rain (MADE/SORGAM, MOSAIC, MAM3 and MAM7) (Slim, 1984; Easter et al., 2004)	All aqueous species are scavenged from the cloud top to the ground in both grid-scale and sub-grid clouds (AERO5, AERO6 and AERO7) (CMAQ User's Guide; Fahey et al., 2017)	Aerosol particles between sizes ranging from 0.5 to 1 μm radius are instantly removed with considering cloud fraction, and scavenged rate depends on aerosol and hydrometeor sizes (Slim, 1984; Gong et al., 2003)	Employing a scavenging coefficient approach based on relationships described by Seinfeld and Pandis (1998), considering accretion of in-cloud droplets particles into precipitation and impaction of ambient particles into precipitation	Discrete size-resolved coagulation between hydrometeors and aerosol particles (aerosol-liquid, aerosol-ice and aerosol-graupel) (GATOR2012) (Jacobson, 2003)
Sedimentation of aerosols (Aerosol mechanism)	Sedimentation with considering mass and number concentrations of aerosols at surface (MOSAIC) (Marelle et al., 2017)	Only considering gravitational sedimentation for aerosols (AERO5, AERO6 and AERO7)	Size-resolved sedimentation of aerosol particles above surface layer is computed with the settling velocity (CUACE) (Gong et al., 2003)	Using size-resolved sedimentation velocity to simulate sedimentation of aerosols (AERO5)	Sedimentation of size-resolved aerosols is computed from one model layer to layers below down to the surface, and the sedimentation velocities are calculated by two-step iterative method (GATOR2012) (Bear, 1976; Jacobson, 1997, 2003)

412 * GATOR2012 refers to either the aerosol or cloud microphysics scheme used in Jacobson (2012).
 413

414 Table 4 further lists various aspects with regards to how ARI and ACI being calculated in the
 415 five two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS,
 416 and GATOR-GCMOM) applied in Asia. Note that the information in this table was extracted from
 417 the latest released version of WRF-Chem (version 4.3.3) and WRF-CMAQ (based on WRF v4.3
 418 and CMAQ v5.3.3) as well as relevant references for GRAPES-CUACE (Wang et al., 2015), WRF-
 419 NAQPMS (Wang et al., 2014) and GATOR-GCMOM (Jacobson et al., 2010; 2012). These models
 420 all use the Mie theory to compute ARI effects but differ in representations of aerosol optical
 421 properties and radiation schemes. To simplify the calculation, aerosol species simulated by the
 422 chemistry module/model are put into different groups (Table 4) and the refractive indices of these
 423 groups are directly from the Optical Properties of Aerosols and Clouds (OPAC) database (Hess et
 424 al., 1998) in WRF-Chem and WRF-CMAQ (Table B6 in Appendix B). In WRF-Chem, the aerosol
 425 optical properties (AOD, extinction/scattering/absorption coefficient, single scattering albedo and
 426 asymmetry factor) are calculated in terms of four spectral intervals (listed in Table B6 in Appendix
 427 B) and then inter/extrapolated to 11 (14) SW intervals defined in the GODDARD (RRTMG) scheme.
 428 For SW and LW radiation in both WRF-CMAQ and WRF-Chem, these optical parameters are
 429 computed at each of corresponding spectral intervals in the RRTMG scheme. The aerosol optical
 430 property for LW radiation is considered only at 5 thermal windows (listed in Table B6) in WRF-
 431 CMAQ. No detailed information regarding how aerosol optical property and relevant parameters
 432 being calculated in GRAPES-CUACE and WRF-NAQPMS can be found from the relevant
 433 references.

434 With respect to ACI effects, the simulated aerosol characteristics (such as mass, size
 435 distribution and species) are utilized for the calculation of cloud droplet activation and aerosol
 436 resuspension based on the Köhler theory (Abdul-Razzak and Ghan, 2002) in several (one)
 437 microphysics schemes (scheme) in WRF-Chem (GRAPES-CUACE). GATOR-GCMOM is the first
 438 two-way coupled model adding IN activation processes including heterogeneous and homogeneous
 439 freezing (Jacobson et al., 2003). None of the other four two-way coupled models considers the IN
 440 formation processes (including immersion freezing, deposition freezing, contact freezing, and
 441 condensation freezing) but they have been included in some specific versions of WRF-Chem (Keita
 442 et al., 2020; Lee et al., 2020), which are not yet in the latest release version 4.3.3 of WRF-Chem.

443
 444 Table 4. Summary of relevant information regarding calculations of aerosol-radiation interactions (ARI) and aerosol-
 445 cloud interactions (ACI) in two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-
 446 NAQPMS and GATOR-GCMOM) applied in Asia.

Model	ARI		ACI				
	Aerosol species groups	Aerosol size distribution (Aerosol mechanism)	Mixing state [‡]	SW scheme (# of spectral intervals)	LW scheme (# of spectral intervals)	CCN (Microphysics scheme)	IN (Microphysics scheme)
WRF-Chem	1. Water 2. Dust 3. BC 4. OC 5. Sea-salt 6. Sulfate	1. Bulk (GOCART) 2. Modal (MADE/SORGAM, AERO5, MAM3 and MAM7) 3. Sectional (MOSAIC (4bins and 8 bins) and MADRID (8bins))	Internal mixing (Volume averaging, Core-shell, and Maxwell-Garnett)	1. Goddard (11) 2. RRTMG (14)	RRTMG (16)	Activation under a certain supersaturation in an air parcel based on Köhler theory (Morrison, Lin, Thompson, WSM 6/5/3 class and Milbrandt-Yau)	Ice heterogeneous nucleation of mineral dust aerosols in based on classical nucleation theory (Milbrandt-Yau and Morrison) [†]
WRF-CMAQ	1. Water 2. Water-soluble 3. BC 4. Insoluble 5. Sea-salt	Modal (AERO5, AERO6 and AERO7)	Internal mixing (Core-shell)	RRTMG (14)	RRTMG (16)	None	None
GRAPES-CUACE	1. Nitrate 2. Dust 3. BC 4. OC 5. Sea-salt 6. Sulfate 7. Ammonium	Sectional (CUACE (12 bins))	External mixing	Goddard (11)	Goddard (10)	Activation under a certain supersaturation in an air parcel based on Köhler theory (WSM 6-class)	None
WRF-NAQPMS	1. Nitrate 2. Dust 3. BC 4. OC 5. Sea-salt 6. Sulfate	Modal (AERO5)	External mixing	Goddard (11)	RRTM (16)	Activation under a certain supersaturation in an air parcel based on Köhler theory (Lin)	None

7. Ammonium
8. Other primary particles
GATOR-GCMOM
1. Water
2. Dust
3. BC
4. HCO_3^-
5. SOA
6. Sulfate
...
42. $\text{MgCO}_3(s)$

Sectional (GATOR2012*)
(17-30 bins)

Internal mixing
(Core-shell[†])

Toon* (318)

Toon* (376)

Activation under a certain supersaturation
in an air parcel based on Köhler theory
(GATOR2012*)

Ice heterogeneous and
homogeneous nucleation
(GATOR2012*)

447 [‡] Specific version of WRF-Chem, WRF-NAQPMS and GOTAR-GCMOM have the ability of simulating aerosol aging (Zhang et al.,
448 2014; Chen et al., 2017; Li et al., 2018; Jacobson, 2012).

449 [†] Some specific versions of WRF-Chem consider IN (Keita et al., 2020; Lee et al., 2020).

450 ^{*}The short- and long-wave radiation calculations in GATOR-GCMOM are based on the algorithm of Toon et al. (1989).

451 ^{*} GATOR2012 refers to either the aerosol or cloud microphysics scheme used in Jacobson (2012).

452

453 How accurately ARI and ACI are simulated also rely on the representation of aerosol
454 composition and size distribution in two-way coupled models. Table 5 presents the treatments of
455 aerosol compositions and size distributions in the five two-way coupled models applied in Asia. As
456 shown in Tables 4 and 5, GATOR-GCMOM considered more detailed aerosol species groups as
457 high as 42 kinds, and others coupled models different numbers of species groups (such as 6, 5, 7, 8
458 aerosol species groups in WRF-Chem, CMAQ, NAQPMS and CUACE, respectively). Three typical
459 representation approaches of size distribution (bulk, modal and sectional methods) are adopted by
460 the five two-way coupled models and WRF-Chem offers all the three approaches, but other models
461 only support one specific option. The Global Ozone Chemistry Aerosol Radiation and Transport
462 (GOCART) model (Ginoux et al., 2001) in WRF-Chem is the only one that is based on a
463 combination of bulk (for water, BC, OC, and sulfate aerosols) and sectional (for dust and sea salt
464 aerosols) approaches. The widely used modal and sectional approaches in five coupled models and
465 their detailed numerical settings of aerosol size distribution (namely, geometric diameter and
466 standard deviation for modal approach or bin ranges for sectional method) are listed in Table 5.
467 Regarding the modal method, same parameter values for Aitken and accumulation modes and
468 geometric diameters for coarse mode in the latest version of WRF-Chem (v4.3.3) and older version
469 of WRF-CMAQ (before v5.2) are set as default, except the standard deviations for coarse mode are
470 slightly different. In the official version of WRF-CMAQ released after v5.2, there are some
471 modifications to the default setting of geometric diameters in Aitken, accumulation and coarse
472 modes, from 0.01 to 0.015 μm , 0.07 to 0.08 μm and 1.0 to 0.6 μm , respectively. For the GRAPES-
473 CUACE model, the parameters of size distribution for certain aerosol species in the accumulation
474 mode were updated from its older version (Zhou et al., 2012) to newer one (Zhang et al., 2021).
475 With respect to the sectional approach, 4 or 8 (from 0.039 to 10 μm), 12 (from 0.005 to 20.48 μm)
476 and 14 (from 0.002 to 50 μm) particle size bins are defined in WRF-Chem, CUACE and GATOR-
477 GCMOM, respectively.

478

479 Table 5. Summary of numerical representations of aerosol size distribution and composition in two-way coupled
480 models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) applied in Asia.

Model	Aerosol mechanism	Modal approach						Compositions	Reference		
		Aitken		Accumulation		Coarse					
		Geometric diameters (μm)	Standard deviations (μm)	Geometric diameters (μm)	Standard deviations (μm)	Geometric diameters (μm)	Standard deviations (μm)				
WRF-Chem v4.3.3	MADE/SORGAM	0.010	1.7	0.07	2.0	1.0	2.5	Water, BC, OC, and sulfate, dust and sea salt	WRF-Chem codes ^{§§}		
WRF-Chem ^Δ	MAM3	0.013 (Sulfate and secondary OM)	1.6 (Sulfate and secondary OM)	0.068 (Sulfate, secondary OM, primary OM, BC, dust and sea salt)	1.8 (Sulfate, secondary OM, primary OM, BC, dust and sea salt)	2.0 (Sea salt), 1.0 (Dust)	1.8 (Sea salt and dust)	Sulfate, methane sulfonic acid (MSA), OM, BC, sea salt and dust	Easter et al. (2004) Liu et al. (2012)		
WRF-Chem ^Δ	MAM7	0.013 (Sulfate and secondary OM and BC)	1.6 (Sulfate, OM and BC)	0.068 (Primary OM) 0.2 (Sea salt) 0.11 (Dust)	1.8 (Sulfate and BC) 1.6 (Primary OM) 1.8 (Sea salt) 1.8 (Dust)	2.0 (Sea salt) 1.0 (Dust)	2.0 (Sea salt) 1.8 (Dust)	Sulfate, methane sulfonic acid (MSA), OM, BC, sea salt and dust	Easter et al. (2004) Liu et al. (2012)		
WRF-CMAQ	AERO5	0.010	1.7	0.07	2.0	1.0	2.2	Water, water-soluble BC,	CMAQ codes*		

(before CMAQ v5.2)								insoluble, sea salt	
WRF- CMAQ (after CMAQ v5.2)	AERO6 and AERO7	0.015	1.7	0.08	2.0	0.60	2.2	Water, water- soluble BC, insoluble, sea salt	CMAQ codes [†]
WRF- NAQPMS	AERO5	0.052	1.9	0.146	1.8	0.80	1.9	Nitrate, dust, BC, OC, sea- salt, sulfate, ammonium, other primary particles	Wang et al. (2014)
GRAPES- CUACE	CUACE	0.10 (BC and OC)	1.7 (BC and OC)	0.25 (Sulfate and nitrate)	1.7 (Sulfate and nitrate)	3.0 (Dust)	1.7 (Dust)	Nitrate, dust, BC, OC, sea- salt, sulfate, ammonium [*]	Zhou et al. (2012)
GRAPES- CUACE	CUACE	Unclear	Unclear	0.37 (BC and OC)	0.42 (BC and OC)	Unclear	Unclear	Nitrate, dust, BC, OC, sea- salt, sulfate, ammonium ⁺	Zhang et al. (2021)
WRF-Chem v4.3.3	MOSAIC	Sectional approach 0.039-0.156, 0.156-0.625, 0.625-2.5, 2.5-10.0 μm (4 bins) 0.039-0.078, 0.078-0.156, 0.156-0.312, 0.312-0.625, 0.625-1.25, 1.25-2.5, 2.5-5.0, 5.0-10.0 μm (8 bins)						Water, BC, OC, sulfate, dust and sea salt	WRF-Chem codes [⊗]
WRF-Chem ^Δ	MADRID	0.0216-10 μm (8 bins)						Water, BC, OC, and sulfate, dust and sea salt	Zhang et al. (2016)
WRF-Chem v4.3.3	GOCART	0.1-1.0, 1.0-1.8, 1.8-3.0, 3.0-6.0, 6.0-10.0 (5 bins for dust) 0.1-0.5, 0.5-1.5, 1.5-5.0, 5.0-10.0 (4 bins for sea salt)						Dust and sea salt	WRF-Chem codes [⊗]
GRAPES- CUACE	CUACE	0.005-0.01, 0.01-0.02, 0.02-0.04, 0.04-0.08, 0.08-0.16, 0.16-0.32, 0.32-0.64, 0.64-1.28, 1.28-2.56, 2.56-5.12, 5.12-10.24, 10.24-20.48 μm (12 bins)						Nitrate, dust, BC, OC, sea- salt, sulfate, ammonium	Zhou et al. (2012)
GATOR- GCMOM	GATOR2012	0.002-50 μm (14 bins)						42 species [‡]	Jacobson (2002, 2012)

481 [⊗]Official released version of WRF-Chem.

482 ^ΔSpecific version of WRF-Chem.

483 ^{*}https://github.com/USEPA/CMAQ/blob/5.1/models/CCTM/aero/aero6/AERO_DATA.F.

484 [†]https://github.com/USEPA/CMAQ/blob/5.2/CCTM/src/aero/aero6/AERO_DATA.F.

485 [‡]More detailed components were presented in the first column of Table 2.

486 ^{*}Initial size distribution is tri-modal log-normal distribution.

487

488 Not only the choice of methodologies for ARI and ACI calculations can impact simulation
489 results, but also the various aspects regarding the setup of modeling studies by applying two-way
490 coupled models. The extra/auxiliary information about model configuration, including horizontal
491 and vertical resolutions, aerosol and gas phase chemical mechanisms, PBL schemes, meteorological
492 and chemical initial conditions (ICs) and boundary conditions (BCs), anthropogenic and natural
493 emissions, were extracted from the 160 papers and presented in Table S4 of Supplement, which is
494 organized in the same order as Table 1.

495

496 For two-way coupled model applications in Asia, horizontal resolutions were set from a few to
497 several hundred kilometers, sometimes with nests, and vertical resolutions were from 15 to about
498 50-70 levels, with only one study performed at 100 levels for studying a fog case (Wang Z. L. et al.,
499 2018). Wang K. et al. (2018) evaluated the impacts of horizontal resolutions on simulation results
500 and found out surface meteorological variables were better modeled at finer resolution but no
501 significant improvements of ACI related meteorological variables and certain chemical species
502 between different grid resolutions. Through applying a single column model and then WRF-Chem
503 with ARI, Wang et al. (2019) unraveled that better representation of PBL structure and relevant
504 variables with finer vertical resolution from the surface to PBL top could reduce model biases
505 noticeably, but balancing between vertical resolution and computational resource was important as
506 well. Among the 160 applications of two-way coupled models in Asia, the frequently used aerosol
507 module and gas-phase chemistry mechanism in WRF-CMAQ (WRF-Chem) were AERO6
508 (MOSAIC and MADE/SOGARM) and CB05 (CBMZ and RADM2), respectively. For PBL

508 schemes, most studies selected YSU in WRF-Chem and ACM2 in WRF-CMAQ. Regarding to
509 meteorological ICs and BCs, the FNL data were the first choice, and outputs from the Model for
510 Ozone and Related Chemical Tracer (MOZART) were used to generate chemical ICs and BCs by
511 most researchers. Georgiou et al. (2018) also unraveled that boundary conditions of dust and O₃
512 played an important role in WRF-Chem simulations. The modeling applications in Asia utilized
513 global (EDGAR), regional (e.g., MIX, INTEX-B, and REAS), and national (e.g., MEIC and JEI-
514 DB) anthropogenic emission inventories. Natural emission sources, such as mineral dust (Shao,
515 2004), biomass burning (FINN (Wiedinmyer et al., 2011) and GFED (Guido et al., 2010)), biogenic
516 VOCs (MEGAN (Guenther et al., 2006)), and sea salt (Gong et al., 1997) were also considered. It
517 should be noted that only one paper by Gao et al. (2017) reported that the WRF-Chem model with
518 the Gridpoint Statistical Interpolation (GSI) data assimilation could improve the simulation
519 accuracy during a wintertime pollution period.

521 4 Overview of research focuses in Asia

522 4.1 Feedbacks of natural aerosols

523 4.1.1 Mineral dust aerosols

525 Due to the fact that dust storm events frequently occurred over Asia during 2000-2010, the
526 research community has focused on dust transportation and associated climatic effects (Choobari et
527 al., 2014; Gong et al., 2003; Lee et al., 2010; Yasunari and Yamazaki, 2009; Zhang et al., 2003a,
528 2003b). Also the detailed processes and physiochemical mechanisms of dust storms had been well
529 understood and reviewed in detail (Chen et al., 2017a; Huang et al., 2014; Shao and Dong, 2006;
530 Uno et al., 2006). To probe into the radiative feedbacks of dust aerosols in Asia, Wang et al. (2013,
531 2010) initiated modeling studies by a two-way coupled model, i.e., the GRAPES-CUAUE model,
532 to simulate direct radiative forcing (DRF) of dust, and revealed that the feedback effects of dust
533 aerosols could lead to decreasing of surface wind speeds and then suppress dust emissions. Further
534 modeling simulations by the same model (Wang and Niu, 2013) indicated that considering dust
535 radiative effects did not substantially improve the model performance of the air temperature at 2
536 meters above the surface (T2), even with assimilating data from in-situ and satellite observations
537 into the model. Subsequently, several similar studies based on another two-way coupled model
538 (WRF-Chem with The Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and
539 Transport scheme) were conducted to investigate dust radiative forcing (including shortwave
540 radiative forcing (SWRF) and longwave radiative forcing (LWRF)) and ARI effects of dust on
541 meteorological variables (PBLH, T2 and WS10) in different regions of Asia (Bran et al., 2018; Chen
542 et al., 2014; Jin et al., 2016a, 2015; Kumar et al., 2014; Liu L. et al., 2016; Su and Fung, 2018a,
543 2018b; Zhou et al., 2018). These studies demonstrated that dust aerosols could induced negative
544 radiative forcing (cooling effect) at top of atmosphere (TOA) as well as the surface (including both
545 Earth's and sea surfaces) and positive radiative forcing (warming effect) in the ATM (Bran et al.,
546 2018; Chen et al., 2014; Kumar et al., 2014; Li and Sokolik, 2018; Li M.M. et al., 2017a; Su and
547 Fung, 2018a; Wang et al., 2013). More thorough analyses of the radiative effects of dust in Asia (Li
548 and Sokolik, 2018; Wang et al., 2013) pointed out that dust aerosols played opposite roles in the
549 shortwave and longwave bands, so that the dust SWRF at TOA and the surface (cooling effects) as
550 well as in the ATM (warming effects) was offset partially by the dust LWRF (warming effects at
551 TOA and the surface but cooling effects in the ATM). It was noteworthy that adding more detailed
552 mineralogical composition into the dust emission for WRF-Chem could alter the dust SWRF at TOA
553 from cooling to warming and then lead to a positive net radiative forcing at TOA (Li and Sokolik,
554 2018). These different conclusions showed some degrees of uncertainties in the coupled model
555 simulations of dust aerosols' radiative forcing that need to be further investigated in the future.

556 Dust aerosols can act not only as water-insoluble cloud condensation nuclei (CCN) (Kumar et
557 al., 2009) but also as ice nuclei (IN) (Lohmann and Diehl, 2006) since they are referred to as ice
558 friendly (Thompson and Eidhammer, 2014). Therefore, activation and heterogeneous ice nucleation
559 parameterizations (INPs) with respect to dust aerosols were developed and incorporated into WRF-
560 Chem to explore ACI effects as well as both ARI and ACI effects of dust aerosols in Asia (Jin et al.,
561 2016a, 2015; Su and Fung, 2018a, 2018b; Wang K. et al., 2018; Zhang Y. et al., 2015b). During dust
562 storms, including the adsorption activation of dust particles played vital roles in the simulations of
563 ACI-related cloud properties and a 45 % of increase of cloud droplet number concentration (CDNC),
564 comparing to a simpler aerosols activation scheme in WRF-Chem (Wang K. et al., 2018). More
565 sophisticated INPs implemented in WRF-Chem that taking dust particles into account as IN resulted

566 in substantial modifications of cloud and ice properties as well as surface meteorological variables
567 and air pollutant concentrations in model simulations (Su and Fung, 2018b; Zhang Y. et al., 2015b).
568 Zhang Y. et al. (2015b) delineated that dust aerosols acting either as CCN or IN made model results
569 rather different regarding radiation, T2, precipitation, and number concentrations of cloud water and
570 ice. Su and Fu (2018b) described that the ACI effects of dust had less impacts on the radiative forcing
571 than its ARI effects and dust particles could promote (demote) ice (liquid) clouds in mid-upper (low-
572 mid) troposphere over EA. With turning on both ARI and ACI effects of dust, less low-level clouds
573 and more mid- and high-level clouds were detected that contributed to cooling at the Earth's surface
574 and in the lower atmosphere and warming in the mid-upper troposphere (Su and Fung, 2018b).
575 Mineral dust particles transported by the westerly and southwesterly winds from the Middle East
576 (ME) affected the radiative forcing at TOA and the Earth's surface and in the ATM by the dust-
577 induced ARI and ACI in the Arabian Sea and the India subcontinent, and subsequently changed the
578 circulation patterns, cloud properties, and characteristics related to the India summer monsoon (ISM;
579 Jin et al., 2015, 2016b). Moreover, the effects of dust on precipitation are not only complex but also
580 highly uncertain, evidencing from several modeling investigations targeting a variety of areas in
581 Asia (Jin et al., 2016a, 2016b, 2015; Su and Fung, 2018b; Zhang Y. et al., 2015b). Less precipitation
582 from model simulations including dust effects was found at EA and dust particles acting mainly as
583 CCN or IN influenced precipitation in a rather different way (Zhang Y. et al., 2015b). A positive
584 response of ISM rainfall to dust particles from the ME was reported by Jin et al. (2015) and less
585 affected by dust storms from the local sources and NWC (Jin et al., 2016a). Jin et al. (2016b) further
586 elucidated that the impacts of ME dust on ISM rainfall were highly sensitive to the imaginary
587 refractive index of dust setting in the model, so that accurate simulations of the dust-rainfall
588 interaction depended on more precise representation of radiative absorptions of dust in two-way
589 coupled models. About 20 % of increase or decrease in rainfall due to the dust effects were detected
590 in different areas over EA from the WRF-Chem simulations (Su and Fung, 2018b). However, it
591 should be mentioned that a few studies that targeting DRF of dust in Asia based on WRF-Chem
592 simulations but without enabling aerosol-radiation feedbacks (Ashrafi et al., 2017; Chen S. et al.,
593 2017b; Tang et al., 2018) were not included in this paper.

594 Along with the modeling research on the effects of dust aerosols on meteorology, their impacts
595 on air quality in Asia were explored using two-way coupled models (Chen et al., 2014; Kumar et
596 al., 2014; Li and Sokolik, 2018; Li M. M. et al., 2017a; Wang et al., 2013). Many early modeling
597 research work involving two-way coupled models with dust only looked into the ARI or direct
598 radiative effects of dust particles, which are described as follows. Taking a spring-time dust storm
599 from the Thar Desert into consideration in WRF-Chem, the modeled aerosol optical depth (AOD)
600 and Angstrom exponent (as indicators of aerosol optical properties and unique proxies of the surface
601 particulate matter pollution) demonstrated that turning on the ARI effects of dust could reduce biases
602 in their simulations, but were underestimated in North India (Kumar et al., 2014). Wang et al. (2013)
603 pointed out that in EA, including the longwave radiative effects of dust in the GRAPES-
604 CUACE/dust model lowered relative errors of the modeled AOD by 15 %, as compared to
605 simulations that only considering shortwave effects of dust. Comparisons against both satellite and
606 in situ observations depicted that the WRF-Chem model was able to capture the general
607 spatiotemporal variations of the optical properties and size distribution of dust particles over the
608 main dust sources in EA, such as the Taklimakan Desert and Gobi Desert, but overestimated AOD
609 during summer and fall and also exhibited positive (negative) biases in the fine (coarse) mode of
610 dust particles (Chen et al., 2014). Besides the ARI effects of dust, the heterogeneous chemistry on
611 dust particles' surface added in WRF-Chem was accounted for 80 % of the net reductions of O_3 ,
612 NO_2 , NO_3 , N_2O_5 , HNO_3 , $\cdot OH$, $HO_2\cdot$ and H_2O_2 when a springtime dust storm striking the Nanjing
613 megacity of EC (Li M. M. et al., 2017a). In CA, AOD was overestimated by WRF-Chem model but
614 its simulation was improved when more detailed mineral components of dust particles were
615 incorporated in the model (Li and Sokolik, 2018). Later on, more investigations started to focus on
616 both ARI and ACI effects of dust aerosols. With consideration of ARI as well as both ARI and ACI
617 of dust particles from the ME, during the ISM period, the WRF-Chem model reproduced AOD's
618 spatial distributions but underpredicted (overpredicted) AOD over the Arabian Sea (the Arabian
619 Peninsula) comparing with satellite observations and AOD reanalysis data (Jin et al., 2016a, 2016b,
620 2015). In EA, Wang K. et al. (2018) demonstrated that including both ARI and ACI effects of dust
621 in WRF-Chem caused lower O_3 concentrations and by incorporating INPs, the WRF-Chem model
622 well simulated the surface PM_{10} concentrations (Su and Fung, 2018a) with reduced (elevated)

623 surface concentrations of OH, O₃, SO₄²⁻, and PM_{2.5} (CO, NO₂, and SO₂) (Zhang Y. et al., 2015b). It
624 is worth noting that how to partition dust particles into fine mode and coarse mode or initialize their
625 size distribution in coupled models can affect simulations in many ways and requires more detailed
626 measurements at the source areas and further modeling studies.

627 4.1.2 Wildfire, sea salt and volcanic ash

629 In the Maritime SEA region, peat and forest fire triggered by El Niño induced drought
630 conditions released huge amount of smoke particles, which promoted dire air pollution problems in
631 the downstream areas, and their ARI effects simulated by WRF-Chem enhanced radiative forcing at
632 the TOA and the atmospheric stability (Ge et al., 2014). Ge et al. (2014) also pointed out the ARI
633 effects of these fires impaired (intensified) sea breeze at daytime (land breeze at nighttime) over this
634 region so that their impacts on cloud cover could be positive or negative in different areas and time
635 period (day or night). Sea salt and volcanic ash are also important natural aerosols for regions near
636 seashores and active volcanoes and surrounding areas but modeling studies of their ARI and ACI
637 effects are relatively scarce in Asia. Based on WRF-Chem simulations, Kedia et al. (2019a)
638 demonstrated that the feedbacks of sea salt aerosols impacted convective and nonconvective
639 precipitation rather variously in different areas of the India subcontinent. Jiang et al. (2019a, 2019b)
640 also used WRF-Chem with/without sea-salt emissions to evaluate the effects of sea salt on rainfall
641 in Guangdong Province of China, but unfortunately, no feedbacks were considered in the
642 simulations. So far there is no investigation targeting aerosol effects of volcanic ash from volcano
643 eruptions in Asia using coupled models.

644 4.2 Feedbacks of anthropogenic aerosols

645 Atmospheric pollutants from anthropogenic sources are the leading causes of heavy pollution
646 events occurring in Asia due to the acceleration of urbanization, industrialization, and population
647 growth in recent decades, particularly in China and India, and their ARI or/and ACI effects on
648 meteorology and air quality had been quantitatively examined using two-way coupled models
649 (Archer-Nicholls et al., 2019; Bharali et al., 2019; Gao M. et al., 2016b; Kumar et al., 2012a, 2012b;
650 Li and Liao, 2014; Wang J. et al., 2014; Wang Z. et al., 2018; Zhang B. et al., 2015; Yao et al., 2017;
651 Zhong et al., 2016). These modeling research work had been primarily focused on the ARI or/and
652 ACI effects of anthropogenic aerosols, their specific chemical components (especially the light-
653 absorbing aerosols, i.e., BC and brown carbon (BrC)) and aerosols originated from different sources.
654 The major findings are outlined as follows, with respect to the effects of anthropogenic aerosol
655 feedbacks on meteorology and air quality.

656 Concerning the meteorological responses, most papers treated anthropogenic aerosols as a
657 whole to explore their effects on meteorological variables based on coupled model simulations with
658 enabling ARI or/and ACI in WRF-Chem, WRF-CMAQ, WRF-CMAQ, GRAPES-CUACE and
659 WRF-NAQPMS (Bai et al., 2020; Gao M. et al., 2016b; Kumar et al., 2012a; Nguyen et al., 2019a,
660 2019b; Wang H. et al., 2015; J. Wang et al., 2014; Wang Z. F. et al., 2014; Zhang B. et al., 2015;
661 Zhang et al., 2018; Zhao et al., 2017). Generally, the main ARI effects of anthropogenic aerosols
662 resulted in decreases of SWRF, T2 and WS10, and PBLH, as well as increases of surface relative
663 humidity (RH2) and temperature in the ATM, which further suppressed PBL development (Gao Y.
664 et al., 2015; Li M. M. et al., 2017b; Nguyen et al., 2019a, 2019b; Xing et al., 2015c; Zhang et al.,
665 2018). Wang H. et al. (2015) utilized GRAPES-CUACE with ARI to study a summer haze case in
666 the NCP area and discovered that the ARI effects made the subtropical high less intense (-14 hPa)
667 to help pollutants in the area to dissipate. In Asia, ACI effects of anthropogenic aerosols on cloud
668 properties and precipitation are relatively complex. On the one hand, anthropogenic aerosols, that
669 being activated as CCN, enhanced CDNC and LWP and then slowed down the precipitation onset,
670 but their impacts on precipitation amounts varied in different seasons and areas in China (Zhao et
671 al., 2017). Targeting a summertime rainstorm in the middle reaches of the Yangtze River (MRYR)
672 in China, sensitivity studies using WRF-Chem unveiled that CDNC, cloud water contents, and
673 precipitation decreased (increased) with low (high) anthropogenic emission scenarios due to the
674 ACI effects and these variations tended to depend on atmospheric humidity (Bai et al., 2020). The
675 modeling investigations with WRF-Chem aiming at the ISM (Kedia et al., 2019) and a disastrous
676 flood event in Southwest China (SWC) (Fan et al., 2015) pointed out that the simulated convective
677 process was suppressed and convective (nonconvective) precipitation was inhibited (enhanced) by

679 the ARI and ACI effects of accumulated anthropogenic aerosols, but these effects could invigorate
680 convection and rainfall in the downwind mountainous area at nighttime (Fan et al., 2015). On the
681 other hand, how anthropogenic aerosols act in the ice nucleation processes is still open to question
682 (Zhao et al., 2019) and these processes need to be represented accurately in two-way coupled models,
683 however until now no study had been performed to simulate the ACI effects of anthropogenic
684 aerosol serving as IN in Asia using two-way coupled models. Therefore, in Asia, further
685 investigations are needed that targeting cloud or/and ice processes involving anthropogenic aerosols
686 (including their size, composition, and mixing state) in two-way coupled models. Meanwhile,
687 several studies not only discussed aerosol feedbacks but also focused on the additional effects of
688 topography or urban heat island on meteorology (Wang D. et al., 2019; Zhong et al., 2017, 2015).
689 Utilizing the GATOR-GCMOM model at global and local scales, Jacobson et al. (2015, 2019)
690 explored the impacts of landuse changes due to the unprecedented expansions of megacities, such
691 as Beijing and New Delhi in Asia, from 2000 to 2009 on meteorology and air quality.

692 Hitherto there were several attempts to ascertain the effects of different chemical components
693 of anthropogenic aerosols on meteorology in Asia (Archer-Nicholls et al., 2019; Ding et al., 2019;
694 Ding et al., 2016; Gao J. et al., 2018; Huang et al., 2015; Wang Z. et al., 2018). First of all, Asia is
695 the region in the world with the highest BC emissions due to burning of large amount of fossil fuels
696 and biomass and this has increasingly attracted many researchers to probe into the ARI or/and ACI
697 effects of BC (Boucher et al., 2013). As the most important absorbing aerosol, BC induced the
698 largest positive, positive and negative mean DRF at the TOA, in the ATM, and at the surface,
699 respectively, over China during 2006 (Huang et al., 2015). Ding et al. (2016) and Wang Z. et al.
700 (2018) further applied WRF-Chem with feedbacks to investigate how aerosol-PBL interactions
701 involving BC suppressed the PBL development, which deteriorated air quality in Chinese cities and
702 was described as “dome effect” (namely BC warms the atmosphere and cools the surface, suppresses
703 the PBL development and eventually results in more accumulation of pollutants). This “dome effect”
704 of BC promoted the advection-radiation fog and fog-haze formation in the YRD area through
705 altering the land-sea circulation pattern and increasing the moisture level (Ding et al., 2019). Gao J.
706 et al. (2018) also pointed out BC in the ATM modified the vertical profiles of heating rate and
707 equivalent potential temperature in Nanjing, China. In India, the ARI effects of BC enhanced
708 convective activities, meridional flows, and rainfall in North-East India during the pre-monsoon
709 season but could either enhance or suppress precipitation during the monsoon season in different
710 parts of the India subcontinent (Soni et al., 2018). Moreover, the ARI effects of BC on surface
711 meteorological variables were larger than its ACI effects in EC (Archer-Nicholls et al., 2019; Ding
712 et al., 2019). Besides BC, the BrC portion of organic aerosols (OA) emitted from agriculture residue
713 burning (ARB) were included in WRF-Chem with the parameterization scheme suggested by Saleh
714 et al. (2014) and the model simulations in EC revealed that at the TOA, the net DRF of OA was -
715 $0.22 \text{ W} \cdot \text{m}^{-2}$ (absorption and scattering DRF were $+0.21 \text{ W} \cdot \text{m}^{-2}$ and $-0.43 \text{ W} \cdot \text{m}^{-2}$ respectively), but
716 the BC's DRF was still the highest ($+0.79 \text{ W} \cdot \text{m}^{-2}$) (Yao et al., 2017). As mentioned above, it is
717 obvious that ARI and ACI effects of different aerosol components are substantially distinctive, and
718 many other aerosol compositions (e.g., sulfate, nitrate and ammonium) besides BC and BrC should
719 be taken into considerations in future modeling studies in Asia.

720 ARB is a common practice in many Asian countries after harvesting and before planting and
721 can deteriorate air quality quickly as one of the most important sources of anthropogenic aerosols,
722 so that it has been attracting much attention among the public and scientists worldwide (Chen J. et
723 al., 2017; Hodshire et al., 2019; Koch and Del Genio, 2010; Reid et al., 2005; Yan et al., 2018).
724 Recently, the effects of ARB aerosols on meteorology had widely been explored using the two-way
725 coupled model (WRF-Chem) in many Asian countries and regions, such as EC (Huang et al., 2016;
726 Li M. et al., 2018; Wu et al., 2017; Yao et al., 2017), South China (SC) (Huang et al., 2019), and
727 South Asia (SA) (Singh et al., 2020). In general, when ARB occurred, the WRF-Chem simulations
728 from all the studies showed that the changes in radiative forcing induced by ARB aerosols were
729 greater than by those from other anthropogenic sources, especially in the ATM. Also all the modeling
730 studies indicated that ARB aerosols reduced (increased) radiative forcing at the surface (in the ATM),
731 cooled (warmed) the surface (the atmosphere), and increased (decreased) atmospheric stability
732 (PBLH). Furthermore, the WRF-Chem simulations with ARI demonstrated that light-absorbing
733 carbonaceous aerosols (CAs) from ARB caused daytime (nighttime) precipitation decreased
734 (increased) over Nanjing in EC during a post-harvest ARB event (Huang et al., 2016). Yao et al.
735 (2017) pointed out their WRF-Chem simulations in EC exhibited larger DRE induced by BC from

ARB at the TOA than previous studies. Lately, several modeling studies using WRF-Chem had targeted the effects of ARI and both ARI and ACI due to ARB aerosols from countries in the Indochina, SEA, and SA regions during the planting and harvesting time (Dong et al., 2019; Huang et al., 2019; Singh et al., 2020; Zhou et al., 2018). Zhou et al. (2018) investigated how ARB aerosols from SEA mixed with mineral dust and other anthropogenic aerosols while being lifted to the mid-low troposphere over the source region and transported to the YRD area and then affected meteorology and air quality there. The influences of ARI and ACI caused by ARB aerosols from Indochina were contrary, namely, the ARI (ACI) effects made the atmosphere over SC warmer (cooler) and drier (wetter), and the ARI effects hindered cloud formation and suppressed precipitation there (Huang et al., 2019). Dong et al. (2019) found the warming ARI effects of ARB aerosols were smaller over the source region (i.e., SEA) than the downwind region (i.e., SC) with cloudier conditions. Annual simulations regarding the ARI effects of ARB aerosols from SA (especially Myanmar and Punjab) indicated that CAs released by ARB reduced the radiative forcing at the TOA but did not change the precipitation processes much when only the ARI effects were considered in WRF-Chem (Singh et al., 2020).

Besides ARB, to our best knowledge, there were only a few research work quantitatively assessing the effects of anthropogenic aerosols from different emission sources on meteorology using WRF-Chem. Gao M. et al. (2018b) evaluated the responses of radiative forcing in China and India to aerosols from five emission sectors (power, industry, residential, BB, and transportation), and found that the power (residential) sector was the dominate contributor to the negative (positive) DRF at the TOA over both countries due to high emissions of sulfate and nitrate precursors (BC) and the total sectoral contributions were in the order of power > residential > industry > BB > transportation (power > residential > transportation > industry > BB) for China (India) during 2013. To pinpoint the ARI and ACI effects, Archer-Nicholls et al. (2019) reported that during January 2014, the aerosols from the residential emission sector induced larger SWRF ($+1.04 \text{ W}\cdot\text{m}^{-2}$) than LWRF ($+0.18 \text{ W}\cdot\text{m}^{-2}$) at the TOA and their DRF ($+0.79 \text{ W}\cdot\text{m}^{-2}$) was the largest, followed by their semidirect effects ($+0.54 \text{ W}\cdot\text{m}^{-2}$) and indirect effects ($-0.29 \text{ W}\cdot\text{m}^{-2}$) over EC. This study further emphasized a realistic ratio of BC to total carbon from the residential emission was critical for accurate simulations of the ARI and ACI effects with two-way coupled models.

In terms of anthropogenic aerosol effects on air quality, the responses of $\text{PM}_{2.5}$ had been widely investigated (Chen et al., 2019a; Gao J. et al., 2018; Gao M. et al., 2016b; Gao Y. et al., 2015; Nguyen et al., 2019a, 2019b; Wang H. et al., 2015; Wang J. et al., 2014; Wang Z. F. et al., 2014; Wu et al., 2019a; Zhang B. et al., 2015; Zhang et al., 2018; Zhao et al., 2017) but less studies explored the responses of O_3 and other species (Kumar et al., 2012b; Li J. et al., 2018; Nguyen et al., 2019a, 2019b; Xing et al., 2017; Zhang B. et al., 2015). As summarized by Wu et al. (2019a) in their Table 1, observations and model simulations with WRF-Chem, WRF-CMAQ, WRF-CMAQ, GRAPES-CUACE, and WRF-NAQPMs all pointed out that the ARI effects promoted higher $\text{PM}_{2.5}$ concentrations in China (Chen et al., 2019a; Gao M. et al., 2016b; Gao Y. et al., 2015; Wang H. et al., 2015; Wang J. et al., 2014; Wang Z.F. et al., 2014; Zhang B. et al., 2015; Zhang et al., 2018) and this was also true in other areas of Asia (e.g., India, EA, Continental SEA) (Gao M. et al., 2018b; Nguyen et al., 2019a, 2019b) during different seasons. At the same time, all the modeling investigations revealed that the positive aerosol-meteorology feedbacks could further exacerbate pollution problems during heavy haze episodes. Based on WRF-Chem simulations, the ACI effects on $\text{PM}_{2.5}$ was negligible comparing to the ARI effects over EC (Zhang B. et al., 2015) but was subject to a certain degree of uncertainty if no consideration of the ACI effects induced by cumulus clouds in the model (Gao Y. et al., 2015). Annual WRF-Chem simulations for 2014 by Zhang et al. (2018) indicated that even though the ARI effects had bigger impacts on $\text{PM}_{2.5}$ during wintertime than the ACI effects, the ARI and ACI impacts on $\text{PM}_{2.5}$ were similar during other seasons and the increase of $\text{PM}_{2.5}$ due to the ACI effects was more noticeable in wet season than dry season. Using the process analysis method to distinguish the contributions of different physical and chemical processes to $\text{PM}_{2.5}$ over the NCP area, Chen et al. (2019a) applied WRF-Chem with ARI and ACI and found that besides local emissions and regional transport processes, vertical mixing contributed the most to the accumulation and dispersion of $\text{PM}_{2.5}$, comparing to chemistry and advection, and the ARI effects changed the vertical mixing contribution to daily $\text{PM}_{2.5}$ variation from negative to positive. Regarding surface O_3 concentrations, all the two-way coupled models with ARI, ACI, and both ARI and ACI predicted reduced photolysis rate and O_3 concentrations under heavy pollution conditions, through the radiation attenuation induced by aerosols and clouds. Further analyses

793 indicated that the ARI effects impacted O_3 positively through reducing vertical dispersions (WRF-
794 CMAQ, Xing et al., 2017), reduced O_3 more during wintertime than summertime in EC (WRF-
795 NAQPMS, Li J. et al., 2018), and suppressed (enhanced) O_3 in dry (wet) season in continental SEA
796 (WRF-CMAQ, Nguyen et al., 2019b). Xing et al. (2017) applied the process analysis method in
797 WRF-CMAQ with ARI and revealed that the impacts of ARI on the contributions of atmospheric
798 dynamics and photochemistry processes to O_3 over China varied in winter and summer months and
799 ARI induced largest changes in photochemistry (dry deposition) of surface O_3 at noon time in
800 January (July). The process analysis in WRF-Chem with ARI and ACI identified that the vertical
801 mixing process played the most important role among the other physical and chemical processes
802 (advection and photochemistry) in surface O_3 growth during 10-14 local time in Nanjing, China
803 (Gao J. et al., 2018). ARI and ACI not only affected $PM_{2.5}$ and O_3 , but also other chemical species.
804 For instance, CO and SO_2 increased due to ARI and ACI over EC (Zhang B. et al., 2015), ARI
805 caused midday (daily average) OH increased (decreased) in July (January) over China (Xing et al.,
806 2017), SO_2 , NO_2 , BC, SO_4^{2-} , NO_3^- were enhanced but OH was reduced over China by ACI (Zhao
807 et al., 2017), and ARI impacted SO_2 and NO_2 positively over EA (Nguyen et al., 2019a). Wu et al.
808 (2019b) further analyzed how the aerosol liquid water involved in ARI and chemical processes (i.e.,
809 photochemistry and heterogeneous reactions) and influenced radiation and $PM_{2.5}$ (esp. secondary
810 aerosols) over NCP during an intense haze event. Moreover, evaluations and sensitivity studies
811 indicated that turning on aerosol feedbacks could improve the model performance for surface $PM_{2.5}$,
812 particularly during severe haze episodes (Li J. et al., 2018; Wang H. et al., 2018; Zhang B. et al.,
813 2015; Zhang et al., 2018).

814 With reference to the feedback effects of anthropogenic aerosol compositions on air quality,
815 most modeling research work with WRF-Chem had focused on the ARI and ACI effects of BC and
816 BrC, especially the “dome effect” that prompted the accumulation of pollutants (aerosols and O_3)
817 near surface and in PBL (Ding et al., 2016; Ding et al., 2019; Gao J. et al., 2018; Li and Liao, 2014;
818 Wang Z. et al., 2018). At the same time, the ARI effects of BC undermined the low-level wind
819 convergence and then led to decrease of aerosols (sulfate and nitrate) and O_3 (Li and Liao, 2014).
820 With the process analysis methodology in WRF-Chem, Gao J. et al. (2018) indicated that comparing
821 to simulations without BC, the BC and PBL interaction slowed the O_3 growth from late morning to
822 early afternoon somewhat before O_3 reaching its maximum value at noon due to less vertical mixing
823 in PBL.

824 Studies on the feedback effects of aerosols from different emission sectors on air quality were
825 relatively limited and mainly involved with ARB emissions and assessments of emission controls
826 during certain major air pollution events. Jena et al. (2015) applied WRF-Chem with aerosol
827 feedbacks and investigated O_3 and its precursors in SA due to regional ARB. Based on WRF-Chem
828 simulations with enabling ARI and ACI, Wu et al. (2017) denoted that aerosols emitted from ARB
829 could be mixed or/and coated with urban aerosols while being transported to cities and contributed
830 to heavy air pollution events there, such as in Nanjing, China. The ARI effects induced by ARB
831 aerosols on O_3 and NO_2 concentrations (-1 % and 2 %, respectively) were small compared to the
832 contribution of precursors emitted from ARB to O_3 chemistry (40 %) in the ARB zone (Li M. et al.,
833 2018). Pollutants emitted from natural and anthropogenic BB over Indochina affected pollution
834 levels over SC and their ACI effects removed aerosols more efficiently than the ARI effects that
835 could make BB aerosols last longer in the ATM (Huang et al., 2019). Gao et al. (2017a) and Zhou
836 et al. (2019) both utilized WRF-Chem to evaluate what role the ARI effects played when dramatic
837 emission reductions implemented during the week of Asia Pacific Economic Cooperation Summit
838 and concluded that the ARI reduction induced by decreased emission led to 6.7-10.9 % decline in
839 $PM_{2.5}$ concentrations in Beijing.

840 841 4.3 Human health effects

842 Poor air quality posts risks to human health (Brunekreef and Holgate, 2002; Manisalidis et al.,
843 2020), therefore, in the past several decades, air quality models had been used in epidemiology
844 related research to establish quantitative relationships between concentrations of various pollutants
845 and burden of disease (including mortality or/and morbidity) as well as associated economic loss
846 (Conti et al., 2017). In Asia, there were several studies that applied coupled air quality models with
847 feedbacks to assess human health effects of air pollutants under historical and future scenarios
848 (Conibear et al., 2018a, 2018b; Gao et al., 2017b; Gao M. et al., 2015; Ghude et al., 2016; Hong et
849 al., 2019; Wang et al., 2017; Xing et al., 2016; Zhong et al., 2019). By applying WRF-Chem with

850 ARI and ACI, M. Gao et al. (2015) estimated the health and financial impacts induced by an intense
851 air pollution event happened in the NCP area during January, 2013 and concluded that the mortality,
852 morbidity, and financial loss over Beijing area were 690, 69070, and 253.8 million US\$,
853 respectively. Targeting the same case, Gao M. et al. (2017b) pointed out that turning on the data
854 assimilation of surface PM_{2.5} observations in WRF-Chem not only improved model simulations but
855 also made the premature death numbers increased by 2 % in the NCP area, comparing to simulations
856 without the PM_{2.5} data assimilation. In India, WRF-Chem simulations with aerosol feedbacks and
857 updated population data revealed that the premature (COPD related) deaths caused by PM_{2.5} (O₃)
858 were 570,000 (12,000), resulting in shortened life expectancy (3.4±1.1 years) and financial
859 expenses (640 million US\$) during 2011 (Ghude et al., 2016). Based on WRF-CMAQ simulations
860 with ARI for 21 years (1990-2010), Xing et al. (2016) pointed out that in EA the population-
861 weighted PM_{2.5} induced mortality had an upward trend from 1990 (+3187) to 2010 (+3548) and the
862 mean mortality caused by ARI-enhanced PM_{2.5} was 3.68 times more than that decreased by ARI-
863 reduced temperature. The same 21 year simulations also showed that from 1990 to 2010, the PM_{2.5}
864 related mortalities in EA and SA rose by 21 % and 85 %, respectively, while they declined in
865 Europe and high-income North America by 67 % and 58 %, respectively (Wang et al., 2017).
866 Conibear et al. (2018a) applied WRF-Chem with ARI to study how different emission sectors
867 affected human health in India and demonstrated that the residential energy use sector played the
868 most critical role among other sectors and could cause 511,000 premature deaths in 2014.
869 Furthermore, Conibear et al. (2018b) investigated future PM_{2.5} pollution levels as well as health
870 impacts in India under different emission scenarios (business as usual and two emission control
871 pathways) and deduced that the burden of disease driven by PM_{2.5} and population factors (growth
872 and aging) in 2050 increased by 75 % under the business as usual scenario but decreased by 9 %
873 and 91 % under the International Energy Agencies New Policy Scenario and Clean Air Scenario,
874 respectively, comparing with that in 2015. The sensitivity study using WRF-Chem with ARI under
875 a variety of emission scenarios, population projections, and concentration-response functions
876 (CRFs) for the years of 2008 and 2050 demonstrated that CRFs (future emission projections) were
877 the main sources of uncertainty in the total mortality estimations related to PM_{2.5} (O₃) in China
878 (Zhong et al., 2019). Applying a suite of models, including WRF-CMAQ with ARI, climate and
879 epidemiology, Hong et al. (2019) inferred that under Representative Concentration Pathway 4.5,
880 the future mortalities could be 12100 and 8900 per year in China led by PM_{2.5} and O₃, respectively,
881 and the climate-driven weather extremes could add 39 % and 6 % to future mortalities due to stable
882 atmosphere and heat waves, respectively. Ten Hoeve and Jacobson (2012) applied GATOR-
883 GCMOM and a human exposure model to estimate the local and worldwide health effects induced
884 by the 2011 Fukushima nuclear accident and a hypothetical one in California of US.
885

886 **5 Effects of aerosol feedbacks on model performance**

887 Even though there are a certain number of research papers using two-way coupled models to
888 quantify the effects of aerosol feedbacks on regional meteorology and air quality in Asia, model
889 performances impacted by considering aerosol effects varied to some extent. This section provides
890 a summary of model performance by presenting the SI of meteorology and air quality variables as
891 shown in Table S2. These SI were collected from the selected papers that supplying these indices
892 and being defined as papers with SI (PSI) (listed in Tables B2-B3 of Appendix B). As
893 aforementioned in Section 3, investigations of ACI effects were very limited and no former studies
894 simultaneously exploring aerosol feedbacks with and without both ARI and ACI turned on. Here,
895 we only compared the SI for simulations with and without ARI in the same study, as summarized in
896 Appendix Tables B4-B5. It should be pointed out that all the reported evaluation results either from
897 individual model or inter-model comparison studies were extracted and put into the Table S2.
898

899 **5.1 Model performance for meteorology variables**

900 With certain emissions, accurate simulations of meteorological elements are critical to air
901 quality modeling and prediction (Appel et al., 2017; Bauer et al., 2015; Saylor et al., 2019; Seaman,
902 2000). Targeting meteorological variables, we summarized their SI and further analyzed the
903 variations of SI on different simulated time scales and among multiple models.
904

905 **5.1.1 Overall performance**

906 Figure 3 shows the compiled statistical indicators (correlation coefficient (R) is in black, and
907 mean bias (MB) and root mean square error (RMSE) are in blue) of T2 (°C), RH2(%) and specific
908 humidity (SH2, g·kg⁻¹) at 2 meters, and WS10 (m·s⁻¹) from PSI (a-d), and simulations with and
909 without ARI (marked as ARI and NO-ARI in e-h). In this figure and following figures, NP and NS
910 are number of publications and samples with SI, respectively and summed up in Appendix Table
911 B2. In these two tables, we also listed the NS of positive (red upward arrow) and negative (blue
912 downward arrow) biases for the meteorological and air quality variables in parentheses in the MB
913 column. Note that NS in Fig. 3e-h and Appendix Table B4 counted the samples of SI provided by
914 the simulations simultaneously with and without ARI. Also, the 5th, 25th, 75th and 95th percentiles
915 of SI are illustrated in box-and-whisker plots, and the dashed line in the box is the mean value (not
916 median) and the circles are outliers.

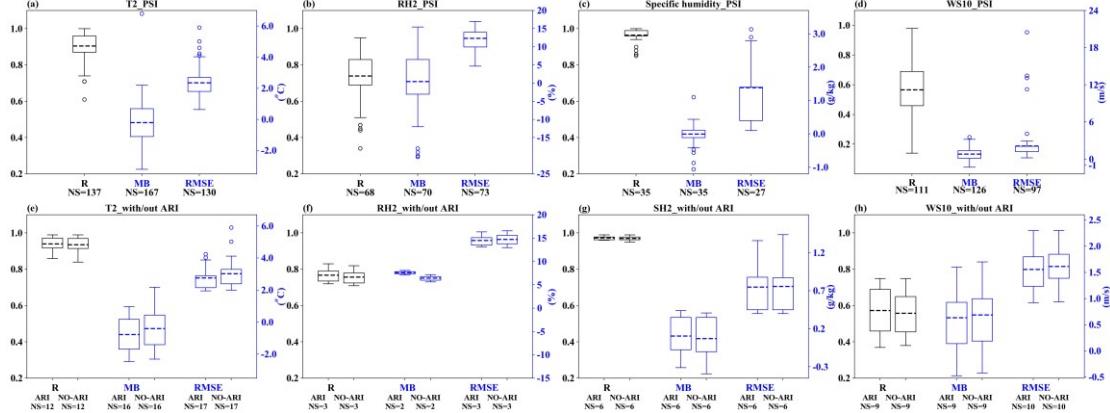
917 The evaluations for T2 (Fig. 3a) from PSI revealed that in Asia coupled models performed
918 rather well for temperature (mean R = 0.90) with RMSE ranging from 0.64 to 5.90 °C, but 60 % of
919 samples showed the tendency towards temperature underestimations (mean value of MB = -0.20 °C)
920 with the largest average MB (-0.31 °C) occurring during winter months (70 samples). The
921 underestimations of temperature had been reported not only from modeling studies by using WRF
922 or coupled models, but also in Asia, Europe and North America (Brunner et al., 2015; Gao et al.,
923 2019; García-Díez et al., 2013; Makar et al., 2015a; Yahya et al., 2015). The WRF simulations in
924 China (Gao et al., 2019) and US (EPA, 2018) also showed wintertime cold biases of T2 but in Europe
925 warm biases were reported (García-Díez et al., 2013). This temperature bias was probably related
926 to the impacts of model resolutions (Kuik et al., 2016), urban canopies (Liao et al., 2014) and PBL
927 schemes (Hu et al., 2013). With the ARI turned on in the coupled models, modeled temperatures
928 (limited papers with 12 samples) were improved somewhat and the mean correlation coefficient
929 increased from 0.93 to 0.95 and RMSE decreased slightly (Fig. 3e), but average MB of temperature
930 was decreased from -0.98 to -1.24 °C. In short, temperatures from PSI or simulations with/without
931 ARI turned on agreed well with observations but were mostly underestimated, and the negative bias
932 of T2 simulated by models with ARI turned on got worse and reasons behind it will be explained in
933 Section 6.

934 Figures 3b-c illustrate that RH2 was simulated reasonably well (mean R = 0.73) and the
935 modeled SH2 was also well correlated with observations (R varied between 0.85 and 1.00). RH2
936 and SH2 from more than half of samples had slightly positive and negative mean biases with average
937 MB values of 0.4 % and -0.01 g·kg⁻¹, respectively. The overestimations of RH2 could be caused by
938 the negative bias of T2 (Cuchiara et al., 2014). Compared with results without ARI effects, statistics
939 of RH2 and SH2 from simulations with ARI showed better R and RMSE. However, the increased
940 positive mean biases (average MBs of RH2 and SH2 were from 6.4 % to 7.6 % and from 0.07 g·kg⁻¹
941 to 0.11 g·kg⁻¹, respectively) indicated that turning on ARI could cause further overprediction of
942 humidity variables. Overall, the modeled RH2 and SH2 were in good agreement with observations
943 with slight over- and under-estimations, respectively, and the limited studies showed that RH2 and
944 SH2 simulated by models with ARI turned on had marginally larger positive biases relative to the
945 results without ARI.

946 Compared with the correlation coefficients of T2, RH2 and SH2, mean R (0.59) of WS10 was
947 smallest with a large fluctuation ranging from 0.14 to 0.98 (Fig. 3d). The meta-analysis also
948 indicated that most modeled WS10 tended to be overestimated (81 % of the samples) with the
949 average MB value of 0.79 m·s⁻¹, and the mean RMSE value was 2.76 m·s⁻¹. The general
950 overpredictions of WS10 by WRF (Mass and Ovens, 2011) and coupled models (Gao M. et al.,
951 2018a; Gao Y. et al., 2015) had been explained with possible reasons such as out-of-date
952 geographical data, coarse model resolutions and lacking of better representations of urban canopy
953 physics. The PSI with ARI effects suggested that the correlation of wind speed was slightly
954 improved (mean R from 0.56 to 0.57) and the average RMSE and positive MB decreased by 0.003
955 m·s⁻¹ and 0.051 m·s⁻¹, respectively (Fig. 3h). The collected SI indicated relatively poor performance
956 of modeled WS10 (most wind speeds were overestimated) compared to T2 and humidity, but turning
957 on ARI in coupled models could improve WS10 simulations somewhat.

958 Besides the SI discussed above, very limited papers reported the normalized mean error (NME)
959 (%) of surface meteorological variables (T2, SH2, RH2 and WS10) simulated by two-way coupled
960 models (WRF-Chem and WRF-CMAQ) in Asia, which is summarized in Appendix Table B7. The
961 evaluations with two-way coupled models in Asia showed that the overall mean percent errors of

962 T2, SH2, RH2 and WS10 were 22.71%, 10.32%, 13.94%, and 51.28%, respectively. The ranges of
 963 NME (%) values were quite wide for T2 (from -0.48 to 270.20 %) and WS10 (from 0.33 to 112.28%)
 964 reported by the limited studies. Note that no NME of surface meteorological variables simulated by
 965 two-way coupled models simultaneously with and without enabling the ARI effects was mentioned
 966 in these studies.



967
 968 Figure 3. Quantile distributions of R, MB and RMSE for simulated surface meteorological variables by the five
 969 coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) (a-d) and
 970 comparisons of statistical indices with/out ARI (e-h) in Asia.

971 5.1.2 Comparisons of SI for meteorology using different coupled models

972 Also, to examine how different coupled models (i.e., WRF-Chem, WRF-CMAQ, WRF-
 973 NAQPMS, GRAPES-CUACE and GATOR-GCMOM) performed in Asia with respect to
 974 meteorological variables, the SI were extracted from PSI in term of these five coupled models and
 975 displayed in Fig. 4. The SI for T2, RH2, SH2, and WS10 from WRF-NAQPMS, GRAPES-CUACE
 976 and GATOR-GCMOM simulations were missing or with rather limited samples so that the
 977 discussions here only focused on the WRF-Chem and WRF-CMAQ simulations. Moreover, the SI
 978 sample size from studies involving WRF-Chem was generally larger than that involving WRF-
 979 CMAQ, except for SH2.

980 As seen in Fig. 4a, the modeled T2 by both WRF-CMAQ and WRF-Chem was well correlated
 981 with observations but WRF-CMAQ (mean R = 0.95) outperformed WRF-Chem (mean R = 0.90) to
 982 some extent. On the other hand, WRF-CMAQ underestimated T2 (mean MB = -1.39 °C) but WRF-
 983 Chem slightly overestimated it (mean MB = 0.09 °C) (Fig. 4e). The RMSE of modeled T2 by both
 984 models was at the similar level with mean RMSE values of 2.51 °C and 2.31 °C by WRF-CMAQ
 985 and WRF-Chem simulations, respectively (Fig. 4i).

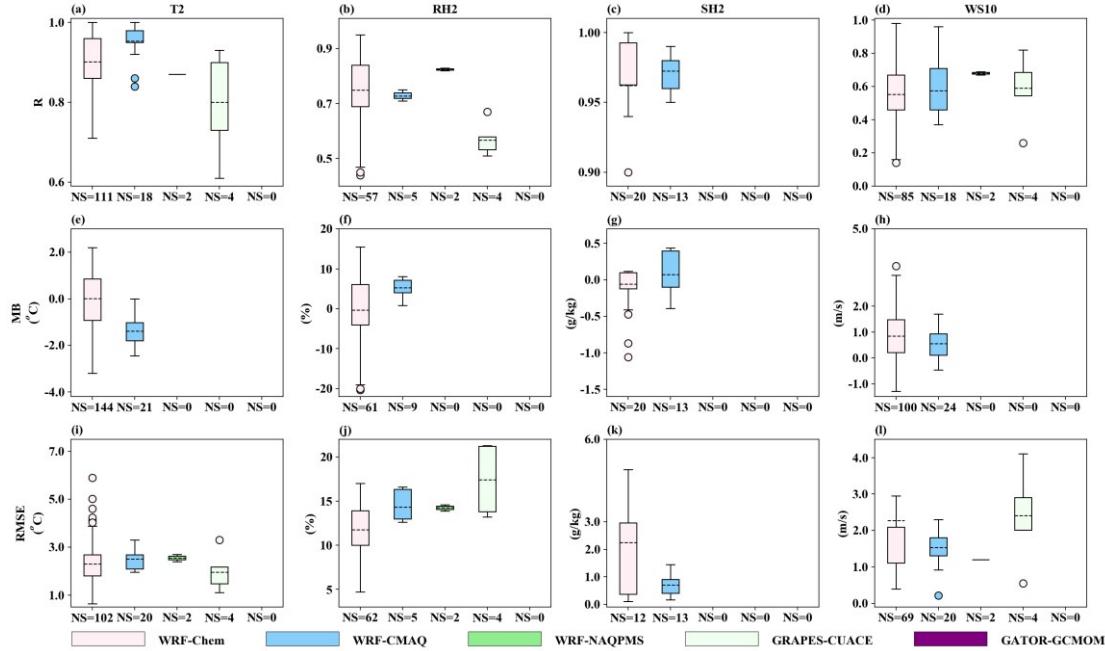
986 Both WRF-Chem and WRF-CMAQ performed better for SH2 (mean R = 0.96 and 0.97,
 987 respectively) than RH2 (mean R = 0.75 and 0.73, respectively) (Figures 4b and 4c), which might be
 988 due to the influence of temperature on RH2 (Bei et al., 2017). Also the modeled RH2 (SH2) by
 989 WRF-Chem correlated better (worsen) with observations than those by WRF-CMAQ. The mean
 990 RMSE of modeled RH2 (Fig. 4j) by WRF-Chem (11.1 %) was lower than that by WRF-CMAQ
 991 (14.3%) but the mean RMSE of modeled SH2 (Fig. 4k) by WRF-Chem (2.25 g·kg⁻¹) higher than
 992 that by WRF-CMAQ (0.71 g·kg⁻¹). It was seen in Figures 4f and 4d that WRF-CMAQ overestimated
 993 RH2 and SH2 (average MB were 5.30 % and 0.07 g·kg⁻¹, respectively), and WRF-Chem
 994 underpredicted RH2 (average MB = -0.32 %) and SH2 (average MB = -0.06 g·kg⁻¹). Generally, the
 995 modeled RH2 and SH2 were reproduced more reasonably by WRF-Chem than those by WRF-
 996 CMAQ.

997 The modeled WS10 by both WRF-Chem and WRF-CMAQ (Fig. 4d) correlated with
 998 observations on the same level with the mean R of 0.56. The mean RMSE of modeled WS10 by
 999 WRF-Chem and WRF-CMAQ were 1.54 m·s⁻¹ and 2.28 m·s⁻¹, respectively, as depicted in Fig. 4l.
 1000 Both models overpredicted WS10 to some extend with average MBs of 0.55 m·s⁻¹ (WRF-CAMQ)
 1001 and 0.84 m·s⁻¹ (WRF-Chem), respectively. These results demonstrated that overall WRF-CMAQ
 1002 and WRF-Chem had similar model performance of WS10.

1003 In general, WRF-CMAQ performed better than WRF-Chem for T2 but worse for humidity
 1004 (RH2 and SH2), and both models' performance for WS10 was very similar. WRF-Chem
 1005 overestimated T2, RH2 and WS10 and underestimated SH2 slightly, while WRF-CMAQ

1007 overpredicted humidity and WS10 but underpredicted T2. Compared to WRF-Chem and WRF-
 1008 CMAQ, the very few SI samples indicated that for the meteorological variables excluding SH2,
 1009 WRF-NAQPMS simulations matched with observations better than GRAPES-CUACE simulations
 1010 but more applications and statistical analysis of these two models are needed to make this kind of
 1011 comparison conclusive.

1012



1013
 1014 Figure 4. Quantile distributions of the statistical indices for simulated surface meteorological variables by WRF-
 1015 Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM in Asia.
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1018 5.2 Model performance for air quality variables

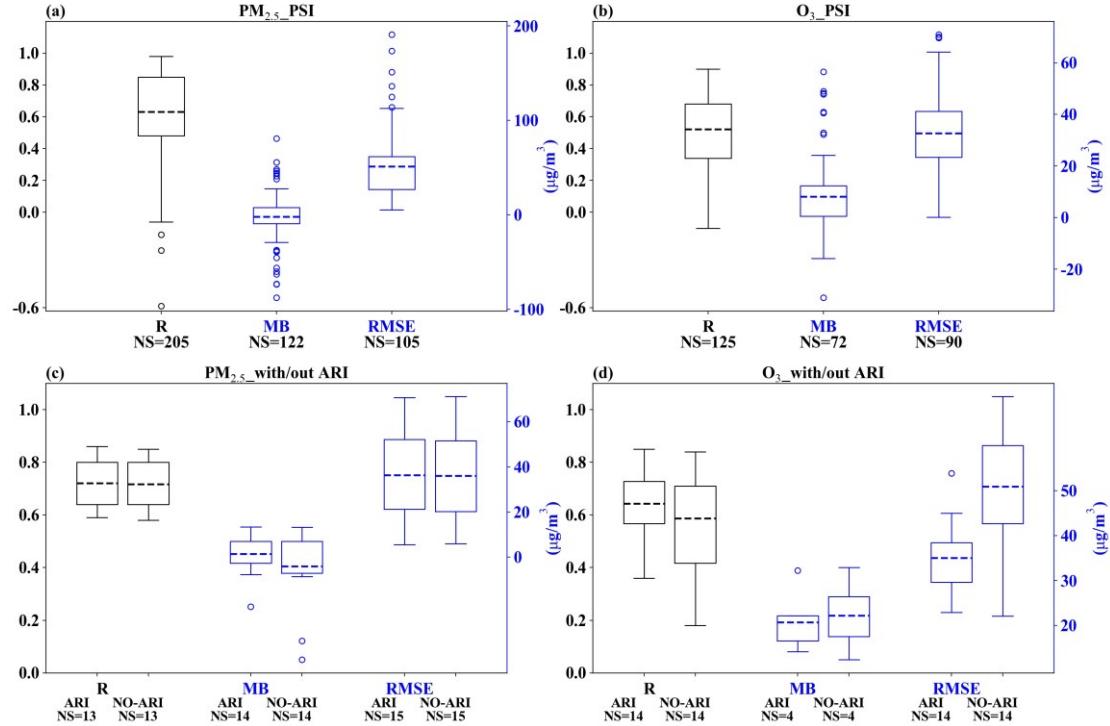
1019 5.2.1 Overall performance

1020 The results of the overall statistical evaluation for the online air quality simulations are
 1021 presented in Figure 5, and all labels and colors indicating SI were the same as those for
 1022 meteorological variables. In this figure and following figures, NP and NS are number of publications
 1023 and samples with SI, respectively and summed up in Appendix Table B3. In Fig. 5a, the correlation
 1024 between the simulated and observed $\text{PM}_{2.5}$ concentrations from PSI showed that in Asia coupled
 1025 models performed relatively well for $\text{PM}_{2.5}$ (mean $R = 0.63$), but RMSE was between -87.60 and
 1026 80.90 and more than half of samples of simulated $\text{PM}_{2.5}$ were underestimated (mean MB = -2.08
 1027 $\mu\text{g}\cdot\text{m}^{-3}$). Note that NS in Fig. 5c-d and Appendix Table B5 counted the samples of SI provided by
 1028 the simulations simultaneously with and without ARI. With the ARI turned on in the coupled models,
 1029 modeled $\text{PM}_{2.5}$ concentrations (limited papers with 15 samples) were improved somewhat and the
 1030 mean R slightly increased from 0.71 to 0.72 and mean absolute MB decreased from 4.10 to 1.33
 1031 $\mu\text{g}\cdot\text{m}^{-3}$ (Fig. 5c), but RMSE of $\text{PM}_{2.5}$ concentrations slightly increased from 35.40 to 36.20 $\mu\text{g}\cdot\text{m}^{-3}$.
 1032 In short, $\text{PM}_{2.5}$ with/without ARI agreed well with observations but were mostly underestimated,
 1033 and $\text{PM}_{2.5}$ bias simulated by models became overpredicted.

1034 Compared with $\text{PM}_{2.5}$, mean R (0.59) of O_3 was relatively smaller (Fig. 5b). The statistical
 1035 analysis also showed the most modeled O_3 concentrations tended to be overestimated (76 % of the
 1036 samples) with the average MB value of $8.05 \mu\text{g}\cdot\text{m}^{-3}$, and the mean RMSE value was $32.65 \mu\text{g}\cdot\text{m}^{-3}$.
 1037 The 14 PSI with ARI effects suggested that the correlation of O_3 was slightly improved (mean R from
 1038 0.58 to 0.64) and the average RMSE and MB were decreased by $15.93 \mu\text{g}\cdot\text{m}^{-3}$ and $1.55 \mu\text{g}\cdot\text{m}^{-3}$,
 1039 respectively (Fig. 5d). The collected studies indicated relatively poor performance of modeled O_3
 1040 compared to $\text{PM}_{2.5}$, but turning on ARI in coupled models improved O_3 simulations somewhat.

1041 In addition to the SI analyzed above and similar to the surface meteorological variables, the
 1042 NME (%) of $\text{PM}_{2.5}$ and O_3 is listed in Table B7. The limited studies with WRF-Chem and WRF-
 1043 CMAQ indicated that the overall mean percent errors of $\text{PM}_{2.5}$ and O_3 were 47.63% (from 29.55 to
 1044 104.70 %) and 43.03% (from 21.10 to 127.00 %), respectively. With the ARI effects enabled in
 1045 WRF-Chem in different seasons over the China domain, the NME (%) of $\text{PM}_{2.5}$ increased slightly

1046 during most seasons, except during a spring month with little change (Zhang et al., 2018). Another
 1047 study by Nguyen et al. (2019b) revealed that the NME (%) of $\text{PM}_{2.5}$ and O_3 simulated by WRF-
 1048 CMAQ became a little worse in SEA comparing to the simulations without ARI.

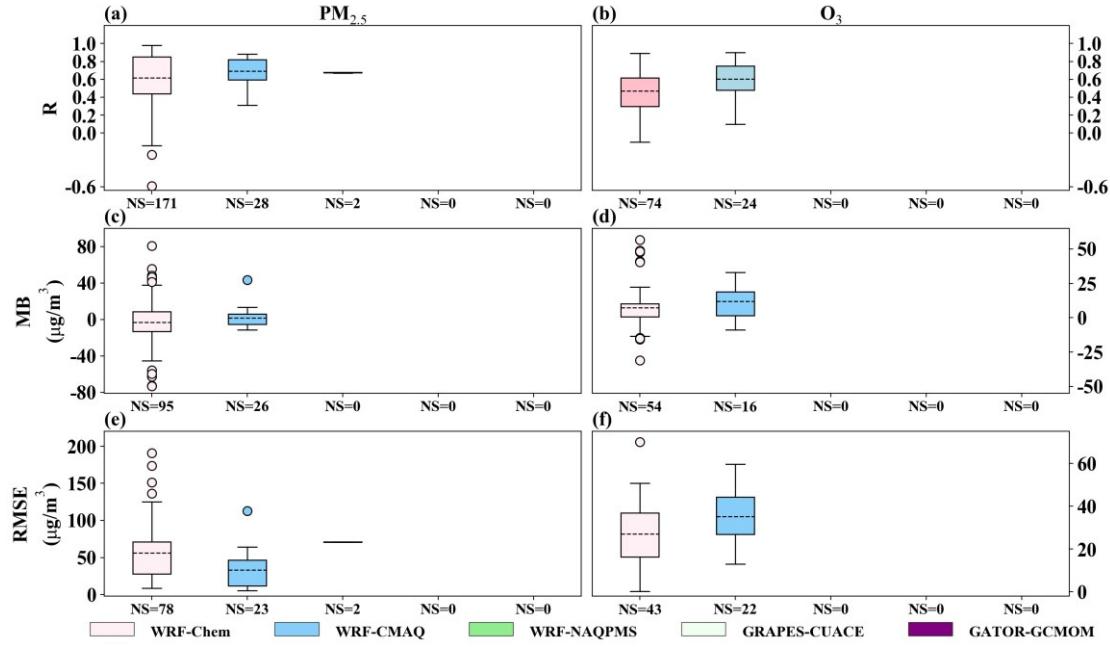


1049 Figure 5. Quantile distributions of statistical indices for simulated $\text{PM}_{2.5}$ and O_3 (a-b) by the five two-way coupled
 1050 models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) and comparisons
 1051 of statistical indices with/out ARI (c-d) in Asia.

1052 5.2.2 Comparisons of SI for air quality using different coupled models

1053 Figure 6 showed the SI for $\text{PM}_{2.5}$ and O_3 from different coupled models, and only WRF-Chem
 1054 and WRF-CMAQ simulations were discussed for the same reason as in Section 5.1.2. The modeled
 1055 $\text{PM}_{2.5}$ by WRF-CMAQ (mean $R = 0.69$) outperformed WRF-Chem (mean $R = 0.62$) to some extent
 1056 (Fig. 6a) and the RMSE of modeled $\text{PM}_{2.5}$ by WRF-CMAQ ($33.24 \mu\text{g} \cdot \text{m}^{-3}$) was smaller than that by
 1057 WRF-Chem ($56.16 \mu\text{g} \cdot \text{m}^{-3}$). With respect to MB, WRF-CMAQ overestimated $\text{PM}_{2.5}$ (mean $\text{MB} =$
 1058 $+1.60 \mu\text{g} \cdot \text{m}^{-3}$) but WRF-Chem slightly underestimated it (mean $\text{MB} = -3.12 \mu\text{g} \cdot \text{m}^{-3}$) (Fig. 6c). Figure
 1059 6b showed that the modeled O_3 by WRF-CMAQ (0.60) correlated better with observations than
 1060 those by WRF-Chem (0.47), but the mean RMSE of modeled O_3 (Fig. 6f) by WRF-Chem ($27.13 \mu\text{g} \cdot \text{m}^{-3}$)
 1061 was lower than that by WRF-CMAQ ($35.19 \mu\text{g} \cdot \text{m}^{-3}$). It was seen in Figures 6d that both
 1062 WRF-CMAQ and WRF-Chem overestimated O_3 , with mean MBs as 11.98 and $7.21 \mu\text{g} \cdot \text{m}^{-3}$,
 1063 respectively. Generally, the modeled $\text{PM}_{2.5}$ and O_3 were reproduced more reasonably by WRF-
 1064 CMAQ than by WRF-Chem, even though there were much more samples available from WRF-
 1065 Chem simulations than WRF-CMAQ simulations.

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1071 Figure 6. Quantile distributions of R, MB and RMSE of PM_{2.5} and O₃ simulated by WRF-Chem, WRF-CMAQ,
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1075 GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM in Asia.

6 Impacts of aerosol feedbacks in Asia

1076 Aerosol feedbacks not only impact the performances of two-way coupled models but also the
1077 simulated meteorological and air quality variables to a certain extent. In this section, we collected
1078 and quantified the variations (Table S3) of these variables induced by ARI or/and ACI from the
1079 modeling studies in Asia. Due to limited sample sizes in the collected papers, the target variables
1080 only include radiative forcing, surface meteorological parameters (T2, RH2, SH2 and WS10), PBLH,
1081 cloud, precipitation, and PM_{2.5} and gaseous pollutants.

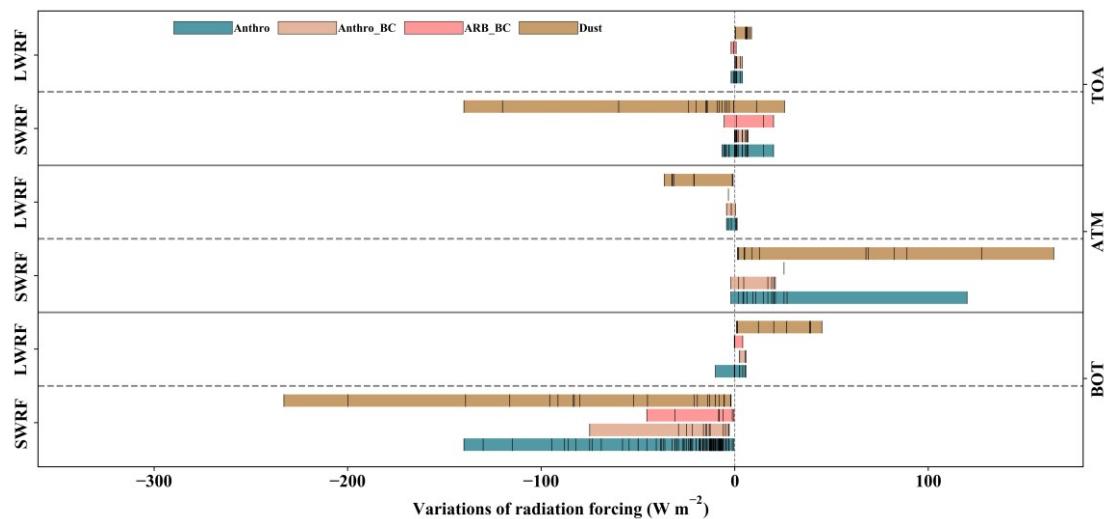
6.1 Impacts of aerosol feedbacks on meteorology

6.1.1 Radiative forcing

1082 With regard to radiative forcing, most studies with two-way coupled models in Asia had
1083 focused on the effects of dust aerosols (Dust), BC emitted from ARB (ARB_BC) and anthropogenic
1084 sources (Anthro_BC), and total anthropogenic aerosols (Anthro). Figure 7 presents the variations of
1085 simulated SWRF and LWRF at the bottom (BOT) and TOA and in the ATM due to aerosol feedbacks,
1086 and detailed information of these variations are compiled in Table S5. In this figure, the color bars
1087 show the range of radiative forcing variations and the black tick marks inside the color bars represent
1088 these variations extracted from all the collected papers. It should be noted that in this figure all the
1089 radiative forcing variations were plotted regardless of temporal resolutions of data reporting and
1090 simulation durations. Apparently in Asia, most studies targeted the SWRF variations induced by
1091 anthropogenic aerosols at the BOT that exhibited the largest differences ranging from -140.00 to -
1092 0.45 W·m⁻², with the most variations (88 % of samples) concentrated in the range of -50.00 to -0.45
1093 W·m⁻². The SWRF variations due to anthropogenic aerosols in the ATM and at the TOA were -2.00
1094 to +120.00 W·m⁻² and -6.50 to 20.00 W·m⁻², respectively. There were much less studies reported
1095 LWRF variations caused by anthropogenic aerosols, which ranged from -10.00 to +5.78 W·m⁻², -
1096 1.91 to +3.94 W·m⁻², and -4.26 to +1.21 W·m⁻² at the BOT and TOA, and in the ATM, respectively.

1097 Considering BC from anthropogenic sources and ARB, they both led to positive SWRF at the
1098 TOA (with mean values of 2.69 and 7.55 W·m⁻², respectively) and in the ATM (with mean values
1099 of 11.70 and 25.45 W·m⁻², respectively) but negative SWRF at the BOT (with mean values of -18.43
1100 and -14.39 W·m⁻², respectively). The responses of LWRF to Anthro_BC and ARB_BC at the BOT
1101 (in the ATM) on average were 4.01 and 0.72 W·m⁻² (-1.89 and -3.24 W·m⁻²), respectively, and weak
1102 at the TOA (+0.92 and -0.53 W·m⁻², respectively). The SWRF variations induced by dust were in
1103 the range of -233.00 to -1.94 W·m⁻² and -140.00 to +25.70 W·m⁻², and +1.44 to +164.80 W·m⁻² at
1104 the BOT and TOA, and in the ATM, respectively. The LWRF variations caused by dust were the

1108 largest (with mean values of $22.83 \text{ W} \cdot \text{m}^{-2}$ and $+5.20 \text{ W} \cdot \text{m}^{-2}$, and $-22.12 \text{ W} \cdot \text{m}^{-2}$ at the BOT and TOA,
 1109 and in the ATM, respectively), comparing to the ones caused by anthropogenic aerosols and BC
 1110 aerosols from anthropogenic sources and ARB.
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 1113 Figure 7. Variations of shortwave and longwave radiative forcing (SWRF and LWRF) simulated by two-way
 1114 coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) with
 1115 aerosol feedbacks at the bottom and top of atmosphere (BOT and TOA), and in the atmosphere (ATM)
 1116 in Asia.

1117 As shown in Fig. 7, SWRF variations at the BOT caused by total aerosols (sum of Anthro,
 1118 Anthro_BC, ARB_BC and Dust) had been widely assessed in Asia. Therefore, we further analyzed
 1119 their spatiotemporal distributions and inter-regional differences, which are displayed in Fig. 8.
 1120 Figure 8a presents the SWRF variations over different areas of Asia (the acronyms used in Fig. 8
 1121 are listed in Appendix Table B1) at different time scales. In Asia, almost 41 % of the selected papers
 1122 investigated SWRF towards its monthly variations, 36 % towards its hourly and daily variations,
 1123 and 23 % towards its seasonal and yearly variations. Most studies reported aerosol-induced SWRF
 1124 variations were primarily conducted in NCP, EA, China, and India. At the hourly scale, the range of
 1125 SWRF decreases was from -350.00 to $-5.90 \text{ W} \cdot \text{m}^{-2}$ (mean value of $-106.92 \text{ W} \cdot \text{m}^{-2}$) during typical
 1126 pollution episodes, and significant variations occurred in EA. The daily and monthly mean SWRF
 1127 reductions varied from -73.71 to $-5.58 \text{ W} \cdot \text{m}^{-2}$ and -82.20 to $-0.45 \text{ W} \cdot \text{m}^{-2}$, respectively, with relative
 1128 large perturbations in NCP. At the seasonal and yearly scales, the SWRF changes ranged from
 1129 -22.54 to $-3.30 \text{ W} \cdot \text{m}^{-2}$ and -30.00 to $-2.90 \text{ W} \cdot \text{m}^{-2}$ with mean value of -11.28 and $-11.82 \text{ W} \cdot \text{m}^{-2}$,
 1130 respectively, with EA as the most researched area.

1131 To identify the differences of aerosol-induced SWRF variations between high- (Asia) and low-
 1132 polluted regions (Europe and North America), their inter-regional comparisons are depicted in Fig.
 1133 8b. This figure does not include information about temporal resolutions of data reporting and
 1134 durations of model simulations with ARI or/and ACI, but intends to delineate the range of SWRF
 1135 changes due to aerosol feedbacks. The SWRF variations fluctuated from -233.00 to $-0.45 \text{ W} \cdot \text{m}^{-2}$,
 1136 -100.00 to $-1.00 \text{ W} \cdot \text{m}^{-2}$, and -600.00 to $-1.00 \text{ W} \cdot \text{m}^{-2}$ in Asia, Europe, and North America, respectively.
 1137 It should be pointed out that the two extreme values were caused by dust ($-233.00 \text{ W} \cdot \text{m}^{-2}$) in Asia
 1138 and wildfire ($-600.00 \text{ W} \cdot \text{m}^{-2}$) in North America. Overall, the median value of SWRF reductions due
 1139 to ARI or/and ACI in Asia ($-15.92 \text{ W} \cdot \text{m}^{-2}$) was larger than those in North America ($-10.50 \text{ W} \cdot \text{m}^{-2}$)
 1140 and Europe ($-7.00 \text{ W} \cdot \text{m}^{-2}$).
 1141

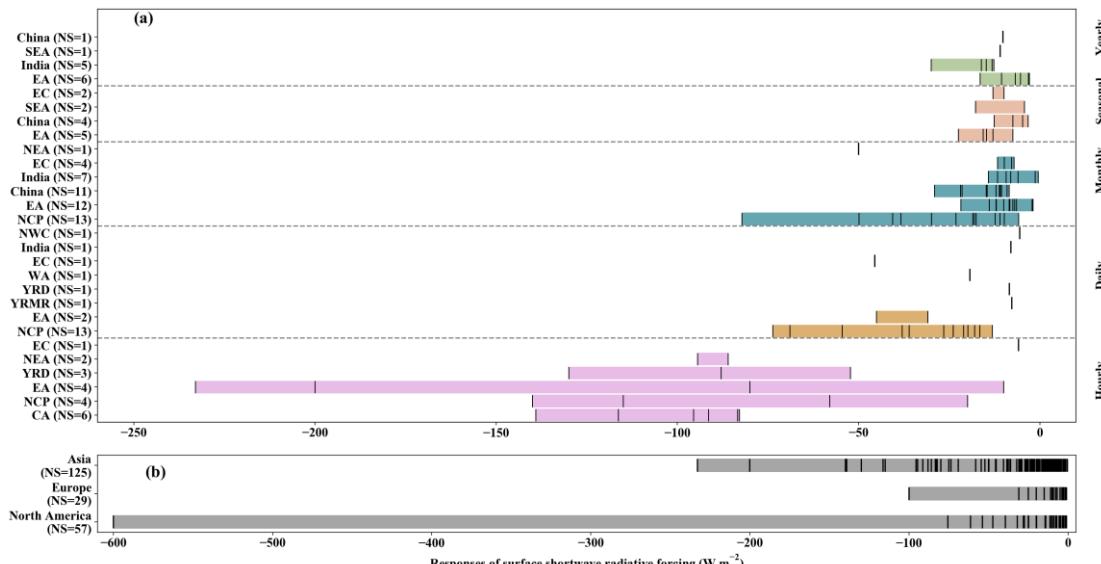


Figure 8. Responses of shortwave radiation forcing to aerosol feedbacks in different areas/periods in Asia (a) and the inter-regional comparisons of its variations in Asia, Europe and North America (b).

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6.1.2 Temperature, wind speed, humidity and PBLH

1148 The impact of aerosols on radiation can influence energy balance, which eventually alter other
 1149 meteorological variables. The summary of aerosol-induced variations of T2, WS10, RH2, SH2 and
 1150 PBLH in different regions of Asia as well as at different temporal scales are provided in Table 6. In
 1151 this table, the minimum and maximum values were collected from the corresponding papers and the
 1152 mean values were calculated with adding all the variations from these papers and then divided by
 1153 the number of samples.

1154 Overall, aerosol effects led to decreases of T2, WS10 and PBLH with average changes of -
 1155 0.65 $^{\circ}\text{C}$, $-0.13 \text{ m}\cdot\text{s}^{-1}$ and -60.70 m , respectively, and increases of humidity (mean $\Delta\text{RH2} = 2.56 \%$)
 1156 in most regions of Asia. On average, the hourly aerosol-induced changes of surface meteorological
 1157 variables (T2, WS10 and RH2) and PBLH were the largest among the different time scales. At the
 1158 hourly time scale, the mean variations of T2, WS10, RH2 and PBLH due to ARI or/and ACI were -
 1159 $-1.85 \text{ }^{\circ}\text{C}$, $-0.32 \text{ m}\cdot\text{s}^{-1}$, 4.60% and -165.84 m , respectively, and their absolute maximum values in EC,
 1160 YRD, NCP and NCP, respectively. Compared to variations at the hourly time scale, smaller daily
 1161 variations of T2, WS10, RH2 and PBLH were caused by aerosol effects, and their mean values were
 1162 $-0.63 \text{ }^{\circ}\text{C}$, $-0.15 \text{ m}\cdot\text{s}^{-1}$, $+2.89 \%$ and -34.61 m , respectively. The largest daily variations of T2, WS10,
 1163 RH2 and PBLH occurred in NCP, EC, EC and SEC, respectively. For other time scales (monthly,
 1164 seasonal and yearly), the respective mean variations of T2, RH2 and PBLH induced by aerosol
 1165 effects were comparable. However, the WS10 perturbations at the monthly time scale were about
 1166 two to three times higher than those at the seasonal and yearly time scales. High variations at the
 1167 monthly, seasonal and yearly time scales were reported in NCP (T2, RH2 and PBLH), EA (T2,
 1168 WS10 and PBLH) and PRD (T2 and PBLH), respectively. In addition, comparing to T2 and PBLH,
 1169 the aerosol-induced variations of WS10 and humidity were less revealed.

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Table 6. Summary of variations of surface meteorological variables and planetary boundary layer height (PBLH) caused by aerosol feedbacks simulated by two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) in different regions of Asia and at different temporal scales.

Region	Time scale	ΔT2 [mean] ($^{\circ}\text{C}$)	ΔWS10 [mean] ($\text{m}\cdot\text{s}^{-1}$)	$\Delta\text{RH2}/\text{SH2}$ [mean]	ΔPBLH [mean] (m)
EC	hours	-8.00 to -0.20 [-2.68]			-300.00 to -50.00 [-175.00]
EA	hours	-3.00 to -2.00 [-2.50]			
YRD	hours	-1.40 to -1.00 [-1.15]	-0.80 to -0.10 [-0.41]		-276.00 to -29.90 [-105.42]
NCP	hours	-2.80 to -0.20 [-1.05]	-0.30 to -0.10 [-0.23]	1.00 % to 12.00 % [4.60 %]	-287.20 to -147.00 [-217.10]
Hourly mean		-1.85	-0.32	4.60%	-165.84
NCP	days	-2.00 to -0.10 [-0.88]	-0.4 to -0.01 [-0.17]	0.51 % to 4.10 % [2.52 %]	-111.40 to -10.00 [-49.07]

EC	days	-0.94 to -0.65 [-0.79]	-0.52 to -0.37 [-0.45]	1.92 % to 9.75 % [5.84 %]	
India	days	-1.60 to 0.10 [-0.75]			
SEC	days	-1.38 to -0.18 [-0.70]	-0.07 to 0.05 [-0.023]	-0.37 % to 6.57 % [2.63 %]	-84.1 to -27.55 [-53.62]
NEA	days	-0.52	-0.08		-46.39
MRYR	days	-0.16	-0.01	0.56 %	-16.46
India	days				-6.90
Daily mean		-0.63	-0.15	2.89 %	-34.61
India	months	-0.45			
NCP	months	-1.30 to -0.06 [-0.43]		1.30 % to 4.70 % [2.53 %]	-109.00 to -5.48 [-36.01]
NEA	months	-0.30	-0.10		-50.00
PRD	months	-0.60 to 0.13 [-0.16]			
EA	months	-0.45 to -0.03 [-0.13]			-35.70 to -13.00 [-24.35]
China	months	-0.89 to 0.60 [-0.12]			-66.60 to -2.30 [-25.67]
EC	months	-0.30 to -0.05 [-0.11]			-13.10 to -6.20 [-9.65]
Monthly mean		-0.24	-0.10	2.53 %	-29.13
EA	seasons	-0.58 to -0.30 [-0.40]	-0.05 to -0.02 [-0.035]		-64.62 to -30.70 [-43.27]
SEA	seasons	-0.39 to -0.03 [-0.21]	-0.06 to -0.01 [-0.035]		-48.33 to -6.71 [-27.52]
Seasonal mean		-0.31	-0.035		-34.61
PRD	years	-0.27			-45.00
TP	years	-0.24			
SEA	years	-0.21	-0.03		-27.25
EA	years		-0.03	0.13 g·kg ⁻¹	-46.47 to -45.00 [-45.74]
EC	years		-0.014	0.21 %	
Yearly mean		-0.24	-0.025	0.21 %	-39.33

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1175 **6.1.3 Cloud and precipitation**

1176 In the included publications, only a few papers focusing on the effects of aerosol feedbacks on
 1177 cloud properties (cloud fraction, LWP, ice water path (IWP), CDNC and cloud effective radius) and
 1178 precipitation characteristics (amount, spatial distribution, peak occurrence and onset time) using
 1179 two-way coupled models in Asia, as shown in Table 7. In this table, the abbreviations representing
 1180 aerosol emission sources (Dust, ARB_BC, Anthro_BC, and Anthro) and regions in Asia are defined
 1181 in Appendix Table B1. The plus and minus signs indicate increase and decrease, respectively.

1182 The variations of cloud properties and precipitation characteristics induced by ARI or/and ACI
 1183 are rather complex and not uniform in different parts of Asia and time periods. BC from both ARB
 1184 and anthropogenic sources reduced cloud fraction through ARI and both ARI and ACI in several
 1185 areas in China. ARI or/and ACI induced by anthropogenic aerosols could increase or decrease cloud
 1186 fraction and affect cloud fraction differently in various atmospheric layers and time periods.
 1187 Considering EA and subareas in China, anthropogenic aerosols tended to increase LWP through ARI
 1188 and ACI as well as ACI alone but decrease LWP in some areas of SC (ARI and ACI) at noon and in
 1189 afternoon during summertime and NC (ACI) in winter. ARI and ACI induced by anthropogenic BC
 1190 aerosols had negative effects on LWP except at daytime in CC. Dust aerosols increased both LWP
 1191 and IWP through ACI in EA, which was reported only by one study. The increase (decrease) of
 1192 CDNC caused by the ARI and ACI effects of anthropogenic (anthropogenic BC) aerosols in EC
 1193 during summertime was reported. Through ACI, anthropogenic aerosols affected CDNC positively
 1194 in EA and China. Compared to anthropogenic aerosols, dust aerosols could have much larger
 1195 positive impacts on CDNC via ACI in springtime over EA. The ACI effects of anthropogenic
 1196 aerosols reduced cloud effective radius over China (January) and EA (July).

1197 Among all the variables describing cloud properties and precipitation characteristics, the
 1198 variations of precipitation amount were studied the most using two-way coupled models in Asia.
 1199 How turning on ARI or/and ACI in coupled models can change precipitation amount is not
 1200 unidirectional and depends on many factors, including different aerosol sources, areas, emission
 1201 levels, atmospheric humidity, precipitation types, seasons, and time of a day. Under the high

emission levels as well as at slightly different humidity levels of $RH > 85\%$ with increasing emissions, the ACI effects of anthropogenic aerosols induced precipitation increase in the MRYR area of China. Over the same area, precipitation decreased due to the ACI effects of anthropogenic aerosols with the low emission levels and $RH < 80\%$. In PRD, wintertime precipitation was enhanced by the ACI effects of anthropogenic aerosols but inhibited by ARI. In SK, summertime precipitation was both enhanced and inhibited by the ACI and ARI effects of anthropogenic aerosols. In locations upwind (downwind) of Beijing, rainfall amount was raised (lowered) by the ARI effects of anthropogenic aerosols but lowered (raised) by ACI. Both ARI and ACI induced by anthropogenic aerosols had positive impacts on total, convective, and stratiform rain in India during the summer season and the increase of convective rain was larger than those of stratiform. Summertime precipitation amounts could be enhanced or inhibited at various subareas inside simulation domains over India, China, and Korea and during day- or night-time due to ARI and ACI of anthropogenic aerosols. Over China, dust-induced ACI decreased (increased) springtime precipitation in CC (western part of NC), and over India, dust aerosols from local sources and ME had positive impacts on total, convective, and stratiform rain through ARI and ACI. Simulations in India also revealed that precipitation could be increased in some subareas but decreased in another and absorptive (non-absorptive) dust enhanced (inhibited) summertime precipitation via ARI and ACI. The ARI (ACI) effects of BC from ARB caused precipitation reduction (increase) in SEC but CAs emitted from ARB (ARB_CAs) caused rainfall enhancement in Myanmar. During pre-monsoon (monsoon) season, ARI induced by anthropogenic BC could lead to +42 % (-5 to -8 %) variations of precipitation in NEI (SI). Considering both ARI and ACI effects, BC from ARB and sea salt aerosols enhanced or inhibited precipitation in different parts of India and BC from anthropogenic sources enhanced (inhibited) nighttime (daytime) rainfall in CC (NC and SC) at the rate of +1 to +4 $mm \cdot day^{-1}$ (-2 to -6 $mm \cdot day^{-1}$) during summer season. With respect to spatial variations, 6.5 % larger rainfall area in PRD was caused by ARI and ACI effects under 50 % reduced anthropogenic emissions. ACI induced by anthropogenic aerosols tended to delay the peak occurrence time and onset time of precipitation by one to nine hours in China and South Korea.

Table 7. Summary of changes of cloud properties and precipitation characteristics due to aerosol feedbacks simulated by two-way coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMs and GATOR-GCMOM) in Asia.

Variables	Variations (aerosol effects)	Simulation time period	Regions	References
Cloud fraction	-7 % low-level cloud (ARB_BC ARI)	Apr., 2013	SEC	Huang et al., 2019
	+0.03 to +0.08 below 850 hPa and at 750 hPa (Anthro ARI & ACI), esp. at early morning and nighttime	Aug., 2008	EC	Gao and Zhang, 2018
	Max -0.06 between 750 hPa and 850 hPa (Anthro ARI & ACI), esp. in afternoon and evening	Aug., 2008	CC	Gao and Zhang, 2018
	-0.02 to -0.06 below 750 hPa (Anthro_BC ARI & ACI), esp. in afternoon	Aug., 2008	SC & NC	Gao and Zhang, 2018
	-0.04 to -0.06 between 750 hPa and 850 hPa (Anthro_BC ARI & ACI), esp. in afternoon	Aug., 2008	CC	Gao and Zhang, 2018
	-6.7 % to +3.8 % (Anthro ARI)	Jun. 6-9 & Jun. 11-14, 2015	SK	Park et al., 2018
	+22.7 % (Anthro ACI)	Jun. 6-9 & Jun. 11-14, 2015	SK	Park et al., 2018
	-0.03 % low-, -0.54 % middle- and -0.58 % high-level cloud (Anthro ACI)	2008 to 2012	PRD	Liu Z. et al., 2018
	+5 to +50 $g \cdot m^{-2}$ (Anthro ARI & ACI)	Aug., 2008	EC	Gao and Zhang, 2018
	+10 to +20 $g \cdot m^{-2}$ (Anthro_BC ARI & ACI) at daytime	Aug., 2008	CC	Gao and Zhang, 2018
Cloud properties	-5 to -40 $g \cdot m^{-2}$ (Anthro ARI & ACI) at noon and in afternoon	Aug., 2008	Part of SC	Gao and Zhang, 2018
	-2 to -20 $g \cdot m^{-2}$ (Anthro_BC ARI & ACI)	Aug., 2008	SC	Gao and Zhang, 2018
	-2 to -30 $g \cdot m^{-2}$ (Anthro_BC ARI & ACI)	Aug., 2008	NC	Gao and Zhang, 2018
	Max+18 $g \cdot m^{-2}$ (Dust ACI)	Mar.-May., 2010	EA	Wang et al., 2018
	+40 to +60 $g \cdot m^{-2}$ (Anthro ACI)	Jan., 2008	SC	Gao et al., 2012
	+40 $g \cdot m^{-2}$ (Anthro ACI)	Jan., 2008	CC	Gao et al., 2012
	Less than +5 $g \cdot m^{-2}$ or -5 $g \cdot m^{-2}$ (Anthro ACI)	Jan., 2008	NC	Gao et al., 2012
LWP	+30 to +50 $g \cdot m^{-2}$ (Anthro ACI)	Jul., 2008	EA	Gao et al., 2012
	+5 to +10 $g \cdot m^{-2}$ (Dust ACI)	Mar. 17-Apr. 30, 2012	EA	Su and Fung, 2018a
CDNC	+20 to +160 cm^{-3} (Anthro ARI & ACI)	Aug., 2008	EC	Gao and Zhang, 2018

	-5 to -60 cm ³ (Anthro_BC ARI & ACI)	Aug., 2008	EC	Gao and Zhang, 2018
	Max +10500 cm ⁻³ (Dust ACI)	Mar.-May., 2010	EA	Wang et al., 2018
	+650 cm ⁻³ (Anthro ACI)	Jan., 2008	EC	Gao et al., 2012
	+400 cm ⁻³ (Anthro ACI)	Jan., 2008	CC & SWC	Gao et al., 2012
	Less than +200 cm ⁻³ (Anthro ACI)	Jan., 2008	NC	Gao et al., 2012
	+250 to +400 cm ⁻³ (Anthro ACI)	Jul., 2008	EA	Gao et al., 2012
Cloud effective radius	More than -4 μ m (Anthro ACI)	Jan., 2008	SWC, CC & SEC	Gao et al., 2012
	More than -2 μ m (Anthro ACI)	Jan., 2008	NC	Gao et al., 2012
	-3 μ m (Anthro ACI)	Jul., 2008	EA	Gao et al., 2012
Enhancement/inhibition of precip. due to high/low Anthro emissions, ACI inhibited (enhanced) precip. at RH < 80 % (> 85 %) with increasing Anthro emissions		Jun. 18-19, 2018	MRYR	Bai et al., 2020
Precipitation (precip.)	-4.72 mm (Anthro ARI) and +33.7 mm (Anthro ACI)	Dec. 14-16, 2013	PRD	Liu Z. et al., 2020
	+2 to +5 % (ARB_CAs ARI)	Mar.-Apr., 2013	Myanmar	Singh et al., 2020
	-1.09 mm·day ⁻¹ (ARB_BC ARI)	Apr., 2013	SEC	Huang et al., 2019
	+0.49 mm·day ⁻¹ (ARB_BC ACI)	Apr., 2013	SEC	Huang et al., 2019
	-0 to -4 mm·day ⁻¹ (Anthro ARI & ACI)	Jun.-Sep., 2010	Indus basin & eastern IGP	Kedia et al., 2019b
	+1 to +3 mm·day ⁻¹ non-convective rain (Anthro ARI & ACI)	Jun.-Sep., 2010	WG of India	Kedia et al., 2019b
	+5 mm·day ⁻¹ non-convective rain (Anthro ARI & ACI)	Jun.-Sep., 2010	NEI	Kedia et al., 2019b
	Increase of total rain (Dust ARI & ACI)	Jun.-Sep., 2010	NI, CI, WG, NEI & central IGP	Kedia et al., 2019b
	Decrease of total rain (Dust ARI & ACI)	Jun.-Sep., 2010	NWI & SPI	Kedia et al., 2019b
	Decrease of total rain (ARB_BC ARI & ACI)	Jun.-Sep., 2010	WG, SPI, NWI, EI & NEI	Kedia et al., 2019b
Increase of total rain (ARB_BC ARI & ACI)		Jun.-Sep., 2010	CI, Central IGP & EPI	Kedia et al., 2019b
Decrease of total rain (Sea salt ARI & ACI)		Jun.-Sep., 2010	EPI, WPI, CPI & SPI	Kedia et al., 2019b
Increase of total rain (Sea salt ARI & ACI)		Jun.-Sep., 2010	NCI & central IGP	Kedia et al., 2019b
-20 to -200mm (Anthro ARI & ACI)		Aug., 2008	SC & NC	Gao and Zhang, 2018
Amount	+20 to +100 mm (Anthro_BC ARI & ACI)	Aug., 2008	CC	Gao and Zhang, 2018
	+1 to +4 mm·day ⁻¹ nighttime precip. (ARI & ACI of Anthro or Anthro_BC)	Aug., 2008	CC	Gao and Zhang, 2018
	-2 to -6 mm·day ⁻¹ daytime precip. (ARI & ACI of Anthro or Anthro_BC)	Aug., 2008	NC	Gao and Zhang, 2018
	-2 to -4 mm·day ⁻¹ daytime precip. (Anthro ARI & ACI)	Aug., 2008	SC	Gao and Zhang, 2018
	-2 to -6 mm·day ⁻¹ daytime precip. (Anthro_BC ARI & ACI)	Aug., 2008	SC	Gao and Zhang, 2018
	-54.6 to +24.1 mm (Anthro ARI)	Jun. 6-9, 2015	SK	Park et al., 2018
	-23.8 to +24.0 mm (Anthro ACI)	Jun. 6-9, 2015	SK	Park et al., 2018
	-63.2 to +27.1 mm (Anthro ARI & ACI)	Jun. 6-9, 2015	SK	Park et al., 2018
	Min -7.0 mm (Anthro ARI)	Jun. 11-14, 2015	SK	Park et al., 2018
	Min -36.6 mm (Anthro ACI)	Jun. 11-14, 2015	SK	Park et al., 2018
	+42 % (Anthro_BC ARI) during pre-monsoon season	Mar.-May., 2010	NEI	Soni et al., 2018
	-5 to -8 % (Anthro_BC ARI) during monsoon season	Jun.-Sep., 2010	SI	Soni et al., 2018
	+1 mm·day ⁻¹ precip. (Dust ACI)	Mar. 17-Apr. 30, 2012	Western part of NC	Su and Fung, 2018b
	-1 mm·day ⁻¹ precip. (Dust ACI)	Mar. 17-Apr. 30, 2012	CC	Su and Fung, 2018b
	+0.95 mm·day ⁻¹ precip. (absorptive Dust ARI & ACI)	Jun.-Aug., 2008	India	Jin et al., 2016a
	-0.4 mm·day ⁻¹ precip. (non-absorptive Dust ARI & ACI)	Jun.-Aug., 2008	India	Jin et al., 2016a
	+0.44 mm·day ⁻¹ total precip. (Dust ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b

	+0.34 mm·day ⁻¹ total precip. (Dust ARI & ACI from ME)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.31 mm·day ⁻¹ total precip. (Anthro ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.32 mm·day ⁻¹ convective precip. (Dust ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.24 mm·day ⁻¹ convective precip. (ARI & ACI of Dust from ME)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.20 mm·day ⁻¹ convective precip. (Anthro ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.12 mm·day ⁻¹ stratiform precip. (Dust ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.10 mm·day ⁻¹ stratiform precip. (ARI & ACI of Dust from ME)	Jun.-Aug., 2008	India	Jin et al., 2016b
	+0.11 mm·day ⁻¹ stratiform precip. (Anthro ARI & ACI over whole study domain)	Jun.-Aug., 2008	India	Jin et al., 2016b
	-48.29 %/+24.87 % precip. in downwind/upwind regions (Anthro ARI)	Jun. 27-28, 2008	Beijing	Zhong et al. 2015
	+33.26 % /-4.64 % precip. in downwind/upwind regions (Anthro ACI)	Jun. 27-28, 2008	Beijing	Zhong et al. 2015
	+0.44 mm·day ⁻¹ precip. (Dust ARI & ACI)	Jun. 1-Aug. 31, 2008	India	Jin et al., 2015
Spatial variation	+6.5 % precip. area (ARI & ACI) with 50% Anthro emissions	Jun. 9-12, 2017	YRD	Liu C. et al., 2019
	1 to 2h delay (Anthro ACI)	Jun. 18-19, 2018	MRYR	Bai et al., 2020
Peak occurrence time	1h delay (ARI & ACI) with 50% Anthro emissions	Jun. 9-12, 2017	YRD	Liu C. et al., 2019
	9h delay (Anthro ACI)	Jun. 7, 2015	Gosan, SK	Park et al., 2018
	4h delay (Anthro ACI)	Jun. 7, 2015	Jinju, SK	Park et al., 2018
	9h delay (Anthro ACI)	Jun. 7, 2015	Gosan, SK	Park et al., 2018
Onset time	2h delay (Anthro ACI)	Jun. 7, 2015	Jinju, SK	Park et al., 2018

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6.2 Impacts of aerosol feedbacks on air quality

1235 Aerosol effects not only gave rise to changes in meteorological variables but also air quality.
 1236 Table 8 (the minimum, maximum and mean values were defined in the same way as in Table 6)
 1237 summarizes the variations of atmospheric pollutant concentrations induced by aerosol effects in
 1238 different regions of Asia and at different time scales. In Asia, most modeling studies with coupled
 1239 models targeted the impacts of aerosol feedbacks on surface PM_{2.5} and O₃ concentrations, with only
 1240 few focusing on other gaseous pollutants.

1241 Simulation results showed that turning on aerosol feedbacks in coupled models generally made
 1242 PM_{2.5} concentrations increased in different regions of Asia at various time scales, which stemmed
 1243 from decrease of shortwave radiation, T2, WS10 and PBLH and increase of RH2. Some studies did
 1244 show negative impacts of aerosol effects on hourly, daily, and seasonal PM_{2.5} at some areas that
 1245 could be attributed to ACI effects, changes in transport and dispersion patterns, reductions in
 1246 humidity levels and secondary aerosol formations (Zhang B. et al., 2015; Zhan et al., 2017; Yang et
 1247 al., 2017; Wang K. et al., 2018). Similar to the perturbations of surface meteorological variables due
 1248 to aerosol effects, the hourly PM_{2.5} variations and the range were the largest compared to those at
 1249 other time scales. The largest PM_{2.5} increases were reported in NCP, SEC, EA, SEA and PRD at the
 1250 hourly, daily, monthly, seasonal and yearly time scales with average values of 23.48 µg·m⁻³, 14.73
 1251 µg·m⁻³, 16.50 µg·m⁻³, 1.12 µg·m⁻³ and 2.90 µg·m⁻³, respectively.

1252 In addition to PM_{2.5}, gaseous pollutants (O₃, NO₂, SO₂, CO and NH₃) are impacted by ARI
 1253 or/and ACI effects as well. As shown in Table 8, general reductions of ozone concentrations were
 1254 reported in Asia across all the modeling domains and time scales based on coupled models'
 1255 simulations. However, the influences of aerosol feedbacks on atmospheric dynamics and stability,
 1256 and photochemistry (photolysis rate and ozone formation regimes) could make ozone concentrations
 1257 increase somewhat in summer months or during wet season (Jung et al., 2019; Nguyen et al., 2019b;
 1258 Xing et al., 2017). The largest hourly, daily, monthly, seasonal, and annual variations of O₃ occurred
 1259 in YRD (-32.80 µg·m⁻³), EC (-5.97 µg·m⁻³), China (-23.90 µg·m⁻³), EA (-4.48 µg·m⁻³) and EA (-
 1260 2.76 µg·m⁻³), respectively. Along with reduced O₃ due to ARI or/and ACI, NO₂ concentrations were
 1261 enhanced with average changes of +12.30 µg·m⁻³ (YRD) at the hourly scale and +0.66 µg·m⁻³ (EA)
 1262 at both the seasonal and yearly scales, which could be attributed to slower photochemical reactions,
 1263 strengthened atmospheric stability and O₃ titration (Nguyen et al., 2019b). Regarding other gaseous
 1264 pollutants, limited studies pointed out daily and annual SO₂ concentrations increased in NEA and

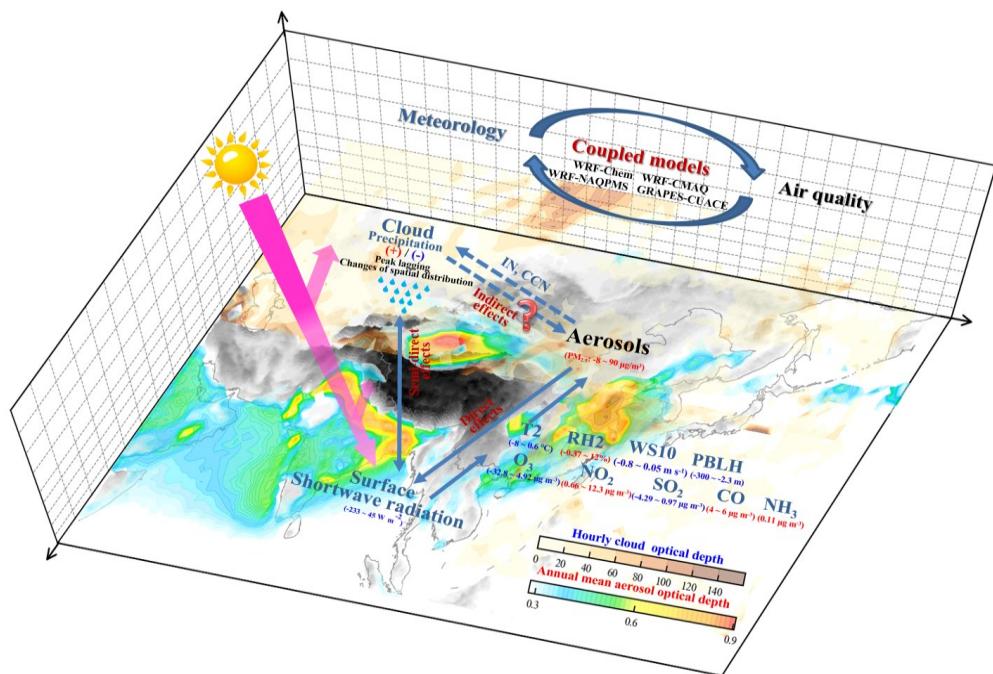
1265 EA due to lower PBLH induced by the ARI effects of anthropogenic aerosols (Jung et al.,2019;
 1266 Nguyen et al., 2019b). The seasonal SO₂ reduction was rather large, which related to higher PBLH
 1267 induced by the ACI effects of dust aerosols in the NCP area of EA (Wang K. et al., 2018). The slight
 1268 increase of seasonal SO₂ was reported in the whole domain of EA due to lower PBLH caused by
 1269 ARI effects of anthropogenic aerosols (Nguyen et al., 2019b). There was only one study depicted
 1270 increased CO (NH₃) concentration in EC (NEA) due to both the ARI and ACI (ARI) effects of
 1271 anthropogenic aerosols but these results may not be conclusive.

1272

1273 Table 8. Compilation of aerosol-induced variations of PM_{2.5} and gaseous pollutants simulated by two-way
 1274 coupled models (WRF-Chem, WRF-CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) in
 1275 different regions of Asia and at different temporal scales.

Region	Time scale	ΔPM _{2.5} [mean] (μg·m ⁻³)	ΔO ₃ [mean] (μg·m ⁻³)	ΔNO ₂ [mean] (μg·m ⁻³)	ΔSO ₂ [mean] (μg·m ⁻³)	ΔCO [mean] (μg·m ⁻³)	ΔNH ₃ [mean] (μg·m ⁻³)
NCP	hours	-3.50 to 90.00 [23.48]					
YRD	hours	7.00 to 30.50 [15.17]	-32.80 to -0.20 [-11.25]	12.30			
Hourly mean		19.32	-11.25	12.30			
SEC	days	-1.91 to 32.49 [14.73]					
NCP	days	-5.00 to 56.00 [14.51]					
EC	days	2.87 to 18.60 [10.74]	-5.97 to -1.45 [-3.71]				
NEA	days	1.75			0.97		0.11
Daily mean		10.43	-3.71		0.97		0.11
India	months	3.00 to 30.00 [16.50]					
EC	months	1.00 to 40.00 [16.33]	-2.40 to -1.00 [-1.70]			4.00 to 6.00 [5.00]	
China	months	1.60 to 33.20 [14.38]	-23.90 to 4.92 [-3.42]				
EA	months	3.60 to 10.20 [5.79]					
Monthly mean		13.25	-2.56			5.00	
SEA	seasons	0.15 to 2.09 [1.12]	-1.92 to 0.26 [-0.83]				
EA	seasons	-8.00 to 2.70 [-0.14]	-4.48 to -1.00 [-2.99]	0.43 to 0.88 [0.66]	-4.29 to 0.72 [-0.42]		
Seasonal mean		0.49	-1.91	0.66	-0.42		
PRD	years	2.90					
EA	years	1.82	-2.76	0.66	0.54		
NCP	years	0.10 to 5.10 [1.70]					
SEA	years	1.21	-0.80				
Yearly mean		1.91	-1.78	0.66	0.54		

1276



1278
 1279 Figure 9. A schematic diagram depicting aerosol-radiation-cloud interactions and quantitative effects of aerosol
 1280 feedbacks on meteorological and air quality variables simulated by two-way coupled models in Asia.
 1281

1282 Two-way coupled models have been applied in US and Europe extensively and then in Asia
 1283 due to frequent occurrences of severe air pollution events accompanied with rapid economic growth
 1284 in the region. Until now, no comprehensive study is conducted to elucidate the recent advances in
 1285 two-way coupled models' applications in Asia. This paper provides a critical overview of current
 1286 status and research focuses of related modeling studies using two-way coupled models in Asia
 1287 between 2010 and 2019, and summarizes the effects of aerosol feedbacks on meteorological and air
 1288 quality variables from these studies.

1289 Through systematically searching peer-reviewed publications with several scientific-based
 1290 search engines and a variety of key word combinations and applying certain selection criteria, 160
 1291 relevant papers were identified. Our bibliometric analysis results (as schematically illustrated in Fig.
 1292 9) showed that in Asia, the research activities with two-way coupled models had increased gradually
 1293 in the past decade and the five two-way coupled models (WRF-Chem, WRF-CMAQ, WRF-
 1294 NAQPMS, GRAPES-CUACE and GATOR-GCMOM) were extensively utilized to explore the ARI
 1295 or/and ACI effects in Asia with focusing on several high aerosol loading areas (e.g., EA, India, China
 1296 and NCP) during wintertime or/and severe pollution events, with less investigations looking into
 1297 other areas and seasons with low pollution levels. Among the 160 papers, nearly 82 % of them
 1298 focused on ARI (72 papers) and both ARI and ACI effects (60 papers), but papers that only
 1299 considering ACI effects were relatively limited. The ARI or/and ACI effects of natural mineral dust,
 1300 BC and BrC from anthropogenic sources and BC from ARB were mostly investigated, while a few
 1301 studies quantitatively assessed the health impacts induced by aerosol effects.

1302 Meta-analysis results revealed that enabling aerosol effects in two-way coupled models could
 1303 improve their simulation/forecast capabilities of meteorology and air quality in Asia, but a wide
 1304 range of differences occurred among the previous studies perhaps due to various model
 1305 configurations (selections of model versions and parameterization schemes) and largest
 1306 uncertainties related to ACI processes and their treatments in models. Compared to US and Europe,
 1307 the aerosol-induced decrease of the shortwave radiative forcing was larger because of higher air
 1308 pollution levels in Asia. The overall decrease (increase) of T2, WS10, PBLH and O₃ (RH2, PM_{2.5}
 1309 and other gaseous pollutant concentrations) caused by ARI or/and ACI effects were reported from
 1310 the modeling studies using two-way coupled models in Asia. The ranges of aerosol-induced
 1311 variations of T2, PBLH, PM_{2.5} and O₃ concentrations were larger than other meteorological and air
 1312 quality variables. For variables of CO, SO₂, NO₂, and NH₃, reliable estimates could not be obtained
 1313 due to insufficient numbers of samples in past studies.

Even though noticeable progresses toward the application of two-way coupled meteorology and air quality models have been made in Asia and the world during the last decade, several limitations are still presented. Enabling aerosol feedbacks lead to higher computational cost compared to offline models, but this shortcoming can be overcome with the new developments of cluster computing technology (i.e., Graphics Processing Unit (GPU)-accelerated computing and cloud computing). The latest advances in the measurements and research of cloud properties, precipitation characteristics, and physiochemical characteristics of aerosols that play pivotal roles in CCN or IN activation mechanisms can guide the improvements and enhancements in two-way coupled models, especially to abate the uncertainties in simulating ACI effects. Special attention needs to be paid to assess the accuracies of different methodologies in terms of ARI and ACI calculations in two-way coupled models in Asia and other regions. Besides the five two-way coupled models mentioned in this paper, more models capable of simulating aerosol feedbacks (such as WRF-CHIMERE and WRF-GEOS-Chem) have become available and projects covering more comprehensive intercomparisons of these coupled models should be conducted in Asia. Future assessments of the ARI or/and ACI effects should pay extra attention to their impacts on dry and wet depositions simulated by two-way coupled models. So far, the majority of two-way coupled models' simulations and evaluations focuses on episodic air pollution events occurring in certain areas, therefore their long-term applications and evaluations are necessary and their real-time forecasting capabilities should be explored as well.

Appendix A

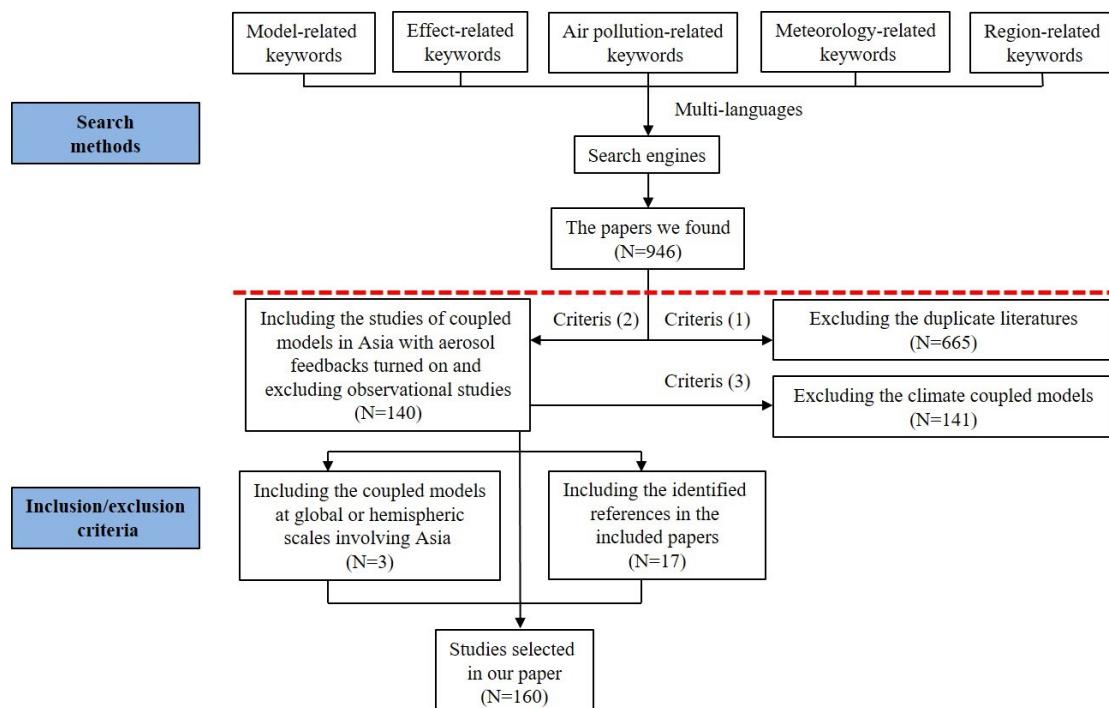


Figure A1. Flowchart of literature search and identification

Appendix B

Table B1. Lists of abbreviations and acronyms

ACI	Aerosol-cloud interactions
AOD	Aerosol optical depth
AQCHEM	the CMAQ's standard aqueous chemistry module
ARB	Agriculture residue burning
ARB_BC	BC emitted from agriculture residue burning
ARB_CAs	Carbonaceous aerosols emitted from agriculture residue burning
ARI	Aerosol-radiation interactions
ATM	In the atmosphere
BB	Biomass burning
BC	Black carbon
BCs	Boundary conditions
BOT	At the bottom

BrC	Brown carbon
CA	Central Asia
CAMx	Comprehensive Air quality Model with extensions
CAs	Carbonaceous aerosols
CC	Central China
CCN	Cloud condensation nuclei
CDNC	Cloud droplet number concentration
CHIMERE	A multi-scale chemistry-transport model for atmospheric composition analysis and forecast
CMAQ	Community Multiscale Air Quality model
CO	Carbon monoxide
CRFs	Concentration-response functions
DRF	Direct radiative forcing
EA	East Asia
EC	East China
EQUISOLV II	the EQUilibrium SOLVer version 2
GATOR-GCMOM	Gas, aerosol, transport, radiation, general circulation, mesoscale, and ocean Model
GOCART	The Global Ozone Chemistry Aerosol Radiation and Transport
GPRAPES-CUACE	Global-regional assimilation and prediction system coupled with the Chinese Unified Atmospheric Chemistry
GSI	Environment forecasting system
H_2O_2	Gridpoint Statistical Interpolation
HNO_3	Hydrogen peroxide
HO_2^{\cdot}	Nitric acid
ICs	Hydroperoxyl
IN	Initial conditions
INPs	Ice nuclei
IPCC	Ice nucleation parameterizations
IPR	Intergovernmental Panel on Climate Change
IWP	Ice particle radius
IWP	Ice water path
LWP	Liquid water path
LWRF	Longwave radiative forcing
MARS-A	the Model for an Aerosol Reacting System-version A
MB	Mean bias
ME	Middle East
MESA-MTEM	the Multicomponent Equilibrium Solver for Aerosols with the Multicomponent Taylor Expansion Method
MICS-Asia	Model Inter-Comparison Study for Asia
MOZART	Model for Ozone and Related Chemical Tracer
MRYR	Middle reaches of the Yangtze River
N	Nitrate
N_2O_5	Nitrogen pentoxide
NAQPMS	Nested Air Quality Prediction Modeling System
NC	North China
NCP	North China Plain
NEA	Northeast Asia
NME	Normalized mean error
NO_2	Nitrogen dioxide
NU-WRF	National aeronautics and space administration Unified Weather Research and Forecasting model
NWC	Northwest China
O_3	Ozone
OA	Organic aerosols
OC	Organic carbon
$\cdot\text{OH}$	Hydroxyl radical
OPAC	Optical Properties of Aerosols and Clouds
PBL	Planetary boundary layer
PBLH	Planetary boundary layer height
$\text{PM}_{2.5}$	Fine particulate matter
PRD	Pearl River Delta
PSI	Papers with statistical indices
R	Correlation coefficient
RADM	the Regional Acid Deposition Mode
RH2	Relative humidity at 2 meters above the surface
RMSE	Root mean square error
RRTM	The Rapid Radiative Transfer Model
RRTMG	The Rapid Radiative Transfer Model for General Circulation Models
S	Sulfate
SA	South Asia
SC	South China
SEA	Southeast Asia
SEC	Southeast China
SH2	Specific humidity at 2 meters above the surface
SI	Statistical indices
SO_2	Sulfur dioxide
SOA	Secondary organic aerosol
SWC	Southwest China
SWRF	Shortwave radiative forcing
T2	Air temperature at 2 meters above the surface
TOA	At the top of atmosphere
TP	Tibetan Plateau
US	the United States
VBS	Volatility basis set

WA	West Asia
WRF	Weather Research and Forecasting model
WRF-Chem	Weather Research and Forecasting model coupled with Chemistry
WRF-CHIMERE	Weather Research and Forecasting model coupled with a multi-scale Chemistry-Transport Model (CTM) for air quality forecasting and simulation
WRF-CMAQ	Weather Research and Forecasting model coupled with Community Multiscale Air Quality model
WRF-NAQPMS	Weather Research and Forecasting model coupled with the Nested Air Quality Prediction Modeling System
WS10	Wind speed at 10 meters above the surface
YRD	Yangtze River Delta

1341

1343

1344 Table B2. The compiled number of publications (NP) and number of samples (NS) for papers that providing
1345 statistical indices (SI) of meteorological variables.

No.*	Meteorological variables																
	T2				RH2				SH2				WS10				
	NP	NS			NP	NS			NP	NS			NP	NS			
		NP	R	MB	RMSE	NP	R	MB	RMSE	NP	R	MB	RMSE	NP	R	MB	RMSE
4	1	5	5 (4↑, 1↓)	5		1	5	5 (1↑, 4↓)	5								
5						1		3 (2↑, 1↓)	3								
7	1	4	4 (3↑, 1↓)			1 (1↑)											
13	1		1 (1↑)				1		1 (1↑)								
15	1	1					1	1									
16	1	1															
20	1	2	2 (1↑, 1↓)	2		1	2	2 (1↑, 1↓)	2								
21	1	0	2 (2↓)	2													
22	1	1	1 (1↓)	1		1	1	1 (1↑)	1								
23	1	1	1 (1↑)			1	1	1 (1↓)									
24	1	1	1 (1↑)			1	1	1 (1↓)									
25	1	1	1 (1↓)														
28	1		1 (1↑)	1				1 (1↓)	1								
29	1	9	9 (6↑, 3↓)	9		1	8			9							
33	1	6	6 (4↑, 2↓)	6													
34	1	2	2 (2↑)	2													
35	1	2		2		1	1		1								
38	1		4 (4↓)	4		1		4 (3↑, 1↓)	4								
50	1		8 (8↓)	8													
56	1	1	1 (1↓)	1		1	1	1 (1↓)	1								
57	1	1				1	1										
61	1	4	4 (4↓)	4		1	4	4 (4↑)	4								
62	1		5 (5↓)	5													
63	1	1															
71	1	1															
72	1	4	4 (3↑, 1↓)	4		1	4	4 (3↑, 1↓)	4								
73	1	1	1 (1↓)	1													
75	1	4	4 (4↑)			1	4	4 (4↑)									
77	1	4	4 (2↑, 2↓)							1	4	3 (3↑)	4	1	4	4 (4↑)	4
79	1		8 (6↑, 2↓)	8													
80	1	8	8 (8↑)	8		1	8	8 (8↓)	8						1	8	8 (6↑, 2↓)
85	1		4 (1↑, 3↓)	4		1		4 (2↑, 2↓)	4						1	4 (4↑)	4
87	1		3 (2↑, 1↓)	3											1	3 (2↑, 1↓)	3
88	1	3	3 (1↑, 2↓)	3		1	3	3 (2↑, 1↓)	3						1	3 (2↑, 1↓)	3
90	1	4	4 (1↑, 3↓)							1	4	4 (4↑)		1	4	4 (4↑)	4
91	1	1	1 (1↓)	1						1	1	1 (1↑)		1	1	1 (1↑)	1
94	1	6	6 (4↑, 2↓)	6		1	6	6 (2↑, 4↓)	6						1	6 (6↑)	6
96	1	16	16 (11↑, 5↓)												1	16 (11↑, 5↓)	
97	1	1	1 (1↓)	1		1	1	1 (1↑)	1						1	1 (1↑)	1
106	1	6	6 (6↓)							1	6	5 (2↑, 3↓)		1	6	6 (6↑)	
109	1	2	2 (2↓)	2		1	3	3 (3↑)	3						1	2	2 (2↑)
112	1		2 (2↓)	2						1		2 (2↓)	2	1		2 (2↑)	2
116	1	2	2 (1↑, 1↓)	0		1	2	2 (1↑, 1↓)							1	1	1 (1↑)
121	1	1	1 (1↓)	1											1	2 (2↑)	2
122	1		2 (2↓)	2		1		2 (2↑)	2						1	4 (4↑)	4
125	1	4	4 (4↓)	4		1	4	4 (4↑)	4						1	4 (4↑)	4
126	1	4	4 (4↓)	4						1	4	4 (2↑, 2↓)	4	1	4	4 (4↑)	4
127	1		2 (2↓)	2											1	2 (2↑)	2
128	1	8	8 (8↑)	8						1	8	8 (5↑, 3↓)	8	1	8	8 (8↑)	8
129	1	1	1 (1↓)	1		1	1	1 (1↑)	1						1	1 (1↑)	1
133	1		1 (1↓)	0		1		4 (4↑)							1	4 (3↑, 1↓)	
143	1	4		4		1	4		4						1	4	4
147	1	2		2		1	2		2						1	2	2
151	1	7	7 (7↓)	7						1	7	7 (3↑, 4↓)	7	1	7	7 (7↓)	7
Total		53	137	167 (67↑, 100↓)	130	30	68	70 (42↑, 28↓)	73	9	35	35 (21↑, 14↓)	27	40	111	126 (104↑, 22↓)	97

1346 Note that the No.* is consistent with the No. in Table 1, and ↑ and ↓ mark over- and underestimations of variables, respectively, along with
1347 their number of samples.

1348

1349 Table B3. The compiled number of publications (NP) and number of samples (NS) for papers that providing
1350 statistical indices (SI) of air quality variables.

No.*	Air quality variables											
	PM _{2.5}				O ₃				SO ₂			
	NP	R	MB	RMSE	NP	R	MB	RMSE	NP	R	MB	RMSE
4	1	5	5 (5↓)	5								
5	1		1 (1↑)	1	1				1 (1↓)	1		
11	1	60										
15	1	1										
21	1		2 (1↑, 1↓)									
22	1	1	1 (1↑)	1								
23	1	1	1 (1↑)	1					1 (1↓)	1		
24	1	1	1 (1↓)	1						1 (1↓)	1	
25	1	1	1 (1↑)	1						1 (1↑)	1	
29	1	9	9 (6↑, 3↓)	9								

33	1	4	4 (4↓)	4	1	4	4 (3↑, 1↓)	4
34	1	2	2 (1↑, 1↓)	2	1	1		
35								
50	1		4 (1↑, 3↓)	4				
56	1	1	1 (1↑)	1				
57	1	1						
59	1	6	6 (6↓)	6	1	6	6 (6↑)	6
61	1	12	12 (12↑)	12				
67	1	10	2 (2↓)	10				
71	1	1						
73	1	2	2 (1↑, 1↓)		1	4	4 (4↑)	
77	1	4						
85	1	3	3 (3↓)					
86	1	4	4 (2↑, 2↓)	4				
88	1	3	3 (1↑, 2↓)	3				
90	1	8	8 (2↑, 6↓)		1	14	14 (14↑)	
91	1	4	4 (1↑, 3↓)	4	1	6	6 (4↑, 2↓)	6
94	1	4	4 (3↑, 1↓)	4				
97	1	1	1 (1↓)	1				
100	1	1			1	1		
106	1	6	6 (2↑, 4↓)		1	8	8 (4↑, 4↓)	
112	1				1			
121					1			5
122	1	4	4 (1↑, 3↓)					
125	1	4	4 (2↑, 2↓)	4	1	4	4 (4↑)	4
126	1	4	4 (2↑, 2↓)	4	1	4	4 (4↑)	4
127	1		1 (1↑)	1				
128	1	8	8 (3↑, 5↓)	8				
129	1	3	3 (2↑, 1↓)	3	1	2	2 (1↑, 1↓)	2
133					1	4	4 (3↑, 1↓)	4
136	1	5	5 (5↓)					
146	1	1			1	20		20
147	1	2		2				
149	1	6		6				
150					1	21		21
151	1	12	6 (6↑)	6	1	24	12 (7↑, 5↓)	12
Total	42	205	122 (55↑, 67↓)	105	21	125	72 (55↑, 17↓)	90

1351 Note that the No.* is consistent with the No. in Table 1, and ↑ and ↓ mark over- and underestimations of variables, respectively, along with
 1352 their number of samples.

1353

1354 Table B4. The compiled number of publications (NP) and number of samples (NS) for papers that simultaneously
 1355 providing the statistical indices (SI) of meteorological variables simulated by coupled models (WRF-Chem, WRF-
 1356 CMAQ, GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) with/out ARI.

No.*	Meteorological variables											
	T2			RH2			SH2			WS10		
	NP	NS		NP	NS		NP	NS		NP	NS	
		R	MB		R	MB		R	MB		R	MB
32	1	3	3 (2↑, 1↓)	3								
78	1		4 (3↑, 1↓)	4								
124	1	2	2 (2↓)	2	1	2	2 (2↑)	2			1	2
125	1	2	2 (2↓)	2				1	2	2 (1↑, 1↓)	2	2 (2↑)
126	1		1 (1↓)	1							1	1 (1↑)
127	1	4	4 (4↓)	4				1	4	4 (3↑, 1↓)	4	4 (4↑)
146	1	1		1	1	1		1			1	1
Total	7	12	16 (5↑, 11↓)	17	2	3	2 (2↑)	3	2	6	6 (4↑, 2↓)	6

1357 Note that the No.* is consistent with the No. in Table 1, and ↑ and ↓ mark over- and underestimations of variables, respectively, along with
 1358 their number of samples.

1359

1360 Table B5. The compiled number of publications (NP) and number of samples (NS) for papers that simultaneously
 1361 providing the statistical indices (SI) of air quality variables simulated by coupled models (WRF-Chem, WRF-CMAQ,
 1362 GRAPES-CUACE, WRF-NAQPMS and GATOR-GCMOM) with/out ARI.

No.*	Air quality variables											
	PM _{2.5}			O ₃								
	NP	NS		NP	NS		NP	NS		NP	NS	
		R	MB		MB	RMSE		R	MB		R	MB
49	1			2 (1↑, 1↓)	2		1		10			10
60	1		4	4 (4↑)	4							
124	1		2	2 (1↑, 1↓)	2		1		2	2 (2↑)	2	
125	1		2	2 (1↑, 1↓)	2		1		2	2 (2↑)	2	
127	1		4	4 (2↑, 2↓)	4							
146	1		1			1						
Total	5	13		14 (9↑, 5↓)	15		3	14		4 (4↑)	14	

1363 Note that the No.* is consistent with the No. in Table 1, and ↑ and ↓ mark over- and underestimations of variables, respectively, along with
 1364 their number of samples.

1365

1366 Table B6. Description of refractive indices and radiation schemes used in the WRF-Chem and WRF-CMAQ in Asia.

Model	Refractive indices of aerosol species groups			Radiation scheme		
	SW	LW	SW scheme (Spectral intervals)	LW	SW scheme (Spectral intervals)	
WRF-Chem	1. Water (1.35+1.524 ⁸ i,	1. Water (1.532+0.336i,	GODDARD (0.175-0.225, 0.225-0.245, 0.245-	RRTMG (10-350, 350-500, 500-630, 630-700,		

1.34+2.494 ⁹ i, 1.33+1.638 ⁹ i, 1.33+3.128 ⁹ i,	1.524+0.360i, 1.420+0.426i, 1.274+0.403i, 1.161+0.321i,	0.260, 0.280-0.295, 0.295-0.310, 0.310-0.320, 0.325-0.400, 0.400-0.700, 0.700-1.220, 1.220-	700-820, 820-980, 980-1080, 1080-1180, 1180- 1390, 1390-1480, 1480-1800, 1800-2080, 2080- 2250, 2250-2390, 2390-2600, 2600-3250 cm ⁻¹
2. Dust (1.55+0.003i, 1.550+0.003i, 1.550+0.003i, 1.550+0.003i)	1.142+0.115i, 1.232+0.047i, 1.266+0.039i, 1.296+0.034i, 1.321+0.034i,	2.270, 2.270-10.00 μ m RRTMG (3.077-3.846, 2.500-3.077, 2.150-2.500, 1.942-2.150, 1.626-1.942, 1.299-1.626, 1.242- 1.299, 0.778-1.242, 0.625-0.778, 0.442-0.625,	
3. BC (1.95+0.79i, 1.95+0.79i, 1.95+0.79i, 1.95+0.79i)	1.342+0.092i, 1.315+0.012i, 1.330+0.013i, 1.339+0.01i,	0.345-0.442, 0.263-0.345, 0.200-0.263, 3.846- 12.195 μ m	
4. OC (1.45+0i, 1.45+0i, 1.45+0i, 1.45+0i)	1.350+0.0049i, 1.408+0.0142i)		
5. Sea salt (1.51+8.66 ⁹ i, 1.5+7.019 ⁹ i, 1.5+1.184 ⁹ i, 1.47+1.54 ⁹ i)	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)		
6. Sulfate (1.52+1.00 ⁹ i, 1.52+1.00 ⁹ i, 1.52+1.00 ⁹ i, 1.52+1.75 ⁹ i) in term of 4 spectral intervals in 0.25- 0.35, 0.35-0.45, 0.55-0.65, 0.998-1.000 μ m	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, 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+0.01i)		
	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 term 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 cm ⁻¹		
1. Water (1.408+1.420 ⁹ i, 1.324+1.577 ⁹ i, 1.277+1.516 ⁹ i, 1.302+1.159 ⁹ i, 1.312+2.360 ⁹ i, 1.321+1.713 ⁹ i, 1.323+2.425 ⁹ i, 1.327+3.125 ⁹ i, 1.331+3.405 ⁹ i, 1.334+1.639 ⁹ i, 1.340+2.955 ⁹ i, 1.349+1.635 ⁹ i, 1.362+3.350 ⁹ i, 1.260+6.220 ⁹ i)	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 (1.443+5.718 ⁹ i, 1.420+1.777 ⁹ i, 1.420+1.060 ⁹ i, 1.420+8.368 ⁹ i, 1.463+1.621 ⁹ i, 1.510+2.198 ⁹ i, 1.510+1.929 ⁹ i, 1.520+1.564 ⁹ i, 1.530+7.000 ⁹ i, 1.530+5.666 ⁹ i, 1.530+1.530+5.000 ⁹ i, 1.530+8.440 ⁹ i, 1.530+3.000 ⁹ i, 1.710+1.100 ⁹ i)	2. Water-soluble (1.570+0.069i, 1.700+0.055i, 1.890+0.128i, 2.233+0.334i, 1.220+0.066i)		
WRF-CMAQ	3. BC (1.570+2.200i, 1.700+2.200i, 1.890+2.200i, 2.233+2.200i, 1.220+2.200i)	RRTMG (3.077-3.846, 2.500-3.077, 2.150-2.500, 1.942-2.150, 1.626-1.942, 1.299-1.626, 1.242- 1.299, 0.778-1.242, 0.625-0.778, 0.442-0.625,	RRTMG (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 cm ⁻¹)
	4. Insoluble (1.482+0.096i, 1.600+0.107i, 1.739+0.162i, 1.508+0.117i, 1.175+0.042i)	0.345-0.442, 0.263-0.345, 0.200-0.263, 3.846- 12.195 μ m	
	5. Sea-salt (1.410+0.019i, 1.490+0.014i, 1.560+0.017i, 1.600+0.029i, 1.402+0.012i)		
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, 2.589+1.771i)	in term of 5 thermal windows at 13.240, 11.20, 9.73, 8.870, 7.830 μ m		

4. Insoluble (1.272+1.165²i, 1.168+1.073²i, 1.208+8.650⁰i, 1.253+8.092³i, 1.329+8.000³i, 1.418+8.000⁰i, 1.456+8.000³i, 1.518+8.000³i, 1.530+8.000⁰i, 1.530+8.000³i, 1.530+8.440⁰i, 1.530+3.000²i, 1.470+9.000²i)
5. Sea-salt (1.480+1.758³i, 1.534+7.462³i, 1.437+2.950⁰i, 1.448+1.276³i, 1.450+7.944⁴i, 1.462+5.382⁴i, 1.469+3.754⁴i, 1.470+1.498⁴i, 1.490+2.050⁷i, 1.500+1.184⁸i, 1.502+9.938³i, 1.510+2.060⁶i, 1.510+5.000⁶i, 1.510+1.000⁷i) in term 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 μm

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Table B7. Summary of normalized mean error (NME) (%) of surface meteorological and air quality variables using two-way coupled models (WRF-Chem and WRF-CMAQ).

T2	SH2	RH2	WS10	PM _{2.5}	O ₃	PM _{2.5} with ARI (ARI) or without ARI (NO)	O ₃ with ARI (ARI) or without ARI (NO)	Model	Region	Reference
0.80, 0.60, 0.60, 0.60	19.10, 16.50, 10.00, 10.10	58.90, 41.60, 44.90, 49.50	37.31, 37.61, 35.77, 34.69, 35.34, 35.41, 45.22, 44.33, 43.09, 39.29, 39.49, 39.07	23.60, 38.50, 55.70, 39.80	37.61, 35.34, 44.33, 39.49 (ARI) 35.77, 35.41, 43.09, 39.07 (NO)			WRF-Chem	EA	Liu X. et al. (2016)
270.20, 22.30, 12.50, 17.60				44.99, 29.55, 37.28				WRF-Chem	EA	Zhang et al. (2018)
15.50, 15.80, 13.90, 9.90	10.40, 10.40, 9.90, 9.90	31.30, 31.30, 32.50, 32.50	49.80, 65.30, 49.80, 65.60, 88.30, 56.90, 88.40, 57.00	127.00, 32.20, 25.40, 126.10, 32.10, 25.00, 79.90, 25.80, 21.40, 45.80, 77.90, 25.60, 21.10, 39.50			WRF-Chem	EA	Zhang Y. et al. (2015a)	
14	11	32	52.70, 58.00, 104.70, 62.00	87.50, 28.60, 23.30, 52.90, 32.40, 28.20			WRF-Chem	EA	Chen Y. et al. (2015)	
-0.48, 0.19, 0.21, 0.05, 0.08, 0.13, 0.05, 0.04, 0.04, 0.05, 0.02, 0.02, 0.06, 0.05, 0.04, 0.02, 16.60, 10.50, 8.90, 12.90, 10.50, 10.20, 6.52, 6.58	0.33, 1.92, 0.71, 0.78, 0.28, 1.72, 0.61, 0.64, 0.24, 1.76, 0.00, 0.45, 0.34, 1.29, 0.44, 0.56						WRF-Chem	NCP	Chen D. et al. (2015)	
15.76, 12.15	112.28, 97.26			36.00, 33.00	31.00, 22.00			WRF-Chem	EA	Wang K. et al. (2018)
				44.00, 44.60, 40.10, 54.30				WRF-Chem	NEA	Park et al. (2018)
				41.48, 41.00, 51.77, 55.70	26.68, 26.71, 34.43, 34.64	41.00, 55.70 (ARI) 41.48, 51.77 (NO)	26.71, 34.64 (ARI) 26.68, 34.43 (NO)	WRF-CMAQ	SEA	Gao M. et al. (2015)
				37.99, 35.06, 38.59, 35.44, 34.39				WRF-CMAQ	China	Nguyen et al. (2019b)
										Chang (2018)

1370

1371 Appendix C

1372 C1 Comparisons of SI at different temporal scales for meteorology

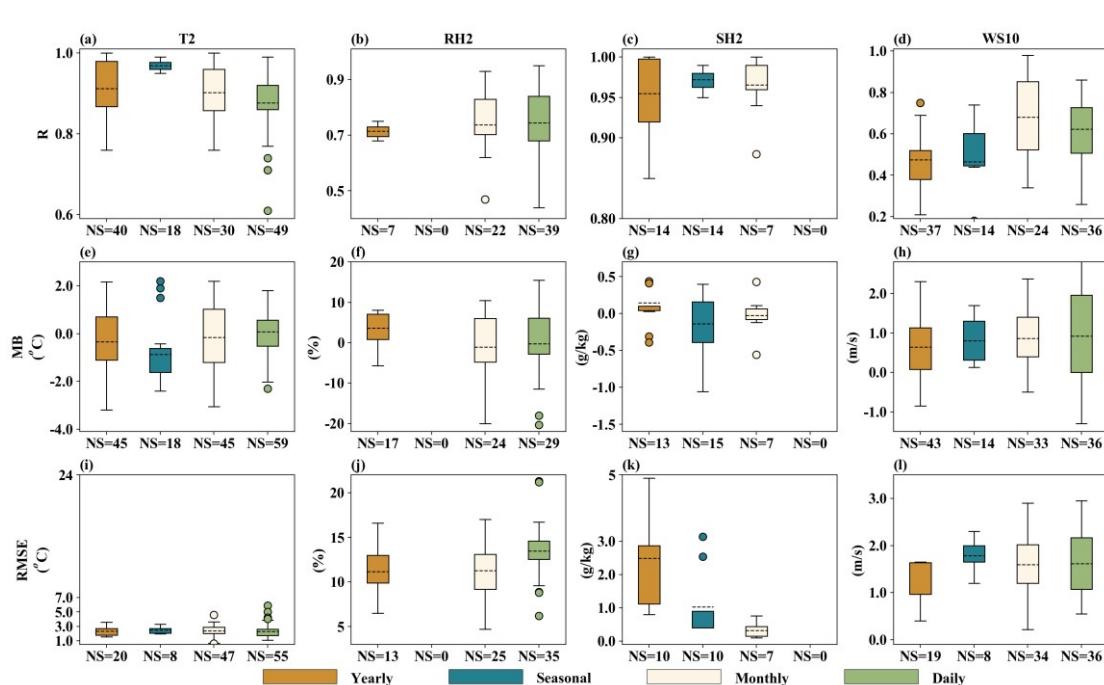
1373 To probe the model performance of simulated T2, RH2, SH2 and WS2 at different temporal
1374 scales, the SI of these meteorological variables from PSI were grouped according to the simulation
1375 time (yearly, seasonal, monthly and daily) and plotted in Figure C1. Note that the seasonal results
1376 contained SI values from simulations lasting more than one month and less than or equal to 3 months.
1377 Here in Figure C1, NP and NS were the number of PSI and samples with SI at different time scales,
1378 respectively, and also their total values were the same as the ones listed in Table S2. The correlation
1379 between simulated and observed T2 (Figure C1a) at the seasonal (mean R= 0.97 with the smallest
1380 sample size), yearly (0.91) and monthly (0.90) scales were stronger than that at the daily scale (0.87),
1381 indicating that long-term simulations of T2 were well reproduced by coupled models. As shown in
1382 Figure C1e, T2 underestimation mentioned above (Fig. 3a) appeared also in the seasonal, monthly
1383 and yearly simulations (average MB = -0.87 $^{\circ}\text{C}$, -0.15 $^{\circ}\text{C}$ and -0.34 $^{\circ}\text{C}$, respectively), but the daily

1384 T2 were overestimated (average MB = 0.07 °C). It should be noted that T2 at the monthly scale was
 1385 underpredicted mainly during winter months (16 samples). Regarding the mean RMSE, its value
 1386 (Figure C1i) at the daily scale was the largest (0.97 °C) in comparison with that at the other temporal
 1387 scales.

1388 Given that no SI was available for RH2 at the seasonal scale, results at other time scales were
 1389 discussed here. Figure C1b presented that simulated RH2 at the daily scale had the best correlation
 1390 coefficient (mean $R = 0.74$), followed by those at the monthly (0.73) and yearly (0.71) scales. Except
 1391 overestimation (average MB = 3.6 %) at the yearly scale (Figure C1f), modeled RH2 were
 1392 underestimated at the monthly (average MB = -1.1 %) and daily (average MB = -0.2 %) scales,
 1393 respectively. Therefore, coupled models calculated RH2 reasonably well in short-term simulations.
 1394 However, at the daily scale, RMSE of modeled RH2 (Figure C1j) was relatively large fluctuation
 1395 ranging from 6.2 % to 21.3 %.

1396 Lacking of SI for SH2 at the daily scale, only those at other time scales were compared. Even
 1397 though NP and NS were very limited, the modeled SH2 (Figure C1c) exhibited especially good
 1398 correlation with observations with the mean R values exceeding 0.95 at the yearly, seasonal and
 1399 monthly scales (0.99, 0.97 and 0.96, respectively) but had the largest mean RMSE (2.09 g·kg⁻¹) at
 1400 the yearly scale (Figure C1k). Also, both over- and under-estimations of modeled SH2 (Fig. C1g)
 1401 were reported at different time scales with average MB values as 0.15 g·kg⁻¹, -0.02 g·kg⁻¹, and -0.14
 1402 g·kg⁻¹ for yearly, seasonal and monthly simulations, respectively. Generally, the long-term
 1403 simulations of SH2 agreed better with observations than the short-term ones.

1404 As seen in Figure C1d, the modeled WS10 at the monthly scale (mean $R = 0.68$) correlated
 1405 with observations better than that at the daily, yearly and seasonal scales (mean $R = 0.62, 0.48$ and
 1406 0.46, respectively). The simulations at all temporal scales tended to overestimate WS10 comparing
 1407 against observations (Figure C1h) and their average MB were 0.80 m·s⁻¹ (seasonal), 0.86 m·s⁻¹
 1408 (monthly), 0.64 m·s⁻¹ (yearly) and 0.62 m·s⁻¹ (daily), respectively. The short-term simulations of
 1409 WS10 better matched with observations compared to the long-term ones. At the same time, the
 1410 largest mean RMSE (1.79 m·s⁻¹) of simulated WS10 (Figure C1l) appeared at the seasonal scale.
 1411



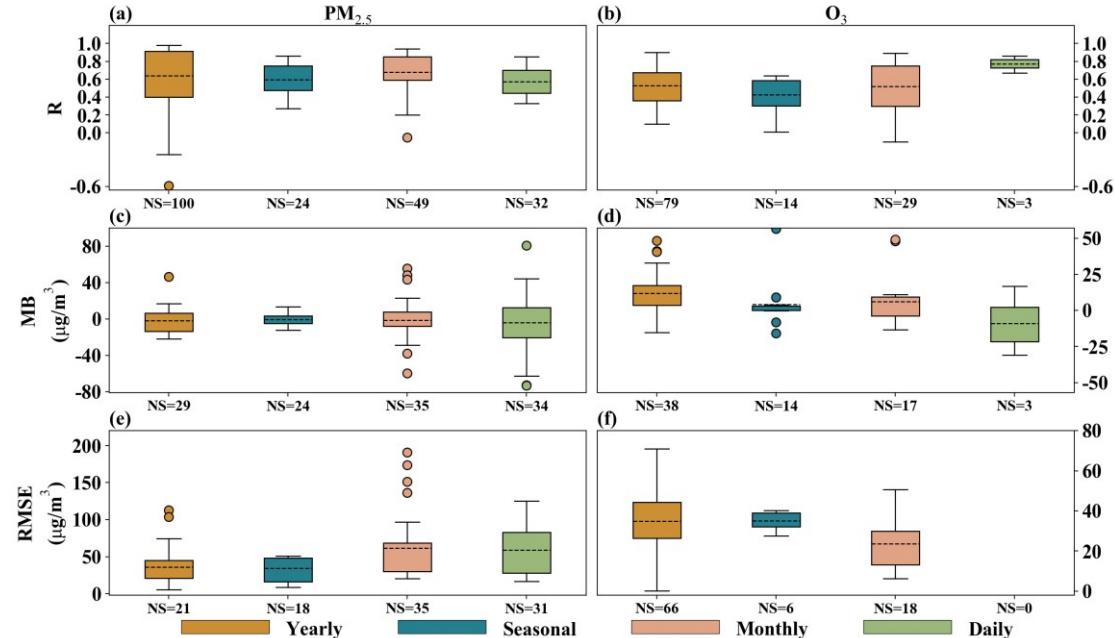
1412
 1413 Figure C1. The statistical indices of modeled meteorological variables at different temporal scales (Yearly, Seasonal,
 1414 Monthly and Daily) from past studies in Asia.
 1415

1416 C2 Comparisons of SI at different temporal scales for air quality

1417 Figure C2 depicted the SI of simulated PM_{2.5} and O₃ at yearly, seasonal, monthly and daily
 1418 scales. The correlation between simulated and observed PM_{2.5} (Figure C2a) at the monthly scale
 1419 (mean $R = 0.68$) was largest compared to those at the yearly (0.64), seasonal (0.59), daily (0.57)
 1420 scales. All the simulated PM_{2.5} were underestimated, with the average daily, monthly, seasonal, and

1421 yearly MB as -4.13 , -1.46 , -0.28 , and $-1.89 \mu\text{g}\cdot\text{m}^{-3}$, respectively (Figure C2c). As displayed in Figure
 1422 C2e, the mean RMSE at the monthly scale was the largest ($61.57 \mu\text{g}\cdot\text{m}^{-3}$).

1423 Regarding to correlation between simulated and observed O_3 (Figure C2b), it was the best at
 1424 the daily scale (mean $R=0.77$). Modeled O_3 were overestimated at the seasonal (average MB =
 1425 $+4.12 \mu\text{g}\cdot\text{m}^{-3}$), monthly (average MB = $+6.11 \mu\text{g}\cdot\text{m}^{-3}$) and yearly (average MB = $+11.71 \mu\text{g}\cdot\text{m}^{-3}$)
 1426 scales, but underestimated at the daily scale (average MB = $-8.89 \mu\text{g}\cdot\text{m}^{-3}$) (Figure C2d). Note that no
 1427 RMSE for O_3 simulation was available at the daily scale, and the RMSE at the yearly scale (Figure
 1428 C2f) had relatively large fluctuation ranging from 0.21 to $71 \mu\text{g}\cdot\text{m}^{-3}$. Therefore, coupled models
 1429 calculated O_3 matched well with observation in short-term simulations.



1430
 1431 Figure C2. The quantile distributions of simulated $\text{PM}_{2.5}$ and O_3 performance metrics at different temporal scales
 1432 from past studies in Asia.
 1433

1434 Data availability

1435 The related dataset can be downloaded from <https://doi.org/10.5281/zenodo.5571076> (Gao et
 1436 al., 2021), and this dataset includes basic information (Table S1), performance metrics (Table S2),
 1437 quantitative effects of aerosol feedbacks on meteorological and air quality variables (Table S3),
 1438 model configuration and setup (Table S4) and aerosol-induced variations of simulated shortwave
 1439 and longwave radiative forcing (Table S5) extracted from collected studies of applications of two-
 1440 way coupled meteorology and air quality models in Asia.

1441

1442 Author contribution

1443 Chao Gao, Aijun Xiu, Xuelei Zhang and Qingqing Tong carried out the data collection, related
 1444 analysis, figure plotting, and manuscript writing; Hongmei Zhao, Shichun Zhang, Guangyi Yang
 1445 and Mengduo Zhang involved with the original research plan and made suggestions to the
 1446 manuscript writing.

1447

1448 Competing interest

1449 The authors declare that they have no conflict of interest.

1450

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