

1 MICS-Asia III: Multi-model comparison of reactive Nitrogen deposition
2 over China

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44 **Abstract:** Atmospheric nitrogen deposition in China has attracted public attention in
45 recent years due to the increasing anthropogenic emission of reactive nitrogen (N_r)
46 and its impacts on the terrestrial and aquatic ecosystems. However, limited long-term
47 and multi-site measurements have restrained the understanding on the mechanism of
48 the N_r deposition as well as the chemical transport model (CTM) improvement. In this
49 study, the performance of the simulated wet and dry deposition for different N_r
50 species, i.e., particulate NO_3^- and NH_4^+ , gaseous NO_x , HNO_3 and NH_3 , have been
51 conducted using the framework of Model Inter-Comparison Study for Asia
52 (MICS-Asia) phase III. Nine Models, including 5 WRF-CMAQ models, 2
53 self-developed regional models, a global model and a RAMS-CMAQ model, have
54 been selected for the comparison. For wet depositions, observation data from 83
55 measurement sites of EANET, CREN, CAUDN, NADMN and DEE of China have
56 been collected and normalized to compare with model results. In general, most
57 models show the consistent spatial and temporal variation of both oxidized N (N_{ox})
58 and reduced N (N_{rd}) wet depositions in China with the NME around at 50%, which is
59 lower than the value of 70% based on EANET observation over Asia. Both the ratio
60 of wet or dry deposition to the total inorganic N deposition (TIN) and the ratios of
61 TIN to their emissions have shown consistent results with the NNDMN estimates.
62 The performance of ensemble results (ENM) was further assessed with satellite
63 measurements. In different regions of China, the results show that the simulated N_{ox}
64 wet deposition was overestimated in North East China (NE) but underestimated in
65 south of China (SE+SW), while the N_{rd} wet deposition was underestimated in all
66 regions by all models. The deposition of N_{ox} has larger uncertainties than the N_{rd}
67 especially in North China (NC), indicating the chemical reaction process is one of the
68 most important factors affecting the model performance. Compared to Critical Load
69 (CL) value, the N_r deposition in NC, SE and SW reached or exceeded reported CL
70 values and resulted in serious ecological impacts. The control of N_{rd} in NC and SW
71 and N_{ox} in SE would be effective mitigations for TIN deposition in these regions. The
72 N_r deposition in the Tibet plateau with a high ratio of TIN/emission (~ 3.0), indicates a
73 significant transmission from outside. Efforts to reduce these transmissions ought to
74 be a paramount goal, due the climatic importance of the Tibet region to sensitive
75 ecosystems throughout China.

76

77 **Keywords:** Nitrogen deposition, multi-model comparison, China, reduced nitrogen,
78 oxidized nitrogen

79 **1 Introduction**

80 Atmospheric Nitrogen (N) deposition is defined as N related gases and particles are
81 deposited via precipitation (wet deposition) and not via precipitation (dry deposition)
82 (Clark and Kremer, 2005). These deposits to the Earth's surface are either close to the
83 sources or in remote regions (e.g. chemical transformation and long-range transport of
84 oxidized and reduced N, N_{ox} and N_{rd} hereafter), located far from human activities and
85 labeled as the N-limited areas (Phoenix et al., 2006;Holtgrieve et al., 2011). Evidence
86 shows that the effects of reactive N ($N_r=N_{ox}+N_{rd}$) deposition to the environment are
87 numerous, including decreased biological diversity, increased soil acidification, and
88 lake eutrophication (Clark and Tilman, 2008;Janssens et al., 2010;Holtgrieve et al.,
89 2011;Phoenix et al., 2006;Galloway et al., 2004). Different human activities disturb
90 the natural N cycle in serious ways (Galloway et al., 2004), for example, using
91 artificial fertilizers to increase crop production (Erisman et al., 2008) or excessively
92 relying on fossil fuels for industrial production. N_r production increased from
93 approximately 15 Tg N yr⁻¹ in 1860 to 187 Tg N yr⁻¹ in 2005 and more than 50% of
94 that N_r has been reported to deposit onto the ground (Nicolas and Galloway, 2008). In
95 the past two decades, high rates of N_r deposition were widely documented in
96 developed countries, such as America (Fenn et al., 1998) and Europe (Dise and
97 Wright, 1995). Great efforts have been made to fight against these negative effects in
98 the USA and the N_{ox} deposition was decreased dramatically in recent years (Li et al.,
99 2016). However, the growing human demand for food and energy at a global scale has
100 resulted in increased emissions of N_r into the environment (Galloway et al., 2008),
101 particularly in large developing countries like China and India (Chen et al., 2019a;Liu
102 et al., 2013).

103 A nationwide estimate of long-term N deposition in China based on the bulk
104 measurements as well as summaries from reported references in 270 sites by Liu et al.
105 (2013), showed an increasing rate of 0.41 kg N ha⁻¹ per year from 1980 to 2010. In
106 contrast to the increasing importance of N_{rd} deposition, due to apparently decreasing
107 N_{ox} resulting from the air quality control policies in the USA in past decades (Li et al.,
108 2016), the ratio of N_{rd}/N_{ox} recorded from bulk/wet deposition decreased from 5:1 in
109 1980 to 2:1 in 2010. This suggests a more and more important role of N_{ox} in China
110 (Liu et al., 2013). The ratio in highly developed regions such as the North China Plain
111 was even lower than 1:1 in recent years (Pan et al., 2012). However, very limited
112 long-term observations in China challenge our capacity to understand and control the
113 increase of N_r deposition. The published long-term N deposition monitoring network,
114 which includes the Acid Deposition Monitoring Network in East Asia (EANET,
115 <http://www.eanet.asia/index.html>), the National wide Nitrogen Deposition Monitoring
116 Network (NNDMN) established in 2010 by the China Agriculture University (CAU)
117 (Xu et al., 2015), and the Chinese Ecosystem Research Network (CERN) in North
118 China Plain, established by the Chinese Academy of Science (Pan et al., 2012), the
119 Acid Rain Monitoring Network, run by the China Meteorological Administration
120 (CMA-ARMN) (Tang et al., 2007;Tang et al., 2010;Ge et al., 2011), and the National
121 Acid Deposition Monitoring Network (NADMN) (Li et al., 2019b), have been
122 identified with many shortcomings. Monitoring sites are widely spread over a large

123 geographical area and therefore data records, due to the high cost of the measurement
124 and unstable financial support, are incomplete. Chemical Transport Model (CTM)
125 simulation is another option to offset these drawbacks and also to quantify long-range
126 transport of deposition in a global or regional map (Seinfeld and Pandis, 2006). It is
127 important to know the accuracy of the CTM before it is employed to investigate the
128 spatial and temporal variation of the depositions. Hayami et al. (2008) and Mann et al.
129 (2014) explain that different parameters in CTMs can result in large variations and the
130 inaccuracies. The multi-model ensemble mean (ENM) shows better performance than
131 any single one (Carmichael et al., 2002; Hayami et al., 2008; Holloway et al.,
132 2008; Wang et al., 2008). Additionally, to better localize applications of CTM,
133 comprehensive evaluations of the strengths and weaknesses of current CTMs for
134 simulating the acid deposition as well as their precursors in a unified framework, with
135 certain regulated rules and the same inputs to models, must be undertaken.

136 Model Inter-Comparison Study for Asia (MICS-Asia) provides an opportunity to
137 investigate the CTMs application with different models in Asia. MICS-Asia was first
138 employed in 1998 with the target of long-range transport and deposition in SO_4^{2-} in
139 the first stage (MICS-Asia phase I) (Carmichael et al., 2002) and sulfur, nitrogen and
140 ozone in the second stage (MICS-Asia phase II) (Carmichael et al., 2008). The
141 findings and methodologies developed in the previous inter-comparison studies
142 contributed to common understandings of the performance and uncertainties of CTM
143 applications in East Asia (Hayami et al., 2008; Carmichael et al., 2008; Han et al.,
144 2008; Wang et al., 2008). The comprehensive multi-model inter-comparison study on
145 acid deposition in China is becoming an urgent issue as the high emissions in China
146 are causing acid deposition in neighboring countries (Lin et al., 2008; Kajino et al.,
147 2011; 2013; Itahashi et al., 2018). In this study, one year simulated N_r depositions, i.e.
148 N_{ox} and N_{rd} in both wet and dry deposition, using the framework of MICS-Asia III
149 (MICS-Asia phase III), have been compared with each other and validated by the
150 observed wet deposition from EANET, NNDMN, CREN and by the Department of
151 Ecological Environment (DEE, formerly known as the Environmental Protect
152 Administration, EPA) over the whole of China. The ENM results were also compared
153 to the Vertical Column Density (VCD) from satellite and the emission inventories.
154 Finally, the uncertainties of the sources of N_r depositions and their ecological impacts
155 have been quantified. The results from this study will not only provide an important
156 reference for establishing a suitable N deposition model, the localized application of
157 CTMs in China will also be tested.

158 **2 Framework of intercomparison in MICS-Asia III**

159 **2.1 Description of the participant models**

160 In phase III of MICS-Asia, 14 chemical transport models (CTM, M1-M14) were used
161 to compare and evaluate current multi-scale air quality models (called topic 1 in
162 MICS-Asia III). The same number index was used to measure aerosols and ozone
163 levels, reported by Chen et al. (2019b) and Li et al. (2019a). However, the fully
164 coupled online Weather Research and Forecasting model with chemistry
165 (WRF-Chem), which has been indexed as M7-M10, was not included in the
166 deposition comparison part in the overview of model inter-comparison and evaluation

167 for acid deposition in Asia (Itahashi et al., 2020). Briefly, the Weather Research and
168 Forecasting model coupled with the Community Multi-scale Air Quality
169 (WRF-CMAQ) has been numbered M1-M6, with a different version of v5.0.2 for M1
170 and M2, v5.0.1 for M3 and v4.7.1 for M4-M6. M11 and M12 are the independent
171 models developed in Japan and China, named as NHM-Chem (Kajino et al., 2019)
172 and the nested air quality prediction model system (NAQPMS), respectively. A global
173 three-dimensional chemical transport model (GEOS-Chem v9.1.3), M13, was also
174 used as the long-range transport and future change prediction model in MICS-Asia III.
175 The last, M14, was represented as the Regional Atmospheric Modeling System
176 coupled with CMAQ (RAMS-CMAQ). It should be noted that the last two models,
177 M13 and M14, were not driven by the “standard” meteorological fields from WRF
178 v3.4.1 model. Basic information about the configuration of each model was
179 summarized in Table 1. More detailed description can also be found in previous
180 studies (Itahashi et al., 2020;Chen et al., 2019b;Li et al., 2019a).

181 **2.2 Model inputs and simulation domain**

182 As mentioned by Chen et al. (2019b), same (“standard”) meteorological fields,
183 emission inventories and boundary conditions have been prepared for the CTMs
184 inter-comparison in MICS-Asia III to reduce the uncertainties from model inputs.
185 However, some models such as M13 and M14, imported “non-standard” inputs due to
186 their specific characteristics. The “standard” meteorological inputs were simulated by
187 WRF v3.4.1 with the initial and lateral boundary conditions from the National Centers
188 for Environmental Prediction (NCEP) Final Analysis (FNL) data. Four dimensional
189 data assimilation (FDDA) nudging was adopted every 6 hours to improve the
190 accuracy of the meteorological parameters simulation. The assimilated meteorological
191 fields from the Goddard Earth Observing System 5 (GEOS5) of the US National
192 Aeronautics and Space Administration (NASA) (<https://gmao.gsfc.nasa.gov>) were
193 used to drive M13. The M14 model was driven by RAMS with the same FNL data for
194 nudging as the “standard” WRF simulation, which was developed by Pielke et al.
195 (1992). For the emission inputs, all the participant model were using the same
196 emission inventory, which included the MIX anthropogenic emissions over Asia
197 developed for MICS-Asia Phase III (Li et al., 2017), the biogenic emissions
198 calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN)
199 version 2.04 (Guenther et al., 2006), and the biomass burning emissions from Global
200 Fire Emission Database (GFED) version 3 (van der Werf et al., 2010). Additionally,
201 SO₂ emissions from volcanoes were collected from the AEROCOM program
202 (https://aerocom.met.no/DATA/download/emissions/AEROCOM_HC/volc, last ac-
203 cess: 11 September 2019, Diehl et al., 2012; Stuefer et al., 2013). MICS-Asia Phase
204 III provided two sets of lateral boundary conditions derived from GEOS-Chem (Bey
205 et al., 2001) and CHASER (Sudo et al., 2002), respectively. The boundary conditions
206 from GEOS-Chem were run with 2.5°×2° resolution and 47 vertical layers, while
207 those from CHASER were run with 2.8°×2.8° and 32 vertical layers. M4, M5, M6,
208 M11 and M12 used the output from CHASER as the boundary conditions, and M1,
209 M13 and M14 were from GEOS-Chem. Only M2 used the default boundary condition
210 field provided in CMAQ.

211 The “standard” simulation domain covers the region of East Asia (15.4°S-58.3°N,
212 48.5°E-160.2°E) with 180×170 grids at 45 km horizontal resolution. M1-M6, M11 and
213 M12 followed “standard” simulation domains, while M13 and M14 employed
214 different modeling domains with 0.5° latitude × 0.667° longitude and 64 × 64 km,
215 respectively. In this study, the analyzed region was only focused in China and all
216 participant models covered it. Therefore, simulated reactive N depositions in each
217 model can be analyzed and compared to show the performance of the participant
218 models. All models output of N depositions have been classified as oxidized N ($N_{ox} =$
219 $gHNO_3 + gNO_x + pNO_3^-$, including gaseous nitrate acid, NO_x and particulate nitrate)
220 and reduced N ($N_{rd} = gNH_3 + pNH_4^+$, including gaseous ammonia and particulate
221 ammonium) for comparison. Several previous studies have reported that the
222 contributions of the other N species, e.g., PAN and isoprene nitrates to total N dry
223 deposition are less important than the inorganic N, e.g., HNO_3 and NO_x (Yuan et al.,
224 2018; Wolfe et al., 2011). Thus, these organic N species were not included in this
225 study.

226 2.3 Observation data

227 China has a large land mass - almost 5,500 km from south to north (3.5°S-53.3°N) and
228 5,200 km from west to east (75.5°E-135°E). It goes from coastal to inland and from a
229 tropical climate to a frigid zone. Only 8 sites located in Guangdong, Fujian, Sichuan
230 and Shanxi in EANET were insufficient to show the real performance of CTMs in
231 China. Besides the 8 EANET sites, data from 83 sites recorded daily, weekly or yearly
232 from the CERN (Pan et al., 2012), NNDMN (Xu et al., 2015; Liu et al., 2013) and
233 DEE in Guangdong, Liaoning and Xinjiang province and Shanghai were employed in
234 this study to compare the simulated wet deposition in MICS-Asia III in China. Figure
235 1 shows the location of the 83 measurement sites as well as the divided regions of the
236 whole of China. There were 50 urban sites and 33 rural sites. More detailed
237 information of each measurement site can also be found in Table S1 in the
238 supplementary documents.

239 The daily wet deposition was measured by a wet-only sampler to collect
240 precipitation samples during the rainfall event in EANET. Analysis methods for NO_3^-
241 and NH_4^+ were based on ion chromatography and checked by ion balance and
242 conductivity agreement. Detailed descriptions can be found in the source document
243 (EANET, 2010). Daily rainwater samples at 10 sites located in the North China Plain
244 were collected using a custom wet-dry automatic collector (APS-2B, Xianglan
245 Scientific Instruments Co., Ltd., Changsha, China) in CREN. Inorganic N, including
246 NO_3^- and NH_4^+ , in the precipitation samples was measured using an ion
247 chromatography system (Model ICS- 90, Dionex Corporation, Sunnyvale, CA, USA)
248 and the standard laboratory procedure of LAPC (Wang et al., 2012). The detection
249 limit (DL) of N for this instrument was $5 \mu g l^{-1}$. A detailed description can be found in
250 the research of Pan et al. (2012). The wet/bulk NO_3^- and NH_4^+ deposition data from
251 25 sites of China Agricultural University Deposition Network (CAUDN), which was
252 renamed as NNDMN in China in 2010, has been collected and reanalyzed as yearly
253 data (Xu et al., 2015; Liu et al., 2013). At all monitoring sites precipitation samples
254 were collected using precipitation gauges (SDM6, Tianjin Weather Equipment Inc.,

255 China) located beside the DELTA systems (ca. 2m, DEnuder for Long-Term
256 Atmospheric sampling). After their collection, the samples were analyzed in CAU's
257 laboratory based on the standard laboratory procedure of CAU (Xu et al., 2015).
258 Routine NO_3^- and NH_4^+ wet depositions collected in each rainfall event at 40 sites
259 were provided by the DEE of Guangdong, Liaoning and Xinjiang provinces as well as
260 Shanghai city. The analytical process was the same used in the laboratory procedure
261 of the China National Environmental Monitoring Centre (CNEMC).

262 The temporal resolutions of the wet deposition data are different from each other,
263 i.e., daily in EANET and CREN, rainfall event collection in DEE and yearly in
264 NNDMN. For the convenience of comparison, all data from daily or rainfall event
265 collecting samples at each type of measurement site has been summarized and
266 interpolated as monthly wet deposition data to compare with the monthly simulation in
267 MICS-Asia III in this study, except the yearly data provided by NNDMN. VCD of
268 NO_2 from SCIAMACHY (<http://www.temis.nl/airpollution/no2col>) and NH_3 from
269 IASI (http://ether.ipsl.jussieu.fr/ether/pubipsl/iasial2/iasi_nh3) have also been used to
270 compare with the total deposition as well as the emissions.

271 **3 Results**

272 **3.1 Validation of wet deposition**

273 **3.1.1 Yearly comparison and monthly variation of oxidized N**

274 Yearly simulated wet deposition of N_{ox} has been evaluated by observed nitrate wet
275 deposition in 83 sites over China. Table 2 lists the statistical parameters of simulated
276 wet deposition of N_{ox} compared with the observed data in rural and urban sites of
277 China. In all sites, M1, M5 and M11 overestimated the yearly wet deposition of N_{ox}
278 with Normalized Mean Bias (NMB) of +30.3%, +55.4% and +67.2%, respectively.
279 M6, M12 and M13 simulated almost comparable results with NMB of -6.8%, +0.6%
280 and +0.1%, respectively. M2, M4 and M14 underestimated the yearly wet deposition
281 of N_{ox} with NMB of -38.7%, -10.7% and -47.4%, respectively. The NME was around
282 50% with the highest 82.2%, in M11, which is lower than that reported over the East
283 Asia with the value of 70% based on EANET observation by Itahashi et al. (2020).
284 However, the correlation coefficient R was around 0.2~0.3 (n=83) which is lower
285 than the East Asia comparison based on the EANET data (0.3~0.4, n=54) (Itahashi et
286 al., 2020). In order to eliminate influences from rainfall uncertainties (R=0.82), the
287 volume weighted mean (VWM) concentration of N_{ox} in precipitation has also been
288 evaluated. In contrast to the low R value of yearly wet deposition of N_{ox} , the
289 correlation R increased to almost 0.5 for the VWM concentrations. To judge the
290 agreement between simulation and observation, the percentages within a factor of 2
291 (FAC2) has been calculated in this study. Approximately 50% of model results
292 corresponded within the percentages within FAC2. M1 and M13 showed better
293 agreement with 60% and 59% within FAC2, while M2 and M14 showed only 36%
294 and 45% agreement within FAC2. The ground surface measurement sites were
295 divided into 49 urban sites and 34 rural sites according to their location. Overall, all
296 the models showed better performance with the R in 0.2~0.4 and FAC2 in 50%~60%
297 in urban sites than that of R in 0.05~0.3 and FAC2 in 40%~50% in rural sites. This
298 difference may not be due to the uncertainties in rainfall simulation, as the simulated

299 VWM concentration of N_{ox} in precipitation may eliminate the rainfall uncertainties,
300 and also shows better agreement in urban locations than that in rural sites (Table 2).

301 Figure 2 shows the percentile box plot the yearly wet deposition of N_{ox} simulated
302 by 9 participant models in five regions of China (i.e., North China (NC), Northeastern
303 China (NE), southeastern China (SE), northwestern China (NW), southwestern China
304 and Tibet Plateau (SW+TP)). Site by site validation of both the yearly wet deposition
305 and VWM concentration of N_{ox} simulated by each model are also displayed in Figure
306 S1. The model results show large intra-region or inter-region uncertainties, especially
307 in NC, NE and SE. The highest wet deposition of N_{ox} simulated by M11 is almost 3~4
308 times the lowest result simulated by M14 in the above regions (Figure 2). Specifically,
309 two models simulate 30-50% higher N_{ox} wet deposition, while four models are 20~40%
310 lower compared to the averaged observations in NC with the averaged value 6.5 kg N
311 $\text{ha}^{-1} \text{ a}^{-1}$. For the wet deposition of N_{ox} in SE and SW+TP, most of the participant
312 models are more than 50% underestimated with the largest underestimation of 75%
313 from M14, even though the precipitation in this region is overestimated. Additionally,
314 the divergence of observed N_{ox} wet deposition between different sites in NC, SE and
315 SW, which was shown as the length of the red box in Figure 2a, 2d and 2e, is
316 significantly larger than the multi-models' results. The scattered distribution of the
317 measurement sites in these regions is responsible for the large divergence in
318 observations. However, most of the participant models failed to capture the large
319 difference, indicating that the coarse grid in MICS-Asia III (45 km) is not suitable for
320 the performance of detailed characterization at a local scale. A global assessment of
321 the ensemble simulated wet depositions in the Task Force on Hemispheric Transport
322 of Atmospheric Pollutants (TF HTAP) by Vet et al. (2014) also indicated the
323 underestimation of the models in a number of sites in North America, Europe, Central
324 Africa and part of East Asia. The under-prediction in Europe was found due to the
325 large under-predictions of precipitation depth, while the reason for the error in East
326 Asia is still unknown. However, most of the models overestimate the wet deposition
327 of N_{ox} in NE. Several models including M1, M5 and M11 simulate more than 10 kg N
328 $\text{ha}^{-1} \text{ a}^{-1}$ N_{ox} wet deposition, almost double the observed value of $5 \text{ kg N ha}^{-1} \text{ a}^{-1}$. Both
329 the multi-models and the observations show very low values of $3\text{-}4 \text{ kg N ha}^{-1} \text{ a}^{-1}$ N_{ox}
330 wet deposition in NW, where the precipitation depth was very low compared to the
331 other regions of China (Figure S1).

332 Regarding to the comparison over the whole of East Asia reported in the overview
333 of acid deposition in MICS-Asia III (Itahashi et al., 2020), similar overestimation was
334 found in M5 and M11 while underestimation in M2, M4 and M14. It should be noted
335 that the EANET sites are mostly located around Japan, Korea and Southeast Asia, and
336 only 8 sites are located in China. The similar performances between the validation in
337 East Asia and China indicated the general underestimation (overestimation) of M2,
338 M4 and M14 (M5 and M11) were reliable in these models. For the rest of models,
339 different results were found between China and East Asia, i.e., the simulated N_{ox} wet
340 deposition in M1 was significant overestimated in China (Figure 6 of Itahashi et al.,
341 2020), but comparable with the observations over the rest of East Asia. Due to the
342 absence of the observations for atmospheric $\text{NO}_2/\text{NO}_3^-$, we cannot validate their

343 model performances directly. Instead, another companion paper (Chen et al., 2019b)
344 reported that most of models overestimated NO_3^- concentrations based on 14 sites in
345 China with most sites located in NC (Figure S5 of Chen et al., 2019b). In summary,
346 the relationship between the atmospheric concentration of NO_3^- and the wet
347 deposition in NC was not obvious, which is also same as that found in East Asia
348 (Itahashi, et al., 2020).

349 Further evaluations in temporal variations both in urban and rural sites of NC and
350 NE are displayed in Figure 3. Generally, all of the models and observations found
351 high levels of depositions in spring and summer and low values in winter in the two
352 regions. High depositions were due to large precipitation depth in the rainy season.
353 However, this was not always true in some urban sites. For example, higher
354 depositions of N_{ox} were observed in May and June with lower rainfall volume than in
355 July and August with higher rainfall in the urban sites of NC. Similar examples were
356 found at urban sites in NE. Previous studies confirmed there is a decreasing trend in
357 variations of chemical components in precipitation as rainfall varies (Aikawa and
358 Hiraki, 2009; Aikawa et al., 2014; Xu et al., 2017). If the rainfall lasts long enough, or
359 if rainfall volume was large enough, the concentrations of chemical components in
360 precipitation remained at low levels and were attributed to the effects of the in-cloud
361 scavenging process. That is, the large rainfall in an event may not cause the high level
362 of monthly wet depositions due to the low level of in-cloud deposition compared to
363 the wet depositions by several different precipitation events, especially in polluted
364 regions in urban sites. Unfortunately, only monthly data of wet depositions and
365 precipitation have been compared in this MICS-Asia III. Detailed comparisons of
366 rainfall events should be considered in the future.

367 **3.1.2 Yearly comparison and monthly variation of reduced N**

368 Simulated wet deposition of N_{rd} in MICS-Asia III has been evaluated using
369 multi-source observations from the same sites referred to in N_{ox} . It is shown in Table 3
370 that all of the models underestimated the N_{rd} wet depositions with the negative NMB
371 both in urban and rural sites. Although little difference between rural and urban sites
372 was found in M11 and M14, a better performance in rural areas was manifested by
373 lower NMB and higher FAC2 in rural sites than the urban sites in most of models
374 (-13.6%~-23.2% vs -37.3%~-45.6% for NMB and 55.9-70.6% vs 42.9-55.1% for
375 FAC2, except M11 and M14). The underestimation of the simulated N_{rd} wet
376 depositions was also found in the VWM concentration of N_{rd} in precipitation with
377 similar NMB and FAC2. However, compared with the wet deposition, the correlation
378 between the simulated and observed N_{rd} VWM concentration in precipitation was
379 significant with the R increased from ~0.3 to ~0.8, which was similar with that shown
380 in N_{ox} . This indicates the simulated VWM concentration of N_{rd} in precipitation by
381 MICS-Asia III has better performance in spatial variation than the simulation of N_{rd}
382 wet deposition over China.

383 The underestimation of N_{rd} wet deposition was also found over the whole of East
384 Asia reported in the overview of acid deposition in MICS-Asia III (Itahashi et al.,
385 2020). This implies the current CTM models might underestimate prediction of N_{rd}
386 wet deposition not only in China but also in the whole of East Asia. The close

387 correlations between the atmospheric concentration of NH_4^+ and wet deposition of N_{rd}
388 with overestimation in the atmosphere but underestimation in precipitation were
389 found over all of East Asia (Itahashi et al., 2020). In this study, the consistent
390 relationships in NC were also found in the results of Chen et al. (2019b)
391 (overestimated NH_4^+ concentration) and in this study (underestimated N_{rd} wet
392 deposition). Bae et al. (2012) reported the below-cloud scavenging process was
393 important in the simulation of N_{rd} wet deposition, which was not explicitly separated
394 in-cloud and below-cloud scavenging but computes it as a whole in the CMAQ model.
395 Note that the wet scavenging process in most of models (including M11 and M12) of
396 MICS-Asia III were similar with that treated in CMAQ module except M13 (Table 1).
397 It is too simple to accurately simulate wet deposition with the absence of accurate
398 below cloud wet scavenging simulation. This would be one reason for the
399 underestimation of N_{rd} wet deposition, especially considering the high concentration
400 of gaseous ammonia in the surface layer of NC (Pan et al., 2018; Kong et al., 2019).
401 Besides, the underestimation NH_3 emission in China would also lead to the
402 underestimation of N_{rd} wet deposition. According to the improved inversion of NH_3
403 emission in China by Kong et al. (2019), the significant underestimation of NH_3
404 emission was found especially in NCP. For the whole China, the priori emission and
405 the inversion emission of NH_3 are 10.3 Tg/year and 13.1 Tg/year, respectively.

406 Specifically, the performance of N_{rd} wet deposition prediction in MICS-Asia III has
407 also been validated in five regions through the percentile box plot in Figure 4. Site by
408 site validation of both the yearly wet deposition and VWM concentration of N_{rd}
409 simulated by each model are displayed in Figure S2. Different from that found in N_{ox} ,
410 almost similar behavior prediction has been found in same models, i.e., CMAQ
411 models in M1~M6, except M12, which was driven by a different meteorological
412 model. Other regional models as well as a global model show significantly different
413 percentile distributions in all regions. Overall, both the medium and mean value of N_{rd}
414 wet deposition were underestimated in NC, SE and SW+TP, while they are found to
415 be similar in NE and NW. The underestimation in NC was largely due to the under
416 prediction in summer time not only in urban sites (Figure 3e) but also in rural sites
417 (Figure 3f). Unfortunately, we cannot obtain convincing temporal variations in SE and
418 SW since the scarcely of monthly data in these two regions – there being only one or
419 two sites in each region. In NE, most models predicted similar temporal variations of
420 N_{rd} wet deposition, especially the high depositions in the summer months.

421 **3.2 Map of wet deposition among participant models**

422 **3.2.1 Wet deposition of oxidized N**

423 Figure 5 shows the map of the distribution of yearly N_{ox} wet deposition simulated by
424 each participant model, the ENM results and the observed results over China. Most
425 models show similar spatial patterns, with high levels of depositions in central to
426 eastern China and low levels in western China. However, the threshold value in the
427 hotspot areas (from light yellow color to orange and red colors) varies significantly

428 among the models and the average is much higher than the N_r deposition threshold
429 value of 10 kg N ha^{-1} to the temperate ecosystems suggested by Bleeker et al.(2011).
430 For example, M1, M5 and M11 simulated very high wet depositions of N_{ox} (almost
431 reaching at 20 kg N ha^{-1}) in the middle Yangtze River and Yangtze River Delta (YRD),
432 basin of Sichuan Province, south of NC and Liaoning Province located in NE. In
433 contrast, M2 and M14 fail to show the relative hotspot N_{wox} in such areas, and M4,
434 M6, M12 and M13 show an obscure hotspot with a small value of 10 kg N ha^{-1} . The
435 significant differences not only exist between different models but also in the same
436 model CMAQ, i.e., M1, M2, M4, M5 and M6. Since most models were driven by the
437 meteoroidal field and standard emission input except M13 (Geos-Chem) and M14
438 (RAMS-CMAQ), the differences in simulated N_{ox} wet deposition should come from
439 the CTMs themselves, such as the diffusion and convection process, the oxidation and
440 chemical transformation as well as the wet scavenging and deposition processes. The
441 comparison of the long lifetime specie CO (Kong et al., 2020) and weak chemical
442 activity specie BC (Chen et al., 2019b) revealed that the model uncertainties are less
443 than other factors, i.e., O_3 (Li et al., 2019) and NO_3^- (Chen et al., 2019b) which have
444 strong chemical activity and short lifetimes in the atmosphere. These results indicate
445 that the chemical reaction process rather than the diffusion and convection process is
446 one of the most important factors affecting the model uncertainties in MICS-Asia III.

447 **3.2.2 Wet deposition of reduced N**

448 Figure 6 shows the map distribution of the reduced N (N_{rd}) wet deposition over China.
449 All of the models show a similar spatial pattern with high values in central and eastern
450 China but low level of deposition in NW and northwestern of NE. Compared with the
451 N_{ox} , few differences of the simulated N_{rd} wet deposition were found among 9 models
452 except M11, which predicted significantly lower values. N_{ox} wet deposition of five
453 agricultural dominant provinces - Shandong, Henan, Hubei, Hunan and Anhui - is
454 higher than the threshold value of 10 kg N ha^{-1} , according to the simulated results by
455 most models. Unfortunately, the small number of observations in these areas make it
456 harder to validate their findings. Evidence shows the high level of N_{rd} wet deposition
457 over the threshold based on the observations in Hebei, YRD and Pearl River delta
458 (PRD). Almost all of the models under-predicted in these areas. Liu et al. (2013)
459 reported the important contribution of N_{rd} to the total N deposition in China based on
460 the long-term national scale of observed nitrogen deposition data. In the agricultural
461 predominantly agricultural areas, ammonia emission is the main contributor to N_{rd}
462 deposition (Liu et al., 2011 AE review; Kang et al., 2016).

463 **3.3 Comparisons among participant models for reactive N depositions**

464 **3.3.1 Coefficient of variations for N depositions in MICS-Asia III**

465 Besides the wet deposition of oxidized and reduced N, dry deposition was also an
466 important process for the total deposition in China (Liu et al., 2013;Pan et al., 2012).
467 Coefficient of Variation (hereinafter, CV), defined as the standard deviation divided
468 by mean value of all selected model results, with a large value denoting lower
469 consistency among the models, is applied for model comparison of simulated reactive
470 N depositions both for dry and wet deposition processes in MICS-Asia III. Figure 7
471 shows the distribution of CV for each type of simulated reactive N deposition. Since

472 the low level of mean values of deposition are more likely to be associated with a
473 higher CV, the gridded CV was only calculated in the area with the simulated
474 depositions higher than 0.5 kg N ha^{-1} (hereafter, analyzed value) in this study. As
475 shown in Figure 7, the spatial distribution of CV only covers Eastern China, Southern
476 China and Northeast China, which indicate that the quarterly and yearly fluxes of
477 reactive N deposition in these regions was higher than the analyzed value. For the
478 annual case, the CV value of N_{rd} was lower compared with N_{ox} both for dry and wet
479 depositions. This means the multi-model simulations are more consistent in N_{rd}
480 depositions than in N_{ox} depositions. Specifically, the N_{rd} in wet depositions have the
481 lowest CV values, followed by N_{rd} in dry depositions and then the N_{ox} in wet and dry
482 depositions. This suggests the simulated wet depositions of N_{rd} have fewer
483 uncertainties than other types of reactive N depositions.

484 More complicated patterns were shown in seasonal variations of each type of
485 deposition. The simulated N_{ox} for dry deposition in Figure 7 (a) showed larger
486 uncertainties in southern China (south of 30°N , with the $\text{CV} > 0.4$) than that in
487 northern China (north of 30°N , with the $\text{CV} < 0.3$) in all seasons except summer.
488 Similar spatial and temporal patterns of the CV values were found in N_{rd} dry
489 deposition. It is worth noting that the large CV values with the range of 0.4-0.6 were
490 exhibited in Central China (i.e., Henan, Hebei and Shandong provinces) during
491 summer and autumn in spite of the high flux of N_{rd} dry depositions in these regions
492 (Map distributions of N_{ox} and N_{rd} dry depositions, simulated in 9 participant models,
493 is displayed in Figure S3 and Figure S4 of the supplementary documents). This
494 suggests, importantly, that the uncertainties of the physical and chemical processes in
495 the participant models - including gas-particle equilibrium (Ge et al., 2019), dry
496 deposition parameter scheme (Zhang et al., 2003), transportation as well as the
497 chemical reaction with other acidifying substances (Liu et al., 2019) - in the regions of
498 high emissions originating from agricultural activities in growing seasons may lead to
499 significant deviations of simulated N_{rd} dry depositions.

500 For wet deposition of N_{ox} , large uncertainties were located in southern China in
501 summer and autumn with the CV values higher than 0.6 compared with the CV values
502 lower than 0.4 in other regions (Figure 7c). This high value of CV was not found in
503 the summertime of simulated N_{rd} wet deposition (Figure 7d). Due to the high portion
504 of summertime flux to the total annual wet deposition, high CV values in N_{ox}
505 contributed the most important part of the significantly larger annual CV value than
506 that shown in the N_{rd} case. Due to the same rainfall input for the wet deposition in the
507 framework of MICS-Asia III, except model 13 and 14, the different CV values for N_{rd}
508 and N_{ox} in same region (i.e., lower CV values of N_{rd} wet depositions in NC, SE and
509 Central China) can be attributed to their precursor concentrations in the air mass as
510 well as the different wet scavenging processes (Seinfeld and Pandis, 2006). This will
511 be discussed in the following section.

512 **3.3.2 Comparison of precursors in the air mass**

513 As is well known, depositions both from dry and wet parts of a certain substance were
514 found to have originated from its precursor in the air mass. The uncertainties of the
515 nitrogen related species in the air mass simulated during MICS-Asia III were

516 therefore an important index for estimating the performance of deposition simulations.
517 It should be noted that only concentrations of most of the related species at surface
518 layer have been submitted in MICS-Asia III, except NO₂ vertical column density data
519 (VCD). According to the comparison of CV between the NO₂ concentration at the
520 surface layer and VCD data (Figure S5), it was shown that there is a similar spatial
521 pattern over the whole of China. This indicates that the simulated concentration on the
522 surface layer can partly represent the difference of the whole column among the
523 multi-models, especially in providing a broad overview in China. Thus, only
524 concentration data at the surface layer has been used in this study.

525 Figure 8 shows the distribution of CV for gaseous NO_x, particulate NO₃⁻, gaseous
526 NH₃ and particulate NH₄⁺ in the air mass simulated by the 9 participant models during
527 four seasons as well as the annual mean values. There were significant seasonal
528 variations among the spatial patterns of the CV for each type of the N-related air
529 pollutant. It is interesting to note that not only the seasonal variations but also the
530 spatial patterns of the simulated precursors' CV were reasonably consistent with those
531 previously shown in the deposition part (Figure 7). For example, high CV values were
532 found in the simulation of particulate NO₃⁻ in Southern China during summer,
533 reaching or even exceeding 0.8 in SE China (Figure 8b). The high CV values were
534 also found in summertime N_{ox} wet depositions (Figure 7c). As the most important
535 precursor of N_{ox} wet deposition (Pan et al., 2012), the consistent distribution of CV
536 between the precursor and the deposition is reasonable. Different from the particulate
537 NO₃⁻, very low CV values were shown in particulate NH₄⁺ during summer leading to
538 less deviation of simulated N_{rd} wet deposition than the N_{ox}. However, uncertainties in
539 precursors cannot explain everything. For example, the high CV values of N_{ox} wet
540 deposition in south China corresponds to the low CV values of NO₃⁻ in autumn. Some
541 other factors, such as the scavenging process might be responsible for the
542 unknown-uncertainties. Xu et al. (2017;2019) first compared the below-cloud wet
543 scavenging coefficients based on the different estimation methods and found the large
544 difference even at magnitude level between each method. Thus, a more detailed
545 comparison - such as in-cloud and below-cloud wet scavenging coefficients in each
546 participant model - should be carried out in the next phase of MICS-Asia.

547 For N_{ox} dry depositions, the anomalies of deposition and NO_x concentration in the
548 air are shown in Figure S6 and Figure S7. It shows same variations among the models,
549 i.e., both of higher deposition and concentration in M1, M5, M11, M13, and lower in
550 M2, M4, M6, M12 and M14. This has also been proved in the distribution of CV
551 values in NO_x (Figure 8a) and N_{ox} dry depositions (Figure 7a) during autumn and
552 winter. As the most important precursor of N_{rd} dry deposition, gaseous NH₃ also
553 shows large CV values in central China during summer time (> 0.6). There were also
554 significant high CV values in south of the Yangtze River during the autumn and
555 winter period (0.7-0.8 in south of the Yangtze River vs 0.3-0.5 in north of the Yangtze
556 River). A similar pattern but of uncertain significance was found in the simulated N_{rd}
557 dry deposition (0.3-0.4 vs 0.2-0.3 in Figure 7b). The anomalies of N_{rd} dry deposition
558 and the gaseous NH₃ in the air simulated by each model are shown in Figure S8 and
559 Figure S9. Additionally, the dry deposition velocity (V_d) of N_{rd} - based on the ratio

560 of the dry deposition fluxes and the surface concentration (same as Tan et al., 2020) -
561 are also shown in Figure S10. The results show that the CMAQ models (M1~M6)
562 predicted similar V_d of N_{rd} , and the N_{rd} dry deposition was consistent with the gaseous
563 NH_3 concentration in the air, i.e., overestimation in M1 and M2 but underestimation in
564 M4 and M5. However, among the different models, high V_d of N_{rd} (low V_d of N_{rd}) was
565 corresponds with the overestimation (underestimation) of dry deposition in M11 and
566 M14 (M12 and M13). From the distribution of CV, similar patterns were also
567 displayed both in V_d (Figure S11) and dry deposition of N_{rd} , with low CV value in
568 NCP (0.1-0.4 for N_{rd} dry deposition, 0.1-0.3 for V_d) and high CV value in SE and SW
569 (0.4-0.8 for N_{rd} dry deposition, higher than 0.5 for V_d).

570 **4 Discussion**

571 **4.1 Ensemble results of reactive N deposition and comparison with satellite data**

572 Wang et al. (2008) first presented the ENM depositions of acidified species over East
573 Asia based on MICS-Asia II simulations and found that the ENM is better in
574 simulating wet depositions than each single model. In the phase III of MICS-Asia, the
575 ENM value of wet depositions both for N_{ox} and N_{rd} has also been validated by
576 observations and shown in Figure 5l and Figure 6l. The simulated N_{ox} wet deposition
577 and VWA concentration in rainfall exhibited larger dispersions around 1:1, in line
578 with the correlation coefficients R of 0.23 and 0.54 in 83 sites over China, compared
579 with those found in N_{rd} , which is concentrated around 1:2 line with the correlation
580 coefficients R of 0.32 and 0.8. This implies the ensemble-mean value of simulated N_{ox}
581 wet deposition has large uncertainties, while N_{rd} wet deposition was under-predicted
582 by a factor of two in MICS-Asia III. Compared to each single model, the
583 ensemble-mean shows a higher R value than most single models. However, due to a
584 lack of direct observation of dry deposition, validation for dry and total deposition of
585 reactive N cannot be achieved. Instead, the column densities from satellite and
586 emissions spatial distribution were employed to address the reasonability of the
587 ensemble-mean of four types of reactive N depositions simulated in nine models. As
588 displayed in Figure 9, dry depositions of N_{ox} and N_{rd} are concentrated in NC, YRD
589 and Henan province, which corresponds to the distribution of their emissions and
590 VCDs, respectively. Meanwhile, wet depositions of N_{ox} and N_{rd} are centered in central
591 China provinces, such as Hubei and Hunan, as well as Chengdu regions. There were
592 especially high wet depositions of N_{rd} in southwest of Hubei province and northeast
593 of Chengdu city, where high values of emissions and the VCDs for NH_3 were absent.
594 These regions loaded with high wet depositions were mainly due to the high volume
595 of rainfall (for more details, see Figure S12) and the long-range transport of acidic
596 substances (Ge et al., 2011).

597 ~~Another interesting phenomenon is that the allocations of high values of~~
598 ~~depositions and VCD of N_{ox} are different from that of N_{rd} . As shown in the Figure 9,~~
599 ~~low depositions with high values of VCD for N_{ox} and high depositions with~~
600 ~~comparatively lower level of VCD for N_{rd} co-existed in East China. On a global scale,~~
601 ~~air pollutants must follow the conservation law—that is, the emissions can be divided~~
602 ~~into two parts, i.e., the depositions and their concentrations in the air. Here we apply~~
603 ~~this concept to the entire region of China. We assume that the amount of N_{ox} and N_{rd}~~

604 transported out of the research areas is equivalent under the same atmospheric
605 advection. The emissions of N_{ox} and N_{rd} in China are also comparable ($8.3 \text{ kg N}\cdot\text{ha}^{-1}$
606 and $8.7 \text{ kg N}\cdot\text{ha}^{-1}$ for NO_x and NH_3 , respectively). At the same time, the simulated
607 low deposition of N_{ox} and observed high VCD match exactly with the high deposition
608 in N_{rd} and observed low VCD in central and eastern China. Although there is no
609 directly observed distribution map to verify the total deposition in our models, the
610 close correlation between the observed VCD and deposition can verify the rationality
611 of the simulated total deposition distribution.

612 **4.2 Contributions to the total inorganic N depositions and their potential effects**

613 Total inorganic N deposition (TIN), which includes the reduced and oxidized forms of
614 inorganic N deposition both from wet and dry processes, has been calculated for
615 estimating its ecosystem effects in this study as they were measured in most cases
616 before (Pan et al., 2012; Liu et al., 2013). Figure 10 and Figure 11 show the pathway
617 of each type of N deposition to the TIN from a spatial distribution view and 6 regions
618 statistical results, respectively. The ENM dry depositions of gaseous HNO_3 and NH_3
619 were the two important contributors to the TIN, both of which took part in 18% of
620 TIN over the whole country, while the wet deposition of NO_3^- and NH_4^+ were another
621 two main components with percentages of 23% and 28% (Table 4), respectively.
622 Consistent with that reported in the global assessment under HTAP (Vet et al., 2014)
623 and in the nationwide monitoring network (NNDMN) estimation (Xu et al., 2015), the
624 N_{rd} in China dominated the TIN deposition with the average percentage reached at 52%
625 for the ensemble results, which is slightly lower compared with 60% and 58% in the
626 two previous works. The overall contribution of wet and dry deposition to TIN was
627 almost half and half, which is consistent with that reported in NNDMN by Xu et al.
628 (Xu et al., 2015). Considering total emissions, the depositions in all of China took
629 about 67%, 65% and 66% in the 2010 emission of NH_3 , NO_x and total N
630 (NH_3-N+NO_x-N), respectively. It is interesting to show that the relationship of the
631 gridded average N_{rd} deposition and the N_{ox} deposition with their relevant emissions in
632 six regions (shown in Figure 12 with the slope: 0.56, r^2 :0.97 for N_{rd} and the slope:
633 0.47, r^2 :0.88 for N_{ox}) were consistent with that reported by Xu et al. (Xu et al., 2015)
634 (slope: 0.51, r^2 :0.89 for N_{rd} and slope: 0.48, r^2 :0.81 for N_{ox}). Even the increasing order
635 of the regions from lowest in TP to highest in NC was the same as the previously
636 measurement study. This implicates the spatial distribution as well as the relationships
637 of deposition and emission are comparable with that measured in the NNDMN. Pan et
638 al. (2013) also compared the correlations between the observed depositions and
639 emissions and attributed the inconsistent distribution between them in NCP to the
640 uncertainties of the emission. However, the patterns of depositions were also
641 influenced by the regional transport in addition to the emissions. In this study,
642 significant positive correlations of the simulated N_{ox} (N_{rd}) depositions with the
643 corresponding NO_x (NH_3) emission reflects the control role of the relative emission to
644 the spatial distribution of the depositions. Although most regions were located below
645 a 1:1 ratio of deposition to emission (Figure 12), a few regions, such as TP and NE,
646 were close to or above 1:1 ratio, implicating the impacts of transport on deposition
647 among the regions.

648 For regions, the area-average deposition of TIN was as highest as 29.2 kg N•ha⁻¹
649 and 27 kg N•ha⁻¹ in NC and SE, followed by 15 kg N•ha⁻¹ and 10.1 kg N•ha⁻¹ in SW
650 and NE, respectively. The TIN in NW and TP were as low as 3.1 and 2.7 kg N•ha⁻¹. In
651 the two highest regions of NC and SE, the deposition of TIN was similar but the
652 pathways to them were different. The N_{rd} deposition (53%) and the dry deposition
653 (54%) contributed more than half the TIN in NC, while the N_{ox} deposition (55%)
654 dominated the TIN in SE. Considering the lower ratio of NO_x/NH₃ emission in SE
655 (21.4/21.6, 0.99) than NC (30.4/24.4, 1.25), higher contribution of N_{ox} to TIN in SE
656 indicated a higher nitrogen oxidant ratio (i.e., the ratio of oxidation from NO₂ to NO₃⁻)
657 than NC. Our companion paper (Tan et al., 2020) also revealed the higher nitrogen
658 oxidation ratio in SE as 0.4-0.6, compared with that in NC as 0.2-0.4. For more
659 oxidant N species, i.e., HNO₃ and NO₃⁻, both dry and wet depositions were higher in
660 SE than that shown in NC (5.8 vs. 4.9 for dry deposition of gaseous HNO₃ and 6.9 vs.
661 6.3 for wet deposition of particulate NO₃⁻). While for less oxidant N and the reduced
662 N, all types of depositions - such as dry deposition of gaseous NO_x, gaseous NH₃ as
663 well as the particulate NH₄⁺ - were less in SE than NC, except the wet deposition of
664 particulate NH₄⁺, due to the much higher volume of rainfall in SE (Figure S5). Overall,
665 the oxidant N made the emitted NO_x easier to scavenge in SE with the ratio of
666 N_{ox}-deposition/NO_x-emission reaching 70%, while the reduced N is more likely to be
667 scavenged from its emission with the ratio of 64% in NC. The total ratio of
668 TIN/emission in NC and SE were 53% and 63%, respectively. Compared to the
669 Critical Load (Duan et al., 2001;Zhao et al., 2009;Liu et al., 2011), which is a
670 judgement of the deposited N effects to the ecosystem, the two regions almost reached
671 in some cases and even exceeded to the CL value (Table 4), indicating serious
672 ecological impacts of the N deposition in NC and SE. More attention should be given
673 to controlling N related species, especially the N_{rd} in NC and N_{ox} in SE.

674 In the less developed economic and social area of SW, due to the high emission of
675 NH₃, 60% of the TIN was contributed by N_{rd} deposition. The ratio of NO_x/NH₃
676 emission reached 0.49 as more NH₃ was emitted from agricultural activity than NO_x
677 from fossil fuel consumption. The ratio of wet deposition/TIN was 55%, which was
678 lower than the HTAP comparison during 2000 (60-70%) (Vet et al., 2014), but higher
679 than the results of NNDMN (45%) (Xu et al., 2015). Although socially undeveloped,
680 the TIN deposition was almost as high as the CL value, according to Zhao et al. (Zhao
681 et al., 2009). Besides, the high emission of NH₃, the high ratio of
682 N_{ox}-deposition/NO_x-emission of up to 94% reflects the importance of N_{ox} from high
683 emission areas, such as SE and NC, should attract our attention in this region.
684 Although the N deposition in TP was not as high as CL value - which was the lowest
685 in all regions of China with the value of 2.7 kg N•ha⁻¹ - the N ecological impacts
686 cannot be neglected since the sensitive ecosystem (Shen et al., 2019) as well as the
687 important climatic influence to all of China. Considering the high ratio of
688 TIN/emissions, which were larger than 1:1 - with 3:1 for TIN, 2.71:1 for N_{rd} and 4:1
689 for N_{ox} - the imports from outside the region were responsible for the N deposition in
690 TP.

691 **5 Conclusion**

692 Reactive N depositions over China simulated in the frame work of MICS-Asia III
693 have been compared within each participant model. Wet depositions were also
694 validated by multi-source observations, i.e., recorded data from EANET, CAS,
695 NNDMN and EPA in Guangdong and Liaoning province. Most models show the
696 consistent spatial and temporal variation of both N_{ox} and N_{rd} wet depositions in China
697 with the NME around 50%, which is lower than the value of 70% based on EANET
698 observations over Asia. Coefficient of Variation (CV) was applied for model
699 comparison of dry deposition as well as the related precursor's concentration in the air
700 mass. Consistency of both spatial and temporal variation of CV in deposition and the
701 concentration in air mass indicates that performance of the precursors' simulation was
702 highly correlated with their depositions.

703 Large deposition of ensemble simulation of N_{rd} deposition in eastern China was
704 corresponds with a low level of VCD from satellite measurements, while the case of
705 N_{ox} was just the contrary. The total emission of NO_x and NH_3 was similar at 8 kg
706 $N \cdot ha^{-1}$ in China. This indicates the allocation of both NO_x and NH_3 from the
707 deposition to the surface ground and the amount staying in the atmosphere were
708 conserved from their emission into the air, which also implicates the reasonable
709 simulation of depositions for N_{ox} and N_{rd} in MICS-Asia III.

710 Wet deposition of nitrate and ammonium as well as the dry deposition of Gaseous
711 NH_3 and HNO_3 were the important pathway to TIN deposition with the percentages of
712 18%, 18%, 23% and 28% for ensemble results, respectively. The gridded averaged N_{rd}
713 in China dominated the TIN deposition with the average percentage found to be 52%,
714 which is slightly lower than the reported 60% and 58% in HTAP and NNDMN
715 measurements. The contribution of wet and dry deposition to TIN was almost half and
716 half and consistent with that reported in NNDMN. Even the ratio of TIN/emission
717 was similar with the NNDMN, indicating that the spatial distribution as well as the
718 relationships of deposition and emission are comparable with that measured in the
719 NNDMN.

720 For different regions of China, the simulated N_{ox} wet deposition was overestimated
721 in NE but underestimated in SE and SW, while large uncertainties were shown in NC.
722 Two models simulated 30-50% higher N_{ox} wet deposition, and four models were
723 20~40% lower compared with observations in NC. The large divergences not only
724 exist between different models but also in the same CMAQ model, i.e., M1-M6. For
725 the simulation of N_{rd} wet deposition, all the models under-predicted in all regions,
726 with the largest underestimation in NC and SE. Different from N_{ox} , almost similar
727 behavior prediction of the less oxidative species such as the N_{rd} wet deposition has
728 been found in CMAQ models, indicating the chemical reaction process is one of the
729 most important factors affecting the model uncertainties in MICS-Asia III. Compared
730 to CL value, the reactive N deposition in NC, SE and SW reached or exceeded the
731 reported CL value and indicates serious ecological impacts. The control of N_{rd} in NC
732 and SW and N_{ox} in SE would be effective to mitigate the TIN deposition in these
733 regions. For the lowest reactive N deposition in TP, however, the N ecological impacts
734 cannot be neglected since it has a sensitive ecosystem and it has an important climatic
735 influence on all of China, especially considering the high ratio of TIN/emission,

736 which was mainly caused by outside sources. The joint prevention and control of air
737 pollution in China should be carefully considered and implemented in the future.

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741 **Data availability.**

742 To request observed data for scientific research purposes, please contact Baozhu Ge at
743 the Institute of Atmospheric Physics, Chinese Academy of Sciences, via email
744 (gebz@mail.iap.ac.cn).

745

746 **Author contribution**

747 BG designed the whole structure of this work, and prepared the manuscript with
748 contributions from all co-authors. BG, SI and KS led the deposition analysis group in
749 MICS-Asia III. DX, JW, FF and QT helped with the data processing. JSF, XW, KY,
750 TN, JL, MK, HL, and MZ performed the model simulations and contributed to submit
751 their simulated deposition results. ZW performed the meteorological model
752 simulation and examined the model performance. ML, JW, JK and QW prepared the
753 emission inventory data. YP and XL supported the observation data in China. ZW was
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755 **Competing interests**

756 The authors declare that they have no conflict of interest

757

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765 **Reference:**

- 766 Aikawa, M., and Hiraki, T.: Washout/rainout contribution in wet deposition estimated
767 by 0.5 mm precipitation sampling/analysis, *Atmos Environ*, 43, 4935-4939, 2009.
- 768 Aikawa, M., Kajino, M., Hiraki, T., and Mukai, H.: The contribution of site to
769 washout and rainout: Precipitation chemistry based on sample analysis from
770 0.5 mm precipitation increments and numerical simulation, *Atmos Environ*, 95,
771 165-174, <http://dx.doi.org/10.1016/j.atmosenv.2014.06.015>, 2014.
- 772 Bae, S. Y., Park, R. J., Yong, P. K., and Woo, J. H.: Effects of below-cloud scavenging
773 on the regional aerosol budget in East Asia, *Atmos Environ*, 58, p.14-22, 2012.
- 774 Benitez, J. M. G., Cape, J. N., Heal, M. R., van Dijk, N., and Diez, A. V.: Atmospheric
775 nitrogen deposition in south-east Scotland: Quantification of the organic nitrogen
776 fraction in wet, dry and bulk deposition, *Atmos Environ*, 43, 4087-4094,
777 10.1016/j.atmosenv.2009.04.061, 2009.
- 778 Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q.
779 B., Liu, H. Y., Mickley, L. J., and Schultz, M. G.: Global Modeling of
780 Tropospheric Chemistry with Assimilated Meteorology: Model Description and
781 Evaluation, *J.geophys.res*, 106, 23073–23095, 2001.
- 782 Bleeker, A., Hicks, W. K., Dentener, F., Galloway, J., and Erisman, J. W.: N deposition
783 as a threat to the World’s protected areas under the Convention on
784 Biological Diversity, *Environ Pollut*, 159, 2280-2288, 2011.
- 785 Byun, D., and Schere, K. L.: Review of the governing equations, computational
786 algorithms, and other components of the models-3 Community Multiscale Air
787 Quality (CMAQ) modeling system, *Appl. Mech. Rev.*, 59, 51-77,
788 <https://doi.org/10.1115/1.2128636>, 2006.
- 789 Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. O., Sarwar, G., Pinder, R. W.,
790 Pouliot, G. A., and Houyoux, M.: 5 Model representation of secondary organic
791 aerosol in CMAQv4.7, *Environ. Sci. Technol.*, 44(22), 8553-8560,
792 <https://doi.org/10.1021/es100636q>, 2010.
- 793 Carmichael, G. R., Calori, G., Hayami, H., Uno, I., Cho, S. Y., Engardt, M., Kim, S.
794 B., Ichikawa, Y., Ikeda, Y., Woo, J. H., Ueda, H., and Amann, M.: The
795 MICS-Asia study: model intercomparison of long-range transport and sulfur
796 deposition in East Asia, *Atmos Environ*, 36, 175-199, 2002.
- 797 Carmichael, G. R., Sakurai, T., Streets, D., Hozumi, Y., Ueda, H., Park, S. U., Fung,
798 C., Han, Z., Kajino, M., Engardt, M., Bennet, C., Hayami, H., Sartelet, K.,
799 Holloway, T., Wang, Z., Kannari, A., Fu, J., Matsuda, K., Thongbooncho, N., and
800 Amann, M.: MICS-Asia II: The model intercomparison study for Asia Phase II
801 methodology and overview of findings, *Atmos Environ*, 42, 3468-3490, 2008.
- 802 Carter, W. L.: Implementation of the SAPRC-99 chemical mechanism into the
803 Models-3 framework, Report to the United States Environmental Protection
804 Agency, available at: <http://www.engr.ucr.edu/~carter/pubs/s99mod3.pdf>, 2000,
805 last access: 20 March 2019.
- 806 Chen, C., Park, T., Wang, X., Piao, S., Xu, B., Chaturvedi, R. K., Fuchs, R., Brovkin,
807 V., Ciais, P., Fensholt, R., Tømmervik, H., Bala, G., Zhu, Z., Nemani, R. R., and
808 Myneni, R. B.: China and India lead in greening of the world through land-use

809 management, *Nature Sustainability*, 2, 122-129, 10.1038/s41893-019-0220-7,
810 2019a.

811 Chen, L., Gao, Y., Zhang, M., Fu, J. S., Zhu, J., Liao, H., Li, J., Huang, K., Ge, B.,
812 Wang, X., Lam, Y. F., Lin, C. Y., Itahashi, S., Nagashima, T., Kajino, M., Yamaji,
813 K., Wang, Z., and Kurokawa, J.: MICS-Asia III: multi-model comparison and
814 evaluation of aerosol over East Asia, *Atmos. Chem. Phys.*, 19, 11911-11937,
815 10.5194/acp-19-11911-2019, 2019b.

816 Clark, C. M., and Tilman, D.: Loss of plant species after chronic low-level nitrogen
817 deposition to prairie grasslands, *Nature*, 451, 712-715, 2008.

818 Clark, H., and Kremer, J. N.: Estimating direct and episodic atmospheric nitrogen
819 deposition to a coastal waterbody, *Mar Environ Res*, 59, 349-366, 2005.

820 Colella, P., and Woodward, P. R.: The piecewise parabolic method (PPM) for gas
821 dynamical simulations, *J. Comp. Phys.*, 54, 174–201, 1984.

822 Dise, N. B., and Wright, R. F.: Nitrogen leaching from European forests in relation to
823 nitrogen deposition, *Forest Ecology & Management*, 71, 153-161, 1995.

824 Duan, L., Xie, S. D., Zhou, Z. P., Ye, X. M., and Hao, J. M.: Calculation and mapping
825 of critical loads for S, N and acidity in China, *Water Air Soil Poll*, 130,
826 1199-1204, 2001.

827 Duce, R. A., LaRoche, J., Altieri, K., Arrigo, K. R., Baker, A. R., Capone, D. G.,
828 Cornell, S., Dentener, F., Galloway, J., Ganeshram, R. S., Geider, R. J., Jickells,
829 T., Kuypers, M. M., Langlois, R., Liss, P. S., Liu, S. M., Middelburg, J. J., Moore,
830 C. M., Nickovic, S., Oschlies, A., Pedersen, T., Prospero, J., Schlitzer, R.,
831 Seitzinger, S., Sorensen, L. L., Uematsu, M., Ulloa, O., Voss, M., Ward, B., and
832 Zamora, L.: Impacts of atmospheric anthropogenic nitrogen on the open ocean,
833 *Science*, 320, 893-897, 10.1126/science.1150369, 2008.

834 EANET: Technical Manual for Wet Deposition Monitoring in East Asia,
835 <http://www.eanet.asia/product/manual/techwet.pdf>, 2010.

836 Erisman, J. W., Sutton, M. A., Galloway, J., Klimont, Z., and Winiwarter, W.: How a
837 century of ammonia synthesis changed the world, *Nat Geosci*, 1, 636-639, 2008.

838 Fenn, M. E., Poth, M. A., Aber, J. D., Baron, J. S., Bormann, B. T., Johnson, D. W.,
839 Lemly, A. D., McNulty, S. G., Ryan, D. F., and Stottlemeyer, R.: NITROGEN
840 EXCESS IN NORTH AMERICAN ECOSYSTEMS: PREDISPOSING
841 FACTORS, ECOSYSTEM RESPONSES, AND MANAGEMENT
842 STRATEGIES, *Ecol Appl*, 8, 706-733, 1998.

843 Fountoukis, C., and Nenes, A.: ISORROPIA II: A computationally efficient aerosol
844 thermodynamic equilibrium model for K⁺, Ca²⁺, Mg²⁺, NH₄⁺, Na⁺, SO₄²⁻,
845 NO₃⁻, Cl⁻, H₂O aerosols, *Atmos. Chem. Phys.*, 7, 4639–4659, 2007.

846 Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W.,
847 Seitzinger, S. P., Asner, G. P., Cleveland, C. C., Green, P. A., Holland, E. A., Karl,
848 D. M., Michaels, A. F., Porter, J. H., Townsend, A. R., and Vorosmarty, C. J.:
849 Nitrogen cycles: past, present, and future, *Biogeochemistry*, 70, 153-226, 2004.

850 Galloway, J. N., Townsend, A. R., Erisman, J. W., Bekunda, M., Cai, Z. C., Freney, J.
851 R., Martinelli, L. A., Seitzinger, S. P., and Sutton, M. A.: Transformation of the
852 nitrogen cycle: Recent trends, questions, and potential solutions, *Science*, 320,

853 889-892, 2008.

854 Ge, B., Xu, X., Ma, Z., Pan, X., Wang, Z., Lin, W., Ouyang, B., Xu, D., Lee, J., Zheng,
855 M., Ji, D., Sun, Y., Dong, H., Squires, F. A., Fu, P., and Wang, Z.: Role of
856 Ammonia on the Feedback Between AWC and Inorganic Aerosol Formation
857 During Heavy Pollution in the North China Plain, *Earth Space Sci*, 6, 1675-1693,
858 10.1029/2019ea000799, 2019.

859 Ge, B. Z., Wang, Z. F., Xu, X. B., Tang, J., He, Y. J., Uno, I., and Ohara, T.: Impact of
860 the East Asian summer monsoon on long-term variations in the acidity of summer
861 precipitation in Central China, *Atmos Chem Phys*, 11, 1671-1684, DOI
862 10.5194/acp-11-1671-2011, 2011.

863 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.:
864 Estimates of global terrestrial isoprene emissions using MEGAN (Model of
865 Emissions of Gases and Aerosols from Nature), *Atmos Chem Phys*, 6, 3181-3210,
866 2006.

867 Han, Z., Sakurai, T., Ueda, H., Carmichael, G. R., Streets, D., Hayami, H., Wang, Z.,
868 Holloway, T., Engardt, M., Hozumi, Y., Park, S. U., Kajino, M., Sartelet, K., Fung,
869 C., Bennet, C., Thongboonchoo, N., Tang, Y., Chang, A., Matsuda, K., and
870 Amann, M.: MICS-Asia II: Model intercomparison and evaluation of ozone and
871 relevant species, *Atmos Environ*, 42, 3491-3509, 2008.

872 Hayami, H., Sakurai, T., Han, Z., Ueda, H., Carmichael, G. R., Streets, D., Holloway,
873 T., Wang, Z., Thongboonchoo, N., Engardt, M., Bennet, C., Fung, C., Chang, A.,
874 Park, S. U., Kajino, M., Sartelet, K., Matsuda, K., and Amann, M.: MICS-Asia II:
875 Model intercomparison and evaluation of particulate sulfate, nitrate and
876 ammonium, *Atmos Environ*, 42, 3510-3527, 2008.

877 Holloway, T., Sakurai, T., Han, Z., Ehlers, S., Spak, S. N., Horowitz, L. W.,
878 Carmichael, G. R., Streets, D. G., Hozumi, Y., Ueda, H., Park, S. U., Fung, C.,
879 Kajino, M., Thongboonchoo, N., Engardt, M., Bennet, C., Hayami, H., Sartelet,
880 K., Wang, Z., Matsuda, K., and Amann, M.: MICS-Asia II: Impact of global
881 emissions on regional air quality in Asia, *Atmos Environ*, 42, 3543-3561, 2008.

882 Holtgrieve, G. W., Schindler, D. E., Hobbs, W. O., Leavitt, P. R., Ward, E. J., Bunting,
883 L., Chen, G. J., Finney, B. P., Gregory-Eaves, I., Holmgren, S., Lisac, M. J., Lisi,
884 P. J., Nydick, K., Rogers, L. A., Saros, J. E., Selbie, D. T., Shapley, M. D., Walsh,
885 P. B., and Wolfe, A. P.: A Coherent Signature of Anthropogenic Nitrogen
886 Deposition to Remote Watersheds of the Northern Hemisphere, *Science*, 334,
887 1545-1548, 2011.

888 Holtslag, A. A. M., and Boville, B.: Local versus nonlocal boundary layer diffusion in
889 a global climate model, *J. Clim.*, 6, 1825-1842, 1993.

890 Itahashi, S., Yumimoto, K., Uno, I., Hayami, H., Fujita, S. I., Pan, Y., and Wang, Y.: A
891 15-year record (2001-2015) of the ratio of nitrate to non-sea-salt sulfate in
892 precipitation over East Asia, *Atmos. Chem. Phys.*, 18, 2835-2852,
893 10.5194/acp-18-2835-2018, 2018.

894 Itahashi, S., Ge, B., Sato, K., Fu, J. S., Wang, X., Yamaji, K., Nagashima, T., Li, J.,
895 Kajino, M., Liao, H., Zhang, M., Wang, Z., Li, M., Kurokawa, J., Carmichael, G.
896 R., and Wang, Z.: MICS-Asia III: Overview of model inter-comparison and

897 evaluation of acid deposition over Asia, *Atmos. Chem. Phys.*, 2019, 1-53,
898 10.5194/acp-20-2667-2020, 2020.

899 Janjic, Z.: The step-mountain eta coordinate model: Further developments of the
900 convection, viscous sublayer, and turbulence closure schemes, *Mon. Weather*
901 *Rev.*, 122, 927–945, 1994.

902 Janssens, I. A., Dieleman, W., Luyssaert, S., Subke, J. A., Reichstein, M., Ceulemans,
903 R., Ciais, P., Dolman, A. J., Grace, J., Matteucci, G., Papale, D., Piao, S. L.,
904 Schulze, E. D., Tang, J., and Law, B. E.: Reduction of forest soil respiration in
905 response to nitrogen deposition, *Nat Geosci*, 3, 315-322, 2010.

906 Kajino, M., Ueda, H., Sato, K., and Sakurai, T.: Spatial distribution of the
907 source-receptor relationship of sulfur in Northeast Asia, *Atmos Chem Phys*, 11,
908 6475-6491, 2011.

909 Kajino, M., Sato, K., Inomata, Y., and Ueda, H.: Source–receptor relationships of
910 nitrate in Northeast Asia and influence of sea salt on the long-range transport of
911 nitrate, *Atmos Environ*, 79, 67-78,
912 <http://dx.doi.org/10.1016/j.atmosenv.2013.06.024>, 2013.

913 Kajino, M., Deushi, M., Sekiyama, T. T., Oshima, N., Yumimoto, K., Tanaka, T. Y.,
914 Ching, J., Hashimoto, A., Yamamoto, T., Ikegami, M., Kamada, A., Miyashita, M.,
915 Inomata, Y., Shima, S., Ueda, H., Maki, T., and Mikami, M.: NHM-Chem, the
916 Japan Meteorological Agency’s regional meteorology – chemistry model (v1.0):
917 model description and aerosol representations, *Geosci. Model Dev. Discuss.*,
918 <https://doi.org/10.5194/gmd-2018-128>, 2018.

919 Kajino, M., Deushi, M., Sekiyama, T. T., Oshima, N., Yumimoto, K., Tanaka, T. Y.,
920 Ching, J., Hashimoto, A., Yamamoto, T., Ikegami, M., Kamada, A., Miyashita, M.,
921 Inomata, Y., Shima, S., Takami, A., Shimizu, A., Hatakeyama, S., Sadanaga, Y.,
922 Irie, H., Adachi, K., Zaizen, Y., Igarashi, Y., Ueda, H., Maki, T., and Mikami, M.,
923 NHM-Chem, the Japan Meteorological Agency’s regional meteorology –
924 chemistry model: model evaluations toward the consistent predictions of the
925 chemical, physical, and optical properties of aerosols, *J. Meteor. Soc. Japan*,
926 97(2), 337-374, <http://dx.doi.org/10.2151/jmsj.2019-020>, 2019.

927 Kong, L., Tang, X., Zhu, J., Wang, Z., Fu, J. S., Wang, X., Itahashi, S., Yamaji, K.,
928 Nagashima, T., Lee, H. J., Kim, C. H., Lin, C. Y., Chen, L., Zhang, M., Tao, Z., Li,
929 J., Kajino, M., Liao, H., Sudo, K., Wang, Y., Pan, Y., Tang, G., Li, M., Wu, Q., Ge,
930 B., and Carmichael, G. R.: Evaluation and uncertainty investigation of the NO₂,
931 CO and NH₃ modeling over China under the framework of MICS-Asia III,
932 *Atmos. Chem. Phys.*, 20, 181-202, 10.5194/acp-20-181-2020, 2020.

933 Kong, L., Tang, X., Zhu, J., Wang, Z., Pan, Y., Wu, H., Wu, L., Wu, Q., He, Y., Tian,
934 S., Xie, Y., Liu, Z., Sui, W., Han, L., and Carmichael, G.: Improved Inversion of
935 Monthly Ammonia Emissions in China Based on the Chinese Ammonia
936 Monitoring Network and Ensemble Kalman Filter, *Environ Sci Technol*, 53,
937 12529-12538, 10.1021/acs.est.9b02701, 2019.

938 Li, J., Nagashima, T., Kong, L., Ge, B., Yamaji, K., Fu, J. S., Wang, X., Fan, Q.,
939 Itahashi, S., Lee, H. J., Kim, C. H., Lin, C. Y., Zhang, M., Tao, Z., Kajino, M.,
940 Liao, H., Li, M., Woo, J. H., Kurokawa, J., Wang, Z., Wu, Q., Akimoto, H.,

941 Carmichael, G. R., and Wang, Z.: Model evaluation and intercomparison of
942 surface-level ozone and relevant species in East Asia in the context of MICS-Asia
943 Phase III – Part 1: Overview, *Atmos. Chem. Phys.*, 19, 12993-13015,
944 10.5194/acp-19-12993-2019, 2019a.

945 Li, M., Zhang, Q., Kurokawa, J. I., Woo, J. H., He, K., Lu, Z., Ohara, T., Song, Y.,
946 Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang,
947 S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission
948 inventory under the international collaboration framework of the MICS-Asia and
949 HTAP, *Atmos. Chem. Phys.*, 17, 935-963, 10.5194/acp-17-935-2017, 2017.

950 Li, R., Cui, L., Zhao, Y., Zhang, Z., Sun, T., Li, J., Zhou, W., Meng, Y., Huang, K.,
951 and Fu, H.: Wet deposition of inorganic ions in 320 cities across China:
952 spatio-temporal variation, source apportionment, and dominant factors, *Atmos.*
953 *Chem. Phys.*, 19, 11043-11070, 10.5194/acp-19-11043-2019, 2019b.

954 Li, Y., Schichtel, B. A., Walker, J. T., Schwede, D. B., Chen, X., Lehmann, C. M.,
955 Puchalski, M. A., Gay, D. A., and Jr, C. J.: Increasing importance of deposition of
956 reduced nitrogen in the United States, *Proc Natl Acad Sci U S A*, 113, 5874-5879,
957 2016.

958 Lin, M., Oki, T., Bengtsson, M., Kanae, S., Holloway, T., and Streets, D. G.:
959 Long-range transport of acidifying substances in east Asia - Part II -
960 Source-receptor relationships, *Atmos Environ*, 42, 5956-5967, DOI
961 10.1016/j.atmosenv.2008.03.039, 2008.

962 Liu, M., Huang, X., Song, Y., Tang, J., Cao, J., Zhang, X., Zhang, Q., Wang, S., Xu, T.,
963 Kang, L., Cai, X., Zhang, H., Yang, F., Wang, H., Yu, J. Z., Lau, A. K. H., He, L.,
964 Huang, X., Duan, L., Ding, A., Xue, L., Gao, J., Liu, B., and Zhu, T.: Ammonia
965 emission control in China would mitigate haze pollution and nitrogen deposition,
966 but worsen acid rain, *Proceedings of the National Academy of Sciences*, 116,
967 7760, 10.1073/pnas.1814880116, 2019.

968 Liu, X. J., Duan, L., Mo, J. M., Du, E. Z., Shen, J. L., Lu, X. K., Zhang, Y., Zhou, X.
969 B., He, C. N., and Zhang, F. S.: Nitrogen deposition and its ecological impact in
970 China: An overview, *Environ Pollut*, 159, 2251-2264, 2011.

971 Liu, X. J., Zhang, Y., Han, W. X., Tang, A. H., Shen, J. L., Cui, Z. L., Vitousek, P.,
972 Erisman, J. W., Goulding, K., Christie, P., Fangmeier, A., and Zhang, F. S.:
973 Enhanced nitrogen deposition over China, *Nature*, 494, 459-462, 2013.

974 Liu, H., Jacob, D. J., Bey, I., and Yantosca, R. M.: Constraints from ^{210}Pb and ^7Be on
975 wet deposition and transport in a global three-dimensional chemical, *J. Geophys.*
976 *Res.*, 2001, 106(D11), 12109–12128, 2001.

977 Mann, G. W., Carslaw, K. S., Reddington, C. L., Pringle, K. J., and Henzing, J. S.:
978 Intercomparison and evaluation of global aerosol microphysical properties among
979 AeroCom models of a range of complexity, *Atmospheric Chemistry & Physics*,
980 14, 4679-4713, 2014.

981 Nenes, A., Pandis, S.N., and Pilinis, C.: ISORROPIA: A new thermodynamic
982 equilibrium model for multiphase multicomponent inorganic aerosols, *Aquat.*
983 *Geoch.*, 4, 123–152, 1998.

984 Nicolas, G., and Galloway, J. N.: An Earth-system perspective of the global nitrogen

985 cycle, *Nature*, 451, 293-296, 2008.

986 Pan, Y. P., Wang, Y. S., Tang, G. Q., and Wu, D.: Wet and dry deposition of
987 atmospheric nitrogen at ten sites in Northern China, *Atmos Chem Phys*, 12,
988 6515-6535, 2012.

989 Pan, Y., Tian, S., Zhao, Y., Zhang, L., Zhu, X., Gao, J., Huang, W., Zhou, Y., Song, Y.,
990 and Zhang, Q.: Identifying ammonia hotspots in China using a national
991 observation network, *Environ Sci Technol*, 2018.

992 Phoenix, G. K., Hicks, W. K., Cinderby, S., Kuylenstierna, J. C. I., Stock, W. D.,
993 Dentener, F. J., Giller, K. E., Austin, A. T., Lefroy, R. D. B., Gimeno, B. S.,
994 Ashmore, M. R., and Ineson, P.: Atmospheric nitrogen deposition in world
995 biodiversity hotspots: the need for a greater global perspective in assessing N
996 deposition impacts, *Global Change Biol*, 12, 470-476, 2006.

997 Pielke, R. A., Cotton, W. R., Walko, R. L., Tremback, C. J., Lyons, W. A., Grasso, L.
998 D., Nicholls, M. E., Moran, M. D., Wesley, D. A., Lee, T. J., and Copeland, J. H.:
999 A comprehensive meteorological modeling system—RAMS, *Meteorol Atmos*
1000 *Phys*, 49, 69-91, 10.1007/BF01025401, 1992.

1001 Pleim, J. E., Xiu, A., Finkelstein, P. L., and Otte, T. L.: A coupled land-surface and dry
1002 deposition model and comparison to field measurements of surface heat, moisture,
1003 and ozone fluxes. *Water Air Soil Pollut. Focus*, 1(5), 243–252,
1004 <https://doi.org/10.1023/A:1013123725860>, 2001.

1005 Pleim, J. E.: A combined local and nonlocal closure model for the atmospheric
1006 boundary layer, Part I: Model description and testing, *J. Appl. Meteor. Climatol.*,
1007 46, 1383–1395, 2007a.

1008 Pleim, J. E.: A combined local and nonlocal closure model for the atmospheric
1009 boundary layer. Part II: Application and evaluation in a mesoscale meteorological
1010 model, *J. Appl. Meteorol. Clim.*, 46, 1396–1409, 2007b.

1011 Seinfeld, J. H., and Pandis, S. N.: *Atmospheric chemistry and physics: from air*
1012 *pollution to climate change*, Wiley, New York, 2006.

1013 Shen, H., Dong, S., Li, S., Xiao, J., Han, Y., Yang, M., Zhang, J., Gao, X., Xu, Y., Li,
1014 Y., Zhi, Y., Liu, S., Dong, Q., Zhou, H., and Yeomans, J. C.: Effects of simulated
1015 N deposition on photosynthesis and productivity of key plants from different
1016 functional groups of alpine meadow on Qinghai-Tibetan plateau, *Environ Pollut*,
1017 251, 731-737, <https://doi.org/10.1016/j.envpol.2019.05.045>, 2019.

1018 Sudo, K., Takahashi, M., Kurokawa, J. I., and Akimoto, H.: CHASER: A global
1019 chemical model of the troposphere 1. Model description, *Journal of Geophysical*
1020 *Research*, 107, ACH-1-ACH 7-20, 2002.

1021 Tang, J., Chen, H. B., Yu, X. L., Wang, S., Yao, P., Lv, B., Xu, X. B., and Ding, G.:
1022 Evaluation of results of station inter-comparison with blind samples in Acid Rain
1023 Monitoring Network in China(in Chinese), *Meteoro. Monthly*, 33, 75–83, 2007.

1024 Tang, J., Xu, X., Ba, J., and Wang, S.: Trends of the precipitation acidity over China
1025 during 1992-2006, *Chinese Sci Bul*, 5, 1-9, 2010.

1026 Tan, J., Fu, J. S., Carmichael, G. R., Itahashi, S., Tao, Z., Huang, K., Dong, X., Yamaji,
1027 K., Nagashima, T., Wang, X., Liu, Y., Lee, H. J., Lin, C. Y., Ge, B., Kajino, M.,
1028 Zhu, J., Zhang, M., Hong, L., and Wang, Z.: Why models perform differently on

1029 particulate matter over East Asia? – A multi-model intercomparison study for
1030 MICS-Asia III, *Atmos. Chem. Phys.*, 20, 7393-7410, 10.5194/acp-20-7393-2020,
1031 2020.

1032 van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P.
1033 S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire
1034 emissions and the contribution of deforestation, savanna, forest, agricultural, and
1035 peat fires (1997–2009), *Atmos. Chem. Phys.*, 10, 11707-11735,
1036 10.5194/acp-10-11707-2010, 2010.

1037 Vet, R., Artz, R. S., Carou, S., Shaw, M., Ro, C. U., Aas, W., Baker, A., Bowersox, V.
1038 C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar, J. J., Gillett, R., Forti, M. C.,
1039 Gromov, S., Hara, H., Khodzher, T., Mahowald, N. M., Nickovic, S., Rao, P. S. P.,
1040 and Reid, N. W.: A global assessment of precipitation chemistry and deposition of
1041 sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and
1042 phosphorus, *Atmos Environ*, 93, 3-100, 2014.

1043 Walcek, C. J., and Aleksic, N. M.: A simple but accurate mass conservative
1044 peak-preserving, mixing ratio bounded advection algorithm with fortran code,
1045 *Atmos. Environ.*, 32, 3863–3880, 1998.

1046 Wang, Y., Yu, W., Pan, Y., and Wu, D.: Acid neutralization of precipitation in Northern
1047 China, *J Air Waste Manag Assoc*, 62, 204-211, 2012.

1048 Wang, Y. X., McElroy, M. B., Jacob, D. J., and Yantosca, R. M.: A nested grid
1049 formulation for chemical transport model over Asia: Applications to CO, *J.*
1050 *Geophys. Res.*, 109, D22307, <https://doi.org/10.1029/2004JD005237>, 2004.

1051 Wang, Z. F., Xie, F. Y., Sakurai, T., Ueda, H., Han, Z. W., Carmichael, G. R., Streets,
1052 D., Engardt, M., Holloway, T., Hayami, H., Kajino, M., Thongboonchoo, N.,
1053 Bennet, C., Park, S. U., Fung, C., Chang, A., Sartelet, K., and Amann, M.:
1054 MICS-Asia II: Model inter-comparison and evaluation of acid deposition, *Atmos*
1055 *Environ*, 42, 3528-3542, 2008.

1056 Wesely, M. L.: Parameterization of surface resistance to gaseous dry deposition in
1057 regional numerical models, *Atmos. Environ.*, 16, 1293–1304, 1989.

1058 Wolfe, G. M., Thornton, J. A., Bouvier-Brown, N. C., Goldstein, A. H., Park, J. H.,
1059 McKay, M., Matross, D. M., Mao, J., Brune, W. H., LaFranchi, B. W., Browne, E.
1060 C., Min, K. E., Wooldridge, P. J., Cohen, R. C., Crouse, J. D., Faloona, I. C.,
1061 Gilman, J. B., Kuster, W. C., de Gouw, J. A., Huisman, A., and Keutsch, F. N.:
1062 The Chemistry of Atmosphere-Forest Exchange (CAFE) Model - Part 2:
1063 Application to BEARPEX-2007 observations, *Atmos Chem Phys*, 11, 1269-1294,
1064 10.5194/acp-11-1269-2011, 2011.

1065 Xu, D., Ge, B., Wang, Z., Sun, Y., Chen, Y., Ji, D., Yang, T., Ma, Z., Cheng, N., Hao,
1066 J., and Yao, X.: Below-cloud wet scavenging of soluble inorganic ions by rain in
1067 Beijing during the summer of 2014, *Environ Pollut*, 230, 963-973,
1068 <https://doi.org/10.1016/j.envpol.2017.07.033>, 2017.

1069 Xu, D., Ge, B., Chen, X., Sun, Y., Cheng, N., Li, M., Pan, X., Ma, Z., Pan, Y., and
1070 Wang, Z.: Multimethod determination of the below-cloud wet scavenging
1071 coefficients of aerosols in Beijing, China, *Atmos. Chem. Phys. Discuss.*, 2019,
1072 1-26, 10.5194/acp-2019-680, 2019.

1073 Xu, W., Luo, X. S., Pan, Y. P., Zhang, L., Tang, A. H., Shen, J. L., Zhang, Y., Li, K. H.,
1074 Wu, Q. H., Yang, D. W., Zhang, Y. Y., Xue, J., Li, W. Q., Li, Q. Q., Tang, L., Lu,
1075 S. H., Liang, T., Tong, Y. A., Liu, P., Zhang, Q., Xiong, Z. Q., Shi, X. J., Wu, L.
1076 H., Shi, W. Q., Tian, K., Zhong, X. H., Shi, K., Tang, Q. Y., Zhang, L. J., Huang, J.
1077 L., He, C. E., Kuang, F. H., Zhu, B., Liu, H., Jin, X., Xin, Y. J., Shi, X. K., Du, E.
1078 Z., Dore, A. J., Tang, S., Collett, J. L., Goulding, K., Sun, Y. X., Ren, J., Zhang, F.
1079 S., and Liu, X. J.: Quantifying atmospheric nitrogen deposition through a
1080 nationwide monitoring network across China, *Atmos Chem Phys*, 15,
1081 12345-12360, 2015.

1082 Yamartino, R. J.: Nonnegative, conserved scalar transport using grid-cell-centered,
1083 spectrally constrained Blackman cubics for applications on a variable-thickness
1084 mesh, *Mon. Weather Rev.*, 121, 753–763, 1993.

1085 Yuan, J., Ling, Z., Wang, Z., Lu, X., Fan, S., He, Z., Guo, H., Wang, X., and Wang, N.:
1086 PAN-Precursor Relationship and Process Analysis of PAN Variations in the Pearl
1087 River Delta Region, *Atmosphere-Basel*, 9, 10.3390/atmos9100372, 2018.

1088 Zaveri, R. A., and Peters, L. K.: A new lumped structure photochemical mechanism
1089 for large-scale applications, *J. Geophys. Res.*, 104, 30387–30415, 1999.

1090 Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry
1091 deposition in air-quality models, *Atmos.chem.phys*, 3, 2067-2082, 2003.

1092 Zhao, Y., Duan, L., Xing, J., Larssen, T., Nielsen, C. P., and Hao, J. M.: Soil
1093 Acidification in China: Is Controlling SO₂ Emissions Enough?, *Environ Sci*
1094 *Technol*, 43, 8021-8026, 2009.

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1096

Tables

1097 Table 1. Mechanism and parameterization of deposition part of MICS-Asia III

No	M1	M2	M4	M5	M6	M11	M12	M13	M14
Model- (version) ^a	CMAQ v5.0.2	CMAQ v5.0.2	CMAQ v4.7.1	CMAQ v4.7.1	CMAQ v4.7.1	NAQPS	NHM-Chem	GEOS-Chem	CMAQ v4.7.1
Advection-H ^b	Yamo	Yamo	PPM	PPM	Yamo	WA	WA	TPCORE	PPM
Advection-V ^b	PPM	PPM	PPM	PPM	Yamo	WA	WA	TPCORE	PPM
Diffusion-H ^b	multiscale	multiscale	multiscale	multiscale	multiscale	BD	multiscale	HB	multiscale
Diffusion-V ^b	ACM2	ACM2	ACM2	ACM2	ACM2	K-theory	MYJ	HB	ACM2
Gas-Chemistry ^c	SAPRC-99	SAPRC-99	SAPRC-99	SAPRC-99	SAPRC-99	CBMZ	SAPRC-99	Bey	SAPRC-99
Aerosol-chemistry ^d	AERO6	AERO6	AERO5	AERO5	AERO5	ISORROPIA (v1.7)	ISORROPIA (v2.1)	ISORROPIA (2.1)	ISORROPIA (v1.7)
Cloud & Aqueous ^e	ISORROPIA (v2.1)	ISORROPIA (v2.1)	ISORROPIA (v1.7)	ISORROPIA (v1.7)	ISORROPIA (v1.7)	ISORROPIA (v1.7)	ISORROPIA (v2.1)	ISORROPIA (2.1)	ISORROPIA (v1.7)
Dry dep ^f	ACM-AE6	ACM-AE6	ACM-AE5	ACM-AE5	ACM-AE5	Ge	WC	Jacob	ACM
Wet dep ^g	M3DRY	M3DRY	M3DRY	M3DRY	M3DRY	Wesely	Kajino	Wesely & Wang	M3DRY
Met	Foley	Foley	Foley	Foley	Foley	Ge	Kajino	Liu	Foley
Emission ^h	WRF	GEOS-5	RAMS						
	standard	standard	standard						

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1099 ^a: References for the advection scheme are as follows: Yamo: Yamartino, 1993; PPM: Piecewise Parabolic Method (Colella and Woodward, 1984); WA: Walcek and Aleksic, 1998; TPCORE: Wang et al., 2004.

1100 ^b: References for diffusion scheme are as follows: ACM2: Asymmetric Convective Model version 2 (Pleim, 2007a,b); BD: Byun and Dennis, 1995; HB: Holtlag and Boville, 1993; multiscale: Byun and Schere, 2006; MYJ: Janjic, 1994.

1103 c: References for the gas phase chemistry are as follows: Bey: Bey et al., 2001; CBMZ: Zaveri and Peters, 1999; SAPRC-99: Carter, 2000.
1104 d: References for the aerosol chemistry are as follows: ISORROPIA version 2.1: Fountoukis and Nenes, 2007; version 1.7: Nenes et al., 1998.
1105 e: References for the Cloud & Aqueous are as follows: Ge: Ge et al., 2014; WC: Walcek, 1986 and Carlton, 2007; Jacob: Jacob, 2000;
1106 f: References for the dry deposition scheme are as follows: M3DRY: Pleim et al., 2001; Kajino: Kajino et al., 2018; Wang: Wang et al., 2004;
1107 Wesely: Wesely, 1989.
1108 g: References for the wet deposition scheme are as follows: Foley: Foley et al, 2010; Ge: Ge et al., 2014; Kajino: Kajino et al., 2018; Liu: Liu et
1109 al., 2001.
1110 h: “standard” indicates the basic emission inventories in Phase III.
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Table 2. Statistical parameters of oxidized N deposition for urban, rural and whole China.

Urban (N=49)	OBS	M1	M2	M4	M5	M6	M11	M12	M13	M14
	Oxidized N deposition (kg N ha ⁻¹)									
R	0.24	0.25	0.28	0.27	0.26	0.25	0.40	0.37	0.22	
NMB%	16.8%	-44.5%	-19.3%	38.5%	-15.8%	48.2%	-5.3%	-7.2%	-52.7%	
NME%	56.4%	60.4%	51.3%	64.0%	51.0%	67.1%	46.9%	44.2%	59.1%	
FAC2%	63.3%	32.7%	51.0%	57.1%	57.1%	59.2%	57.1%	61.2%	40.8%	
Mean	7.1	3.9	5.7	9.9	6.0	10.5	6.7	6.6	3.4	
	Oxidized N concentration in rainfall (mg N/L)									
R	0.60	0.57	0.60	0.62	0.59	0.49	0.61	0.52	0.50	
NMB%	26.8%	-37.9%	-11.3%	49.3%	-6.7%	75.9%	2.7%	19.3%	-31.3%	
NME%	57.6%	51.4%	46.0%	69.0%	47.5%	94.8%	47.4%	65.5%	60.0%	
FAC2%	59.2%	42.9%	59.2%	51.0%	59.2%	51.0%	61.2%	49.0%	34.7%	
Mean	0.9	0.5	0.8	1.3	0.8	1.5	0.9	1.0	0.6	
	Oxidized N deposition (kg N ha ⁻¹)									
Rural (N=34)										
R	0.09	0.05	0.09	0.14	0.09	0.28	0.26	0.23	0.30	
NMB%	55.4%	-27.8%	5.1%	86.9%	9.9%	102.5%	11.5%	13.6%	-37.6%	
NME%	83.7%	57.8%	59.9%	103.3%	60.5%	110.1%	54.4%	56.0%	50.3%	
FAC2%	55.9%	41.2%	50.0%	35.3%	47.1%	38.2%	55.9%	55.9%	50.0%	
Mean	5.4	3.9	5.7	10.2	6.0	11.0	6.1	6.2	3.4	
	Oxidized N concentration in rainfall (mg N/L)									
R	0.43	0.41	0.44	0.46	0.44	0.48	0.47	0.35	0.43	
NMB%	20.5%	-43.0%	-17.1%	45.2%	-13.4%	63.2%	-9.4%	-0.2%	-43.2%	
NME%	65.4%	55.6%	54.2%	76.3%	53.8%	89.2%	53.8%	62.7%	53.3%	
FAC2%	44.1%	38.2%	41.2%	41.2%	44.1%	41.2%	47.1%	32.4%	41.2%	

Mean	0.9	1.0	0.5	0.7	1.2	0.7	1.4	0.8	0.9	0.5
All sites (N=83)										
Oxidized N deposition (kg N ha ⁻¹)										
R	0.2	0.2	0.17	0.21	0.21	0.19	0.24	0.37	0.33	0.23
NMB%	30.3%	30.3%	-38.7%	-10.7%	55.4%	-6.8%	67.2%	0.6%	0.1%	-47.4%
NME%	66.0%	66.0%	59.5%	54.3%	77.8%	54.3%	82.2%	49.5%	48.3%	56.0%
FAC2%	60.2%	60.2%	36.1%	50.6%	48.2%	53.0%	50.6%	56.6%	59.0%	44.6%
Mean	6.4	8.4	3.9	5.7	10.0	6.0	10.7	6.5	6.4	3.4
Oxidized N concentration in rainfall (mg N/L)										
R	0.53	0.53	0.51	0.54	0.56	0.53	0.48	0.56	0.46	0.46
NMB%	24.2%	24.2%	-40.0%	-13.7%	47.6%	-9.5%	70.7%	-2.3%	11.3%	-36.2%
NME%	60.8%	60.8%	53.1%	49.4%	72.0%	50.1%	92.5%	50.0%	64.4%	57.3%
FAC2%	53.0%	53.0%	41.0%	51.8%	47.0%	53.0%	47.0%	55.4%	42.2%	37.3%
Mean	0.9	1.1	0.5	0.7	1.3	0.8	1.5	0.8	1.0	0.5

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Table 3. Same as Table 2 but for reduced N deposition.

Urban (N=49)	OBS	M1	M2	M4	M5	M6	M11	M12	M13	M14		
				Reduced N deposition (kg N ha ⁻¹)								
R	0.30	0.31	0.33	0.33	0.34	0.32	0.41	0.33	0.49	0.05		
NMB%	-38.2%	-43.0%	-45.6%	-45.6%	-43.9%	-37.3%	-73.5%	-38.8%	-38.8%	-60.2%		
NME%	50.7%	52.2%	52.9%	52.9%	51.4%	49.8%	73.5%	50.0%	46.3%	64.1%		
FAC2%	53.1%	44.9%	42.9%	42.9%	46.9%	51.0%	16.3%	51.0%	55.1%	34.7%		
Mean	10.9	6.2	5.9	5.9	6.1	6.8	2.9	6.7	6.7	4.3		
				Reduced N concentration in rainfall (mg N/L)								
R	0.83	0.83	0.84	0.84	0.84	0.83	0.77	0.86	0.75	0.56		
NMB%	-38.0%	-42.6%	-42.2%	-42.2%	-40.6%	-36.1%	-73.8%	-41.1%	-22.1%	-48.8%		
NME%	44.5%	47.5%	47.4%	47.4%	46.0%	43.7%	73.8%	43.7%	46.0%	62.1%		
FAC2%	57.1%	51.0%	42.9%	42.9%	51.0%	55.1%	10.2%	57.1%	44.9%	24.5%		
Mean	1.5	0.9	0.9	0.9	0.9	1.0	0.4	0.9	1.2	0.8		
				Reduced N deposition (kg N ha ⁻¹)								
Rural (N=34)												
R	0.29	0.29	0.28	0.28	0.30	0.32	0.27	0.28	0.52	0.44		
NMB%	-14.4%	-22.0%	-21.1%	-21.1%	-18.3%	-13.6%	-62.5%	-23.2%	-19.0%	-46.2%		
NME%	48.0%	47.7%	48.1%	48.1%	46.5%	47.2%	68.5%	45.8%	40.8%	49.4%		
FAC2%	70.6%	55.9%	58.8%	58.8%	61.8%	67.6%	23.5%	61.8%	73.5%	52.9%		
Mean	9.0	7.7	7.1	7.1	7.3	7.7	3.4	6.9	7.3	4.8		
				Reduced N concentration in rainfall (mg N/L)								
R	0.79	0.79	0.81	0.81	0.82	0.80	0.74	0.82	0.69	0.55		
NMB%	-27.5%	-34.2%	-31.9%	-31.9%	-29.7%	-26.4%	-69.0%	-33.8%	-20.2%	-47.4%		
NME%	37.7%	40.6%	39.0%	39.0%	36.6%	37.1%	69.6%	37.9%	40.5%	56.9%		
FAC2%	52.9%	52.9%	52.9%	52.9%	52.9%	55.9%	17.6%	52.9%	64.7%	44.1%		

Mean	1.3	1.0	0.9	0.9	0.9	1.0	0.4	0.9	1.1	0.7
All sites (N=83)										
Reduced N deposition (kg N ha ⁻¹)										
R	0.26	0.27	0.26	0.27	0.28	0.30	0.29	0.48	0.20	0.20
NMB%	-29.6%	-35.3%	-36.7%	-34.5%	-28.6%	-69.5%	-33.1%	-31.6%	-55.1%	-55.1%
NME%	49.7%	50.6%	51.2%	49.6%	48.9%	71.7%	48.5%	44.3%	58.7%	58.7%
FAC2%	60.2%	49.4%	49.4%	53.0%	57.8%	19.3%	55.4%	62.7%	42.2%	42.2%
Mean	10.1	7.1	6.4	6.6	7.2	3.1	6.8	6.9	4.5	4.5
Reduced N concentration in rainfall (mg N/L)										
R	0.81	0.81	0.82	0.83	0.81	0.75	0.84	0.73	0.56	0.56
NMB%	-34.0%	-39.4%	-38.2%	-36.4%	-32.3%	-71.9%	-38.3%	-21.3%	-48.3%	-48.3%
NME%	41.9%	44.9%	44.2%	42.3%	41.2%	72.2%	41.5%	43.9%	60.1%	60.1%
FAC2%	55.4%	51.8%	47.0%	51.8%	55.4%	13.3%	55.4%	53.0%	32.5%	32.5%
Mean	1.4	0.9	0.9	0.9	1.0	0.4	0.9	1.1	0.7	0.7

1118 Table 4. Types of depositions and its relevant contributions to TIN as well as the
 1119 emissions of reduced and oxidized N in different regions (Unit: kg N/ha/yr).

Regions		NC	NE	NW	SE	SW	TP	China
Types of deposit ion	gHNO3d	4.9	1.8	0.8	5.8	2.4	0.2	2.1
	gNH3d	6.7	1.8	0.5	3.7	3.0	0.5	2.0
	gNOxd	1.2	0.3	0.1	1.0	0.3	0.0	0.3
	pNH4d	1.9	0.5	0.2	1.5	0.8	0.1	0.6
	pNO3d	1.3	0.4	0.1	1.2	0.4	0.0	0.4
	pNH4w	7.0	2.6	0.8	7.0	5.2	1.3	3.2
	pNO3w	6.3	2.7	0.7	6.9	3.0	0.6	2.6
	N _{rd}	15.6	4.9	1.6	12.2	9.0	1.9	5.9
	N _{ox}	13.6	5.2	1.6	14.9	6.0	0.8	5.4
	Wet TIN	13.3	5.3	1.5	13.9	8.2	1.9	5.8
	Dry TIN	16.0	4.8	1.7	13.2	6.9	0.8	5.5
TIN	29.2	10.1	3.1	27.0	15.0	2.7	11.3	
N _{rd} /TI N %	This study	53	49	52	45	60	70	52
	NNDMN							58
	HTAP							>60
Wet/TI N %	This study	46	52	48	51	55	70	51
	NNDMN	43	46	39	58	45	50	48
	HTAP	40~50	40~60	30~60	~60	60~70	60~70	
Emissi on	N _{rd}	24.4	4.9	2.9	21.6	13.1	0.7	8.7
	N _{ox}	30.4	5.6	3.1	21.4	6.4	0.2	8.3
	TIN	54.8	10.5	5.9	43.0	19.5	0.9	17.1
Dep/E mi %	N _{rd}	64	100	55	56	69	271	67
	N _{ox}	45	93	52	70	94	400	65
	TIN	53	96	53	63	77	300	66
Critical load	SSMB1	10~30	5~20	<5	10~20	>20	10~15	
	Empirical*	>200	<15	<15	50~200	50~200	20~50	
	SSMB2	>50	14-50	<14	20-50	10~30	~14	

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1121 **Figures and captions**

1122 **Figure 1:** Locations of the measurement sites and the distribution of the ID.

1123 **Figure 2:** Percentile Box plot of oxidized N wet deposition simulated in each model
1124 and compared with the observation as well as the rainfalls, with 99% and 1%
1125 represented for the high and low points, 90% and 10% represented for the top and
1126 bottom horizontal lines, 75% and 25% represented for the upper and lower edge of the
1127 box and asterisk and long horizontal line in the middle of the box represented for the
1128 medium and mean value, respectively.

1129 **Figure 3:** Monthly variation of simulated wet deposition of oxidized N compared
1130 with the observations in urban sites (a) and rural sites (b) of NC; urban sites (c) and
1131 rural sites (d) of NE; as well as of reduced N in urban sites (e) and rural sites (f) of
1132 NC; urban sites (g) and rural sites (h) of NE.

1133 **Figure 4:** Same as Figure 2 but for reduced N wet depositions.

1134 **Figure 5:** Distributions of the wet depositions of N_{ox} simulated by M1~M14 (a)~(i),
1135 ENM of the multi-models (j) MICS-Asia III, observation from multi-source
1136 measurements (k) and the comparison between ENM and observations (l) ($kgN \cdot ha^{-1}$).

1137 **Figure 6:** Same as Figure 5 but for N_{rd} .

1138 **Figure 7:** Spatial distribution of CV of (a) N_{ox} dry deposition, (b) N_{rd} dry deposition,
1139 (c) N_{ox} wet deposition and (d) N_{rd} wet deposition in MICS-Asia III on the annual and
1140 seasonal basis.

1141 **Figure 8:** Distribution of CV of NO_x (a), NO_3^- (b), NH_3 (c) and NH_4^+ (d) in the air
1142 mass for seasonal and annual.

1143 **Figure 9:** ENM results for dry deposition (a) and wet deposition (b) of N_{ox} , VCD of
1144 NO_2 from SCIAMACHY (c) and NO_x emission from MICS-Asia (d); ENM results for
1145 dry deposition (e) and wet deposition (f) of N_{rd} , VCD of NH_3 from IASI (g) and NH_3
1146 emission from MICS-Asia (h).

1147 **Figure 10:** ENM results of each process of N deposition flux (a) and the fraction in
1148 TIN (b) in MICS-Asia III. The icons w_N , w_A , d_{NO2} , d_{NH3} , d_{HNO3} ,
1149 $d_{ammonium}$ and $d_{nitrate}$ represented wet deposition of nitrate, wet deposition of
1150 ammonium, dry deposition of NH_3 , dry deposition of HNO_3 , dry deposition of
1151 ammonium and dry deposition of nitrate, respectively.

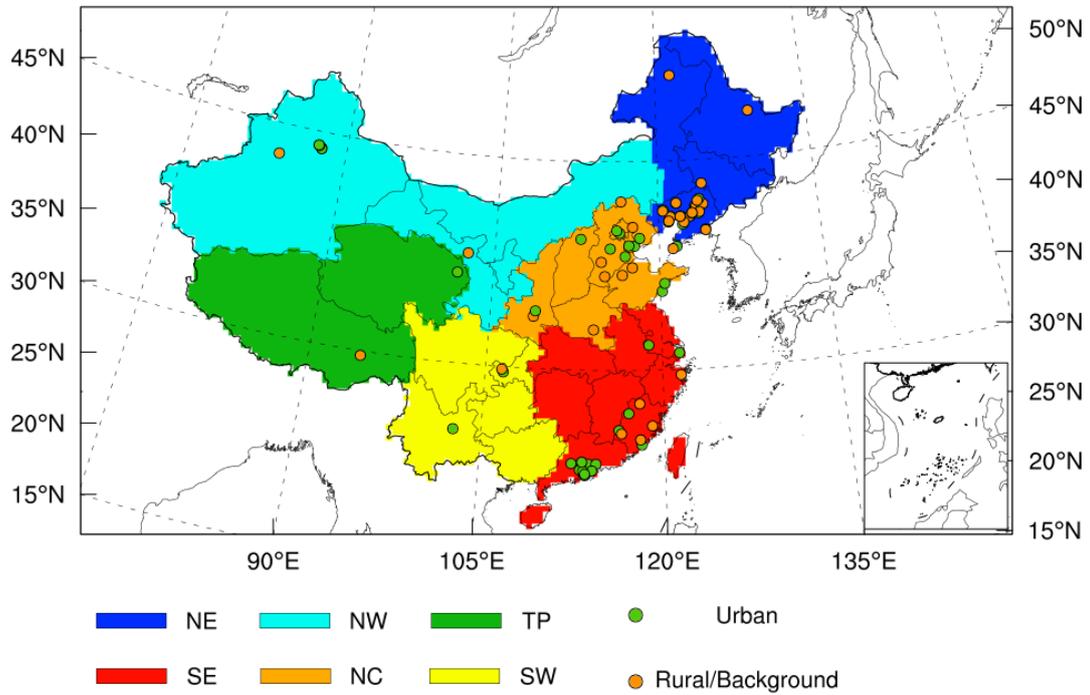
1152 **Figure 11:** Pathway of N species to TIN deposition in different regions from ENM
1153 results (a), and TIN depositions by wet or dry deposited manner (b) in percentile Box
1154 plot; with 90% and 10% represented for the top and bottom horizontal line, 75% and
1155 25% represented for the upper and lower edge of the box and asterisk in the middle of

1156 the box represented for the medium value, respectively.

1157 **Figure 12:** Relationship of N_{rd} deposition vs. NH_3 emission (a) and relationship of
1158 N_{ox} deposition vs. NO_x emission (b) in each region of China.

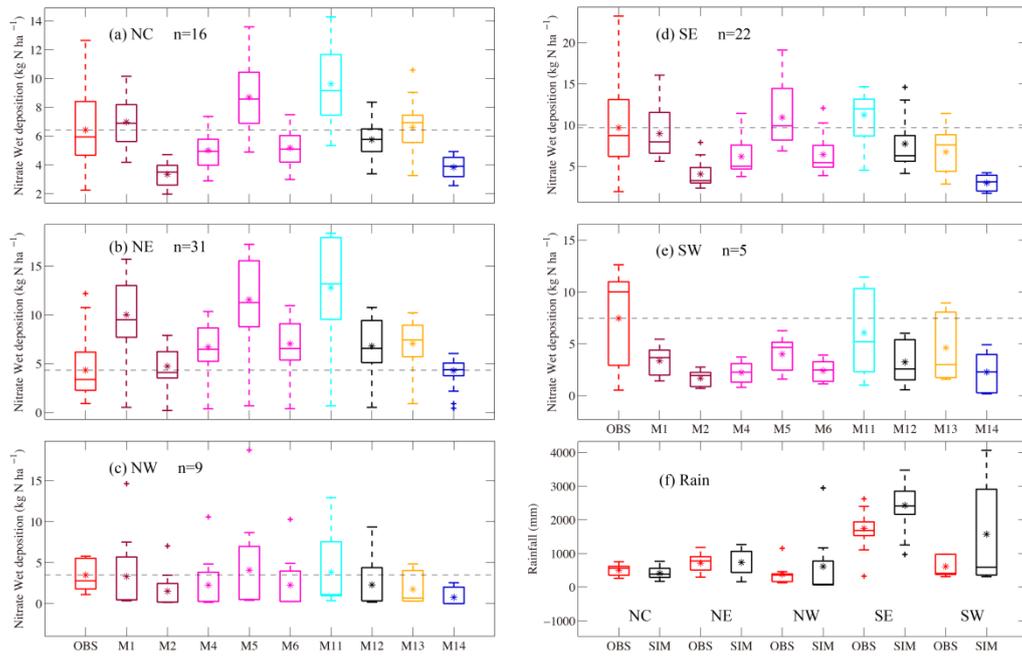
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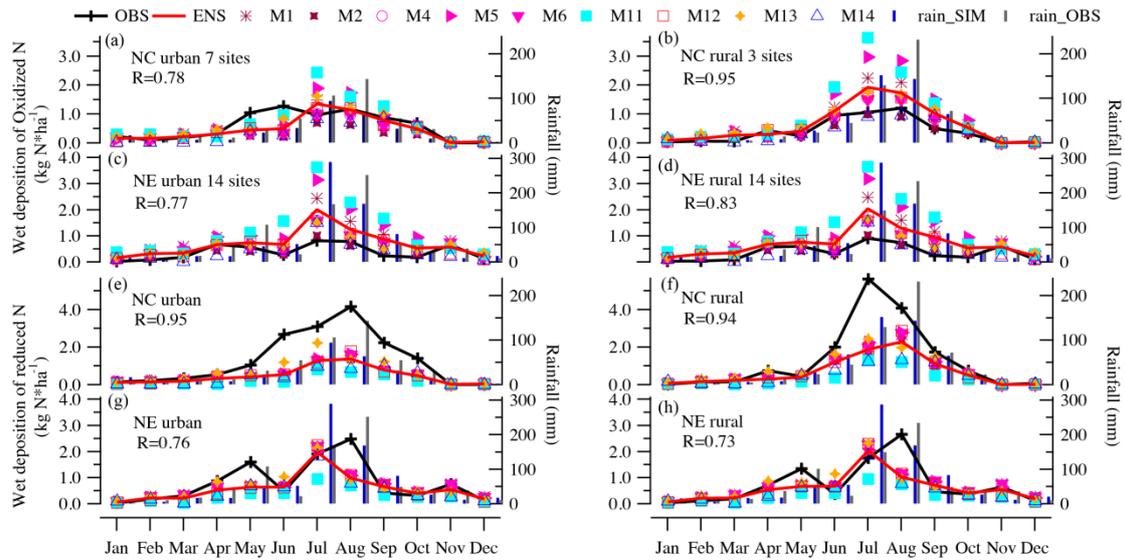
1161

1162 Figure 1: Locations of the measurement sites and the distribution of the ID.



1164

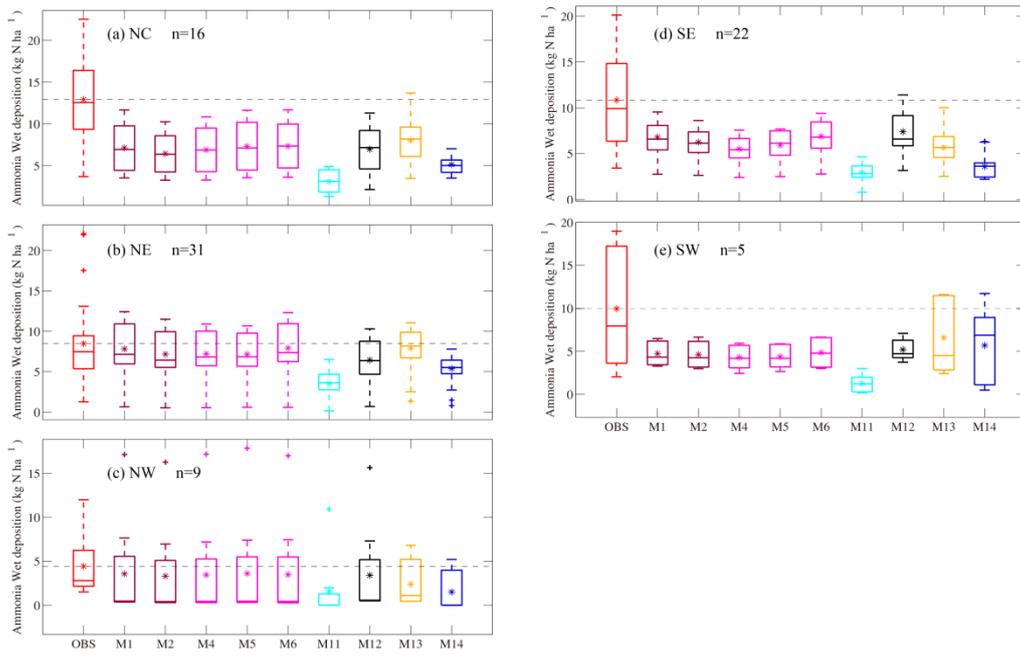
1165 Figure 2: Percentile Box plot of oxidized N wet deposition simulated in each model
 1166 and compared with the observation as well as the rainfalls, with 99% and 1%
 1167 represented for the high and low points, 90% and 10% represented for the top and
 1168 bottom horizontal lines, 75% and 25% represented for the upper and lower edge of the
 1169 box and asterisk and long horizontal line in the middle of the box represented for the
 1170 medium and mean value, respectively.



1171

1172 Figure 3: Monthly variation of simulated wet deposition of oxidized N compared with
 1173 the observations in urban sites (a) and rural sites (b) of NC; urban sites (c) and rural
 1174 sites (d) of NE; as well as of reduced N in urban sites (e) and rural sites (f) of NC;
 1175 urban sites (g) and rural sites (h) of NE.

1176

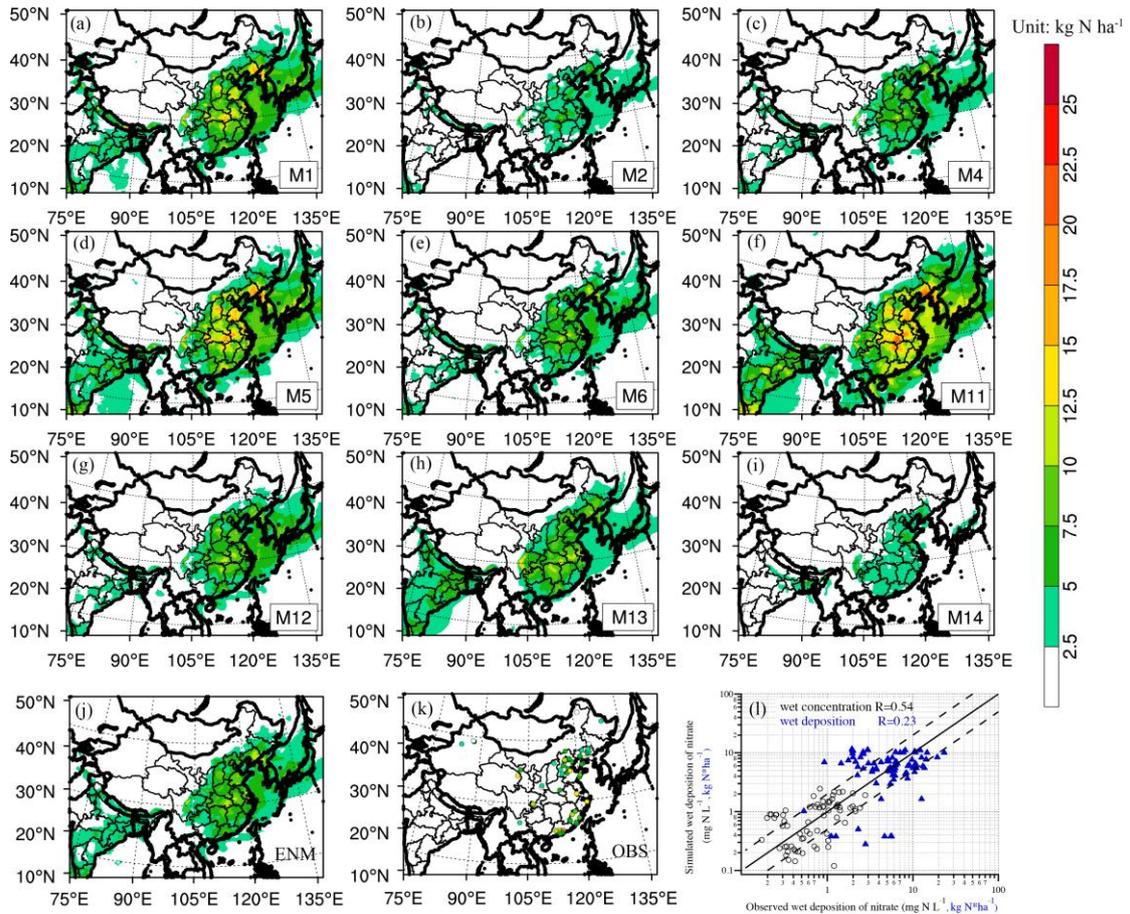


1177

1178 Figure 4: Same as Figure 2 but for reduced N wet depositions.

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1182 Figure 5: Distributions of the wet depositions of N_{ox} simulated by M1~M14 (a)~(i),

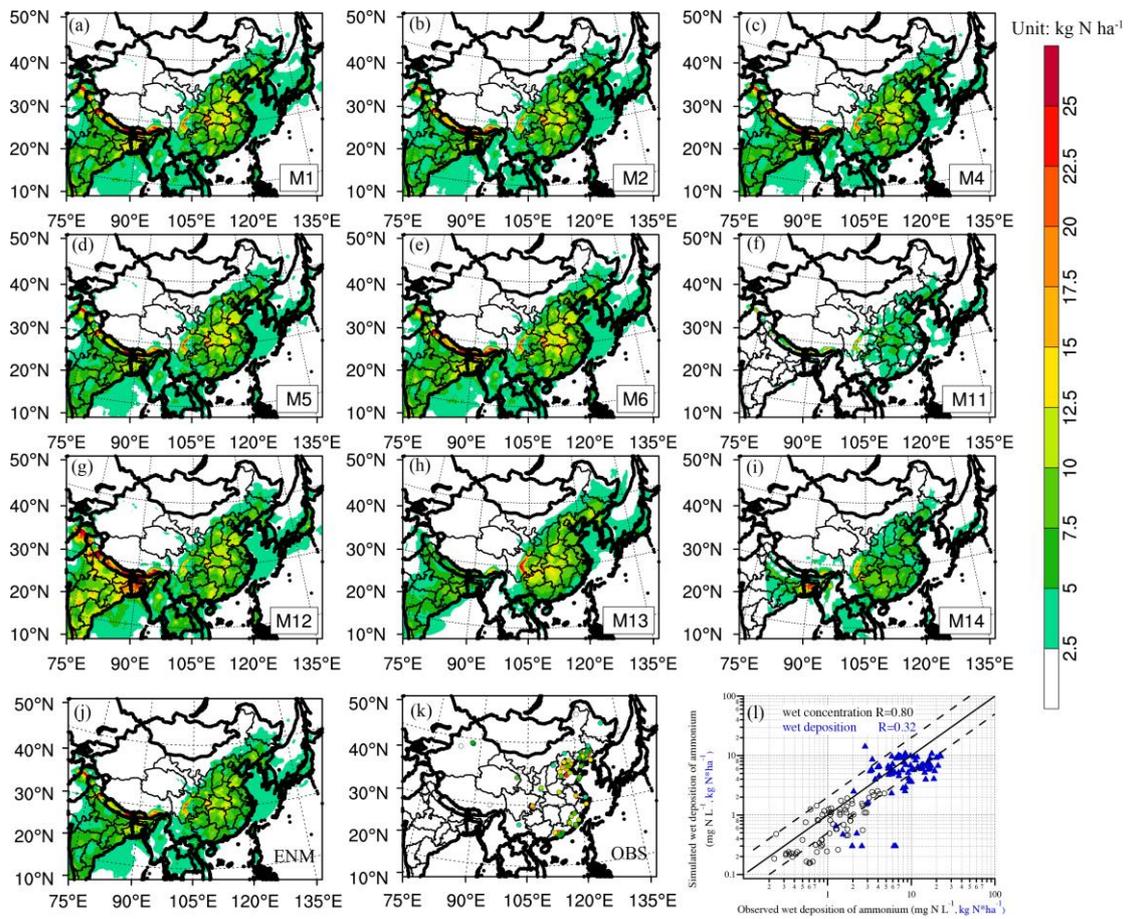
1183 ENM of the multi-models (j) MICS-Asia III, observation from multi-source

1184 measurements (k) and the comparison between ENM and observations (l) (kgN·ha⁻¹).

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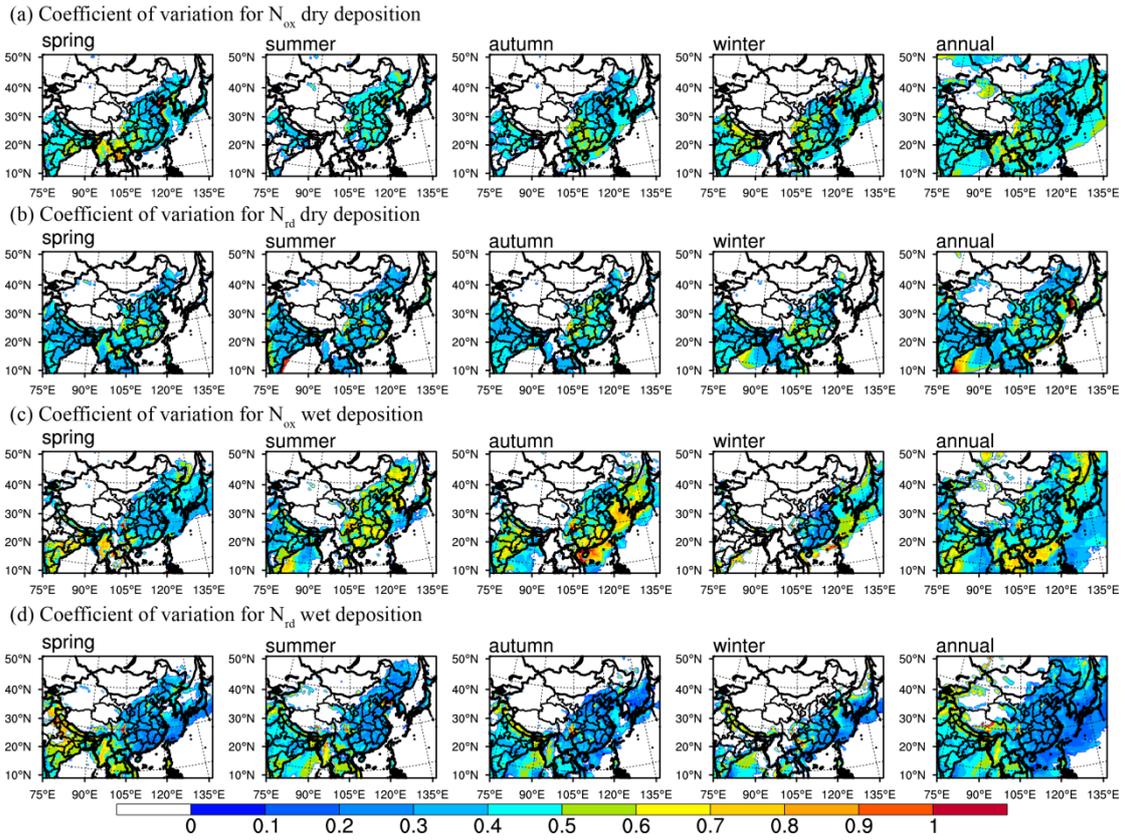


1188

1189 **Figure 6: Same as Figure 5 but for N_{rd} .**

1190

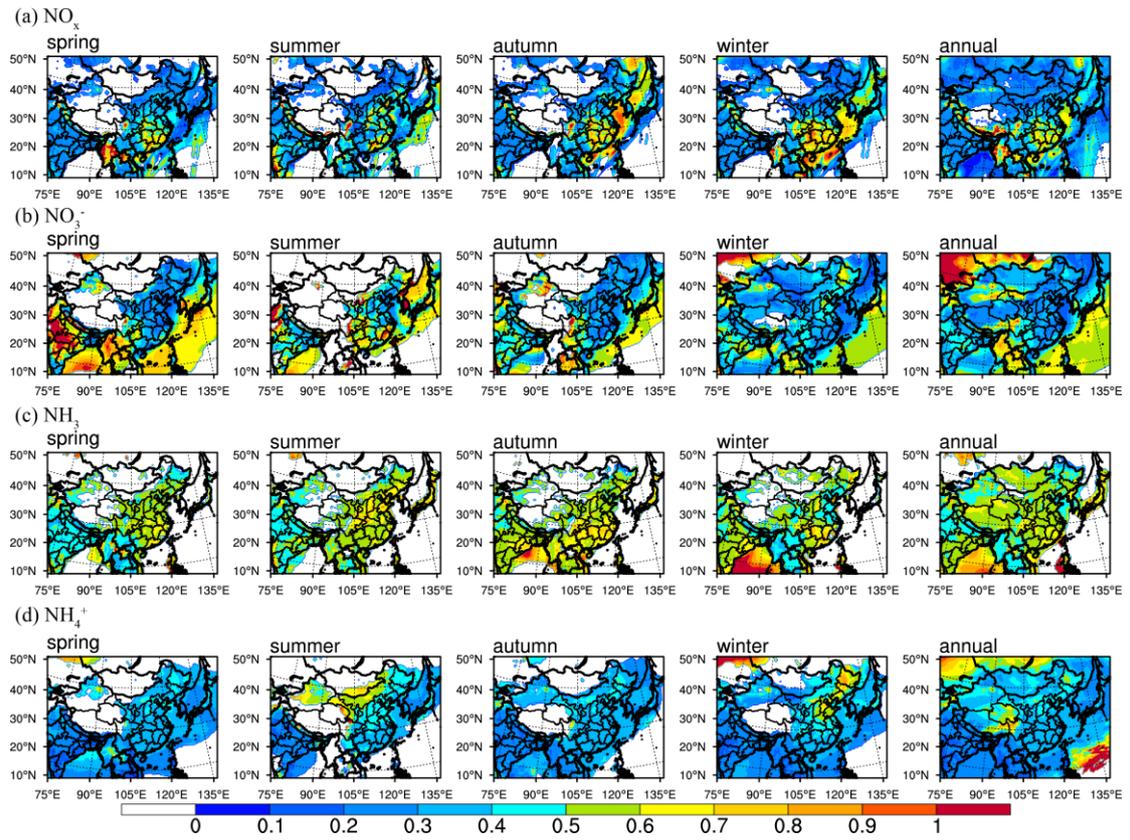
1191



1192

1193 Figure 7: Spatial distribution of CV of (a) N_{ox} dry deposition, (b) N_{rd} dry deposition,
 1194 (c) N_{ox} wet deposition and (d) N_{rd} wet deposition in MICS-Asia III on the annual and
 1195 seasonal basis.

1196

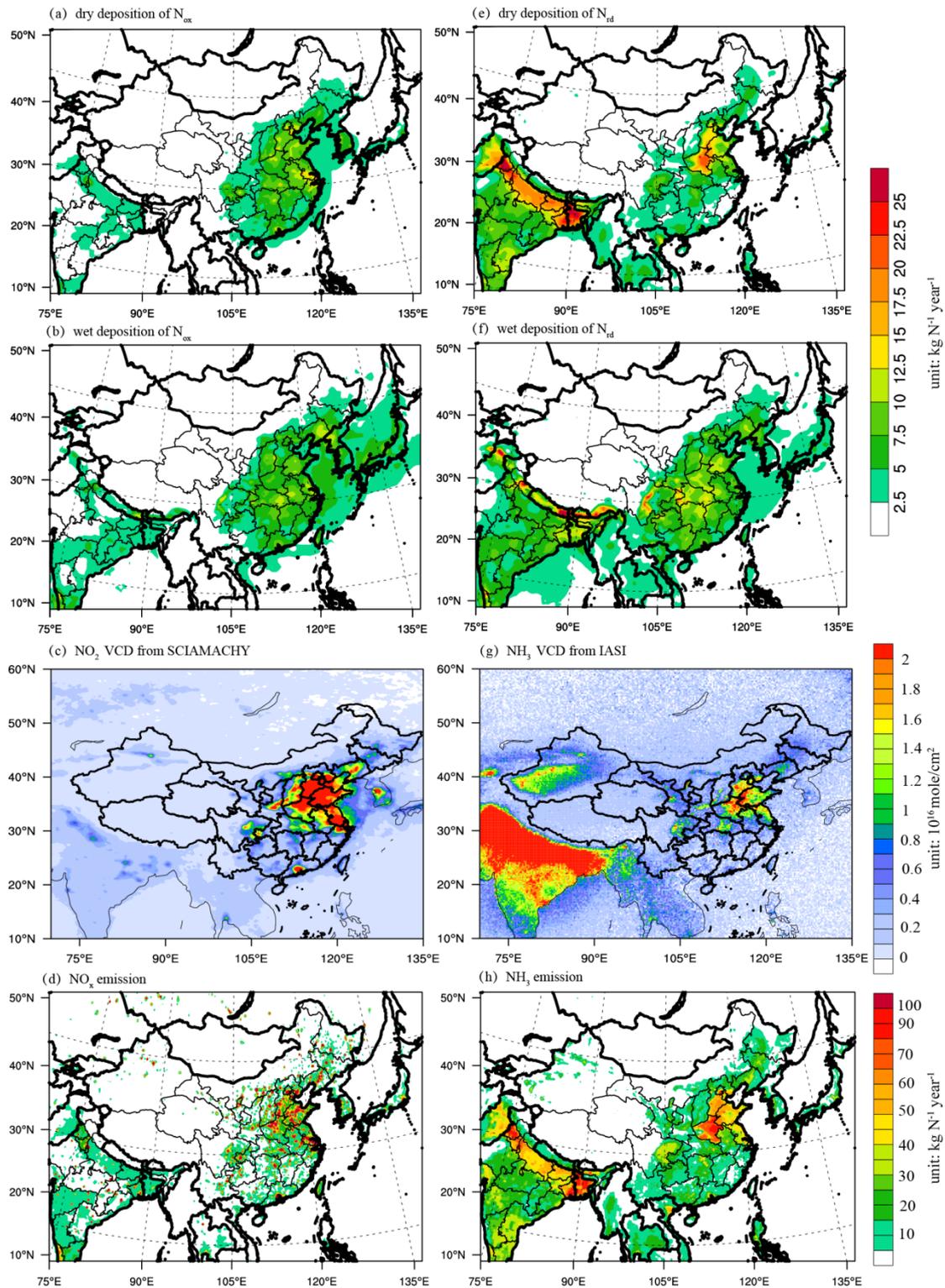


1198

1199 Figure 8: Distribution of CV of NO_x (a), NO_3^- (b), NH_3 (c) and NH_4^+ (d) in the air
 1200 mass for seasonal and annual.

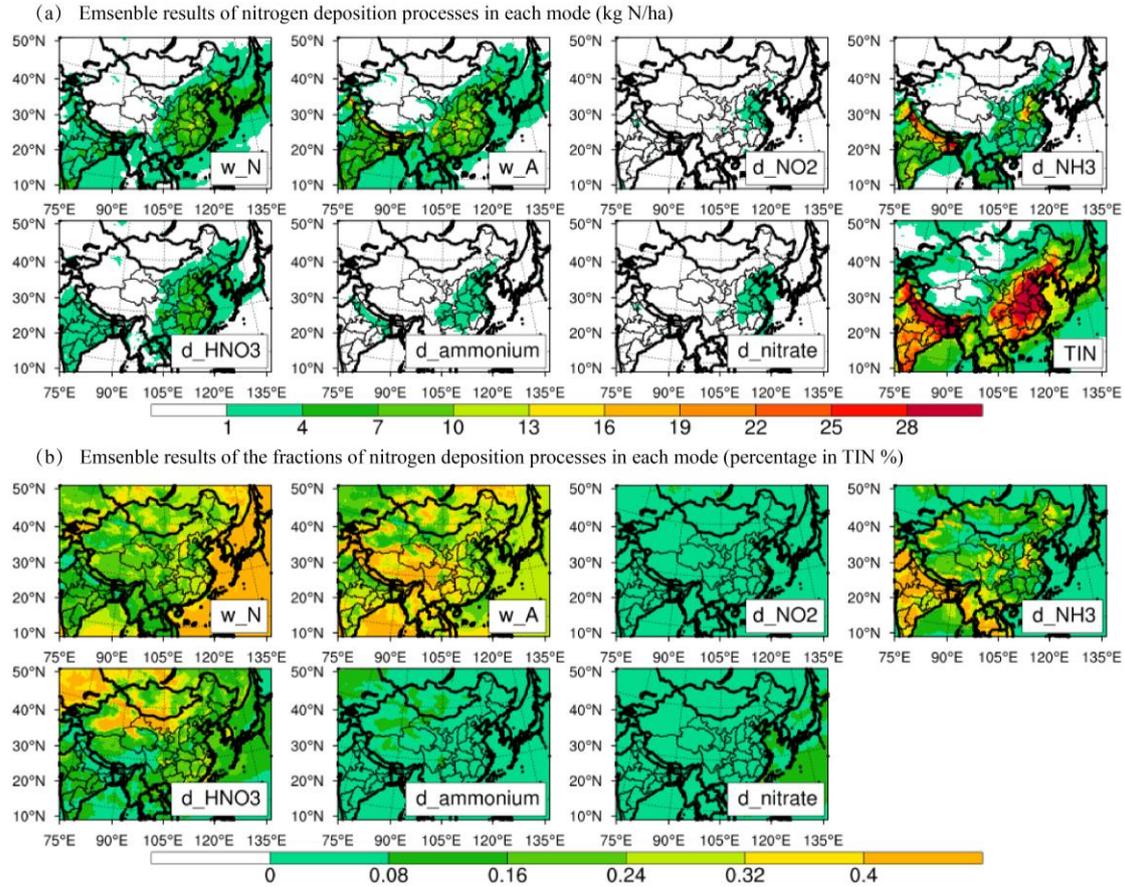
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1203

1204 Figure 9: ENM results for dry deposition (a) and wet deposition (b) of N_{ox} , VCD of
 1205 NO_2 from SCIAMACHY (c) and NO_x emission from MICS-Asia (d); ENM results for
 1206 dry deposition (e) and wet deposition (f) of N_{rd} , VCD of NH_3 from IASI (g) and NH_3
 1207 emission from MICS-Asia (h).

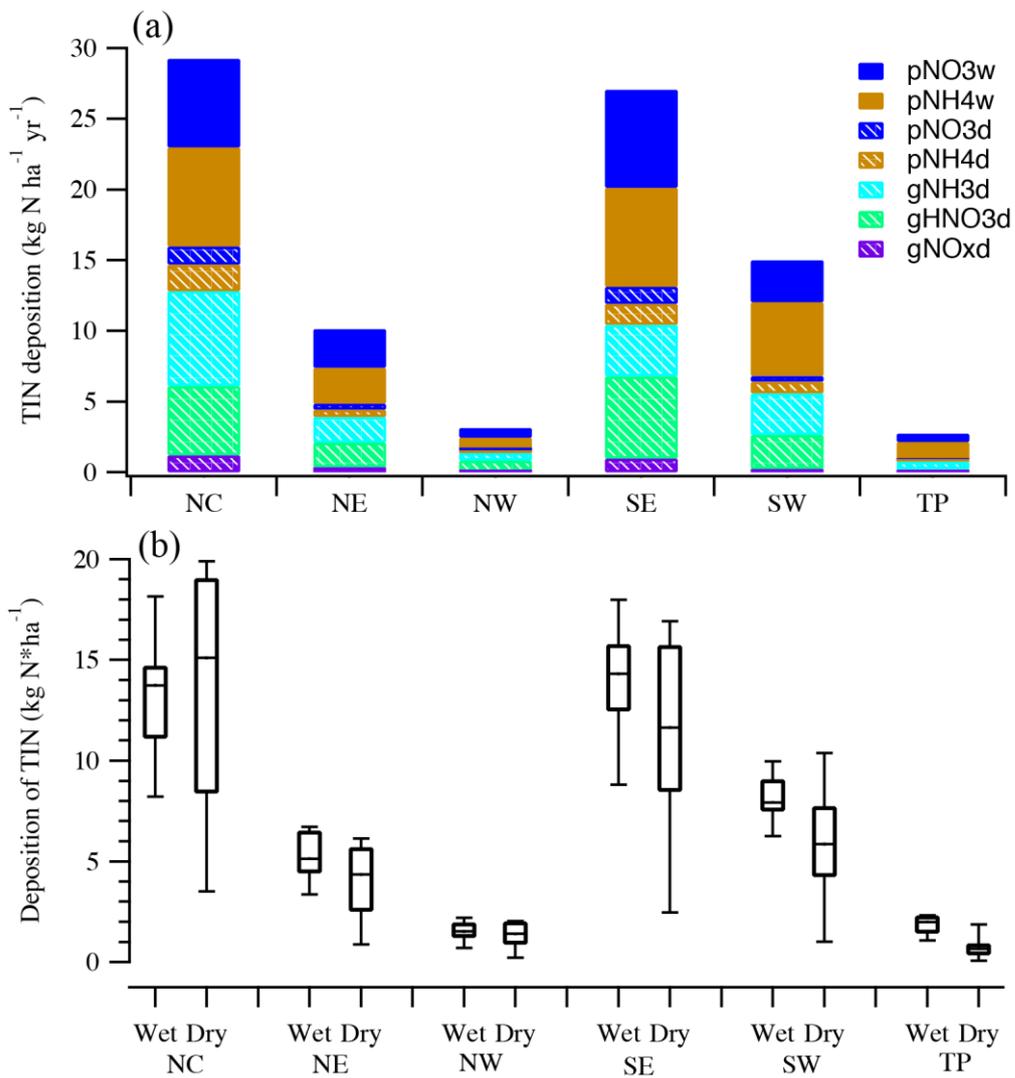


1208

1209 Figure 10: ENM results of each process of N deposition flux (a) and the fraction in
 1210 TIN (b) in MICS-Asia III. The icons w_N, w_A, d_NO2, d_NH3, d_HNO3,
 1211 d_ammonium and d_nitrate represent wet deposition of nitrate, wet deposition of
 1212 ammonium, dry deposition of NH₃, dry deposition of HNO₃, dry deposition of
 1213 ammonium and dry deposition of nitrate, respectively.

1214

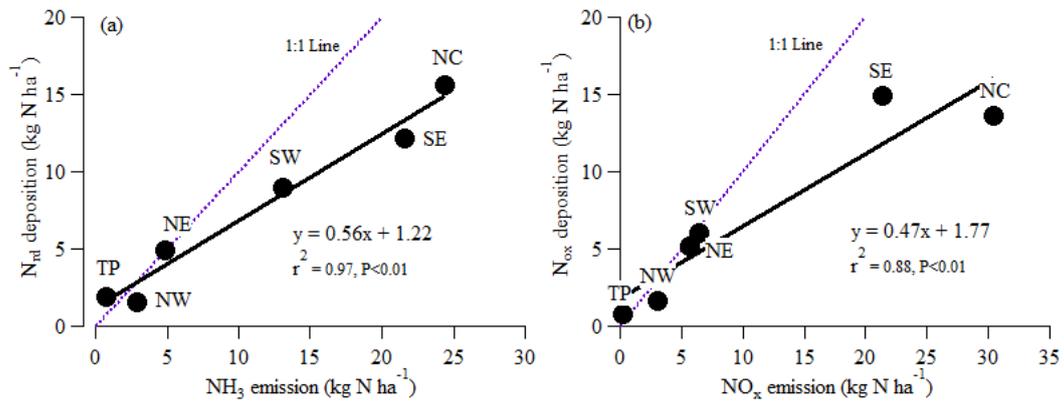
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1216

1217 Figure 11: Pathway of N species to TIN deposition in different regions from ENM
 1218 results (a), and TIN depositions by wet or dry deposited manner (b) in percentile Box
 1219 plot; with 90% and 10% represented for the top and bottom horizontal line, 75% and
 1220 25% represented for the upper and lower edge of the box and asterisk in the middle of
 1221 the box represented for the medium value, respectively.

1222



1223

1224 Figure 12: Relationship of N_{rd} deposition vs. NH_3 emission (a) and relationship of N_{ox}

1225 deposition vs. NO_x emission (b) in each region of China.

1226