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

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Abstract: Atmospheric nitrogen deposition in China has attracted huge public attention in recently years due to the increasing anthropogenic emission of reactive nitrogen (N_r) and its impacts on the terrestrial and aquatic ecosystems. However, limited long-term and multi-site measurements have restrained the understanding on the mechanism of the N_r deposition as well as the chemical transport model (CTM) improvement. In this study, the performance of the simulated wet and dry deposition for different N_r species, i.e., particulate NO_3^- and NH_4^+, gaseous NO_x, HNO_3 and NH_3, have been conducted using the framework of Model Inter-Comparison Study for Asia (MICS-Asia) phase III. Nine Models, including 5 WRF-CAMQ models, 2 self-developed regional models, a global model and a RAMS-CMAQ model, have been selected for the comparison. For wet depositions, observation data from 83 measurement sites of EANET, CREN, CAUDN, NADMN and DEE of China have been collected and normalized to compare with model results. In general, most models showed the consistent spatial and temporal variation of both oxidized N (N_{ox}) and reduced N (N_{rd}) wet depositions in China with the NME around at 50%, which is lower than the value of 70% based on EANET observation over Asia. Both the ratio of wet or dry deposition to the total inorganic N deposition (TIN) and the ratios of TIN to their emissions have shown the consistent results with the NNDMN estimations. The performance of ensemble results (ENM) was further assessed with the measurement from satellite. In different regions of China, the results showed that the simulated N_{ox} wet deposition was overestimated in North East China (NE) but underestimated in south of China (SE+SW), while the N_{rd} wet deposition was underpredicted in all regions by all models. The deposition of N_{ox} have large uncertainties than the N_{rd} especially in North China (NC), indicating chemical reaction process is one of the most importance factors that affecting the model performance. Compared to Critical load (CL) value, the N_r deposition in NC, SE and SW reached or exceeded the reported CL value and exerted serious ecological impacts. The control of N_{rd} in NC and SW and N_{ox} in SE would be effective to mitigate the TIN deposition in these regions. More interestingly, the N_r deposition in Tibet plateau with the high ratio of TIN/emission (~3.0), indicating a significant import from outside should be focused in the future due to its climatical influence to the sensitive ecosystem in whole China.

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

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1 Introduction

Atmospheric Nitrogen (N) deposition is defined that N related gases and particles are deposited via precipitation (wet deposition) and not via precipitation (dry deposition) (Clark and Kremer, 2005). These depositions to the Earth's surface, are either close to the sources or in remote regions (e.g. chemical transformation and long-range transport of oxidized and reduced N, N_{ox} and N_{rd} hereafter), where is located far from human activities and labeled as the N-limited areas (Phoenix et al., 2006;Holtgrieve et al., 2011). Evidences show that the effects of reactive N (N_r=N_{ox}+N_{rd}) deposition to environment are numerous, including decreased biological diversity, increased soil acidification, and lake eutrophication (Clark and Tilman, 2008;Janssens et al., 2010;Holtgrieve et al., 2011;Phoenix et al., 2006;Galloway et al., 2004). Different human activities disturb the natural N cycle in serious manners (Galloway et al., 2004), for example using artificial fertilizer to increase crop production (Erisman et al., 2008) or relying too much fossil fuel for industrial production. The N_r production increased from approximately 15 Tg N yr^{-1} in 1860 to 187 Tg N yr^{-1} in 2005 and more than 50% of them have been reported to deposit onto the ground (Nicolas and Galloway, 2008). In the past two decades, high rates of Nr deposition were widely documented in the developed countries such as America (Fenn et al., 1998) and Europe (Dise and Wright, 1995). Great efforts have been made to fight against these negative effects in USA to decrease NO_x, and the N_{ox} deposition was decreased dramatically in recent years (Li et al., 2016). However, the growing human demand for food and energy at a global scale resulted in an increasing emission of N_r into environment (Galloway et al., 2008), particularly in large developing countries like China and India (Chen et al., 2019a;Liu et al., 2013).

A nationwide estimation of long-term N deposition in China based on the bulk measurements as well as summarizing from reported references in 270 sites by Liu et al. (2013) shown an increasing rate of 0.41 kg N ha^{-1} per year from 1980 to 2010. Different from the increasing importance of N_{rd} deposition due to apparently decreasing of N_{ox} that benefit from the air quality control in USA in past decades (Li et al., 2016), the ratio of N_{rd}/N_{ox} recorded from bulk/wet deposition decreased from 5 in 1980 to 2 in 2010 indicating the more and more important role of N_{ox} in China (Liu et al., 2013). The ratio in highly developed regions such as North China Plain was even lower than 1 in recent years (Pan et al., 2012). However, very limited long-term observations in China challenge our capacity to understand and control the increase of N_r deposition. The published long-term N deposition monitoring network, which includes the Acid Deposition Monitoring Network in East Asia (EANET, http://www.eanet.asia/index.html), the National wide Nitrogen Deposition Monitoring Network (NNDMN) established since 2010 by China Agriculture University (CAU) (Xu et al., 2015), and the Chinese Ecosystem Research Network (CERN) in North China Plain established by Chinese Academy of Science (Pan et al., 2012), the Acid Rain Monitoring Network run by the China Meteorological Administration (CMA-ARMN) (Tang et al., 2007;Tang et al., 2010;Ge et al., 2011), and the National Acid Deposition Monitoring Network (NADMN) (Li et al., 2019b), have been identified with many shortcomings, e.g. the scattered monitoring sites as well as the

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uncompleted recorded data, due to high cost of the measurement against the unstable financial support. Chemical Transport Model (CTM) simulation is another option to offset these drawbacks and also to quantify long-range transport of deposition in a global or regional map (Seinfeld and Pandis, 2006). It is important to know the accuracy of the CTM before it is employed to investigate the spatial and temporal variation of the depositions. Hayami et al. (2008) and Mann et al. (2014) referred that different parameterization set in CTMs may result in large variations, and the multi-model ensemble mean (ENM) shows better performance than most single one (Carmichael et al., 2002;Hayami et al., 2008;Holloway et al., 2008;Wang et al., 2008). Besides, to better localize applications of CTM, the comprehensive evaluations of the strengths and weaknesses of current CTMs for simulating the acid deposition as well as their precursors in a unified framework, with certain regulated rules and same inputs to models, should be more critical and effective.

Model Inter-Comparison Study for Asia (MICS-Asia) gives an opportunity to investigate the CTMs application with different models in Asia. MICS-Asia was initiated in 1998 with the target of long-range transport and deposition in SO_4^{2-} in the first stage (MICS-Asia phase I) (Carmichael et al., 2002) and sulfur, nitrogen and ozone in the second stage (MICS-Asia phase II) (Carmichael et al., 2008). The findings concluded and the methodologies developed in the previous inter-comparison studies undoubtedly contributed to common understandings of the performance and uncertainties of CTMs applications in East Asian (Hayami et al., 2008;Carmichael et al., 2008;Han et al., 2008;Wang et al., 2008). The comprehensive multi-model inter-comparison study on the acid deposition in China is becoming urgent issue as the high emissions in China causing acid deposition in neighboring countries (Lin et al., 2008;Kajino et al., 2011;2013;Itahashi et al., 2018),. In this study, one year simulated N_r depositions, i.e. N_{ox} and N_{rd} in both wet and dry deposited ways to the surface ground, using the framework of MICS-Asia III (MICS-Asia phase III), have been compared with each other and validated by the observed wet deposition from EANET, NNDMN, CREN and the Department of ecological environment (DEE, former named as Environmental Protect Administration, EPA) over the whole China. The rationale of the performance of the ENM results were also discussed by comparing with the Vertical Column Density (VCD) from satellite and the emission inventories. Finally, the uncertainties of the pathways to N_r depositions as well as its ecological impacts have been quantified. The results from this study will not only provide important reference for establishing a suitable N deposition model, the localized application of CTMs in China will also be tested.

2 Framework of intercomparison in MICS-Asia III

2.1 Description of the participant models

In the phase III of MICS-Asia, a total of 14 chemical transport models (CTM, M1-M14) participated in the topic of comparison and evaluation of current multi-scale air quality models (Named as topic 1 in MICS-Asia III). The same number index has been used in the deposition comparison part defined in aerosol and ozone comparison reported by Chen et al. (2019b) and Li et al. (2019a). However, the fully coupled online Weather Research and Forecasting model with chemistry

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(WRF-Chem), which has been indexed as M7-M10, was not included in the deposition comparison part in the overview of model inter-comparison and evaluation for acid deposition in Asia (Itahashi et al., 2019). Briefly, Weather Research and Forecasting model coupled with Community Multi-scale Air Quality (WRF-CMAQ) has been numbered as M1-M6, with different version of v5.0.2 for M1 and M2, v5.0.1 for M3 and v4.7.1 for M4-M6. M11 and M12 were the independent models developed by Japan and China, named as NHM-Chem (Kajino et al., 2019) and the nested air quality prediction model system (NAQPMS), respectively. A global three-dimensional chemical transport model (GEOS-Chem v9.1.3), numbered as M13, was also used as the long-range transport and future change prediction in MICS-Asia III. The last number of M14 was represented as the Regional Atmospheric Modeling System coupled with CMAQ (RAMS-CMAQ). It should be noted that the last two models, i.e., M13 and M14, were not driving by the "standard" meteorological fields from WRF v3.4.1 model. Basic information about the configuration of each model was summarized in Table 1. More detailed description could also be found in previous studies (Itahashi et al., 2019;Chen et al., 2019b;Li et al., 2019a).

2.2 Model inputs and simulation domain

As that mentioned by Chen et al. (2019b), same ("standard") meteorological fields, emission inventories and boundary conditions have been prepared for the CTMs inter-comparison in MICS-Asia III to reduce the uncertainties from model inputs. However, some models such as M13 and M14 were imported "non-standard" inputs due to their specific characteristics. The "standard" meteorological inputs were simulated by WRF v3.4.1 with the initial and lateral boundary conditions from the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) data. Four dimensional data assimilation (FDDA) nudgings have been adopted every 6 hours to improve the accuracy of the meteorological parameters simulation. The assimilated meteorological fields from the Goddard Earth Observing System 5 (GEOS5) of the US 25 National Aeronautics and Space Administration (NASA) (https://gmao.gsfc.nasa.gov) were used to drive M13. The M14 model was driven by RAMS with the same FNL data for nudging as the "standard" WRF simulation, which is developed by Pielke et al. (1992). For the emission inputs, all the participant model were using the same emission inventory, which included the MIX anthropogenic emissions over Asia developed for MICS-Asia Phase III (Li et al., 2017), the biogenic emissions calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.04 (Guenther et al., 2006), and the biomass burning emissions from Global Fire Emission Database (GFED) version 3 (van der Werf et al., 2010). Besides, the SO_2 emissions from volcano were collected from AEROCOM program (https://aerocom.met.no/ DATA/download/emissions/AEROCOM_HC/volc, last access: 11 September 2019, Diehl et al., 2012; Stuefer et al., 2013). MICS-Asia Phase III provided two sets of lateral boundary conditions derived from GEOS-Chem (Bey et al., 2001) and CHASER (Sudo et al., 2002), respectively. The boundary conditions from GEOS-Chem were run with 2.5 °×2 ° resolution and 47 vertical layers, while those from CHASER were run with 2.8 °×2.8 ° and 32 vertical layers. M4, M5, M6, M11 and M12 used the output from CHASER as the boundary conditions, and M1,

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M13 and M14 were from GEOS-Chem. Only M2 used the default boundary condition field provided in CMAQ.

The "standard" simulation domain covers the region of East Asia (15.4°S-58.3°N, 48.5°-160.2°E) with 180×170 grids at 45 km horizontal resolution. M1-M6, M11 and M12 followed "standard" simulation domain, while M13 and M14 employed different modeling domains with 0.5° latitude × 0.667° longitude and 64 × 64 km, respectively. In this study, the analyzed region was only focused in China and all participant models covered it. Therefore, simulated reactive N depositions in each model can be analyzed and compared to show the performance of the participant models. All models output of N depositions have been classified as oxidized N (N_{ox} = $gHNO_3$ + gNO_x + pNO_3^-, including gaseous nitrate acid, NO_x and particulate nitrate) and reduced N (N_{rd} = gNH_3 + pNH_4^+, including gaseous ammonia and particulate ammonium) for comparison.

2.3 Observation data

China has large area with almost 5,500 km from south to north (3.5°-53.3°N) and 5,200 km from west to east (75.5°-135°E), which go through the coastal to inland and through tropical to Frigid Zone. Only 8 sites located in Guangdong, Fujian, Sichuan and Shanxi in EANET were not sufficient to show the real performance of CTMs in China. Besides the 8 EANET sites, 83 sites in total with daily or weekly and even yearly routine recorded data from the CERN (Pan et al., 2012), NNDMN (Xu et al., 2015;Liu et al., 2013) and DEE of Guangdong, Liaoning and Xinjiang province as well as Shanghai have been employed in this study to compare the simulated wet deposition in MICS-Asia III in China. Figure 1 displayed the location of 83 measurement sites as well as the divided regions of the whole China. There were 50 urban sites and 33 rural sites. More detailed information of each measurement site could also be found in Table S1 in the supplementary documents.

The daily wet deposition was measured by wet-only sampler to collect precipitation samples during the rainfall event in EANET. Analysis methods for NO_3^- and NH_4^+ were based on ion chromatography and checked by ion balance and conductivity agreement. Detailed description could be found on manual (EANET, 2010). Daily rainwater samples at 10 sites located in North China Plain were collected using a custom wet-dry automatic collector (APS-2B, Xianglan Scientific Instruments Co., Ltd., Changsha, China) in CREN. Inorganic N, including NO_3^- and NH_4^+, in the precipitation samples was determined using an ion chromatography system (Model ICS- 90, Dionex Corporation, Sunnyvale, CA, USA) and the standard laboratory procedure of LAPC (Wang et al., 2012). The detection limit (DL) of N for this instrument was 5 μg l^{-1}. Detailed description could be found in the research of Pan et al. (2012). The wet/bulk NO_3^- and NH_4^+ deposition data from 25 sites of China Agricultural University Deposition Network (CAUDN), which was renamed as NNDMN in China since 2010, have been collected and reanalyzed as yearly data (Xu et al., 2015;Liu et al., 2013). At all monitoring sites precipitation samples were collected using precipitation gauges (SDM6, Tianjin Weather Equipment Inc., China) located beside the DELTA systems (ca. 2m, DEnuder for Long-Term Atmospheric sampling). After collecting, the samples have been analyzed in CAU's laboratory

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based on the standard laboratory procedure of CAU (Xu et al., 2015). Routine NO_3^- and NH_4^+ wet depositions collected in each rainfall event at 40 sites have been provided by the DEE of Guangdong, Liaoning and Xinjiang province as well as Shanghai city. The analyzed procedure was followed as the laboratory procedure of China National Environmental Monitoring Centre (CNEMC).

All data from daily or rainfall event collecting samples at each type of measurement sites has been summarized and normalized as monthly wet deposition data to compare with the monthly simulation in MICS-Asia III in this study, except the yearly data provided by NNDMN. VCD of NO_2 from SCIAMACHY (http://www.temis.nl/airpollution/no2col) and NH_3 from IASI (http://ether.ipsl.jussieu.fr/ether/pubipsl/iasial2/iasi_nh3) have also been used to compare with the total deposition as well as the emissions.

3 Results

3.1 Validation of wet deposition

3.1.1 Yearly comparison and monthly variation of oxidized N

Yearly simulated wet deposition of N_{ox} has been evaluated by observed nitrate wet deposition in 83 sites over China. Table 2 listed the statistical parameters of simulated wet deposition of N_{ox} compared with the observed data in rural and urban sites of China. In all sites, M1, M5 and M11 overestimated the yearly wet deposition of N_{ox} with Normalized Mean Bias (NMB) of +30.3%, +55.4% and +67.2%, respectively. M6, M12 and M13 simulated almost comparable results with NMB of -6.8%, +0.6% and +0.1%, respectively. M2, M4 and M14 underestimated the yearly wet deposition of N_{ox} with NMB of -38.7%, -10.7% and -47.4%, respectively. The NME was almost around at 50% with highest 82.2% in M11, which is lower than EANET observation over Asia with the value at 70% by Itahashi et al. (2019). However, the correlation coefficients R was around 0.2~0.3 (n=83) which is lower than EANET data (0.3~0.4, n=54) (Itahashi et al., 2019). In order to eliminate influences from rainfall uncertainties (R=0.82), the volume weighted mean (VWM) concentration of N_{ox} in precipitation has also been evaluated. In contrast to the low R value of yearly wet deposition of N_{ox}, the correlation R increased to almost 0.5 for the VWM concentrations. Approximately 50% of model results were corresponded within the percentages within a factor of 2 (FAC2). M1 and M13 performed better agreement with 60% and 59% within FAC2, while M2 and M14 showed only 36% and 45% agreement within FAC2. All of ground surface measurement sites have been divided into 49 urban sites and 34 rural sites according to their location. Overall, all the models showed better performance with the R in 0.2~0.4 and FAC2 in 50%~60% in urban sites than that of R in 0.05~0.3 and FAC2 in 40%~50% in rural sites. This difference may not due to the uncertainties in rainfall simulation, as the simulated VWM concentration of N_{ox} in precipitation may eliminate the rainfall uncertainties, and also shows better agreement in urban than that in rural sites (Table 2).

Figure 2 showed the percentile box plot the yearly wet deposition of N_{ox} simulated by 9 participant models in five regions of China (i.e., North China (NC), Northeastern China (NE), southeastern China (SE), northwestern China (NW), southwestern China and Tibet Plateau (SW+TP)). Site by site validation of both the yearly wet deposition

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and VWM concentration of N_{ox} simulated by each model were also displayed in Figure S1. The model results showed large intra-region or inter-region uncertainties, especially in NC, NE and SE. The highest wet deposition of N_{ox} simulated by M11 was almost 3~4 times of the lowest result simulated by M14 in the above regions (Figure 2). Specifically, two models simulated 30-50% higher of N_{ox} wet deposition, while four models were 20~40% lower compared to the averaged observations in NC with the averaged value 6.5 kg N ha^{-1} a^{-1}. For the wet deposition of N_{ox} in SE and SW+TP, most of the participant models were more than 50% underestimated with the largest underestimation of 75% from M14, even though the precipitation in this region was overestimated. Besides, the divergence of observed N_{ox} wet deposition between different sites in NC, SE and SW, which was shown as the length of the red box in Figure 2a, 2d and 2e, was significantly larger than the multi-models results. The scattered distribution of the measurement sites in these regions was responsible for the large divergence in observations. However, most of the participant models failed to capture the large difference, indicating that the coarse grid in MICS-Asia III (45 km) was not suitable for the performance of detailed characterization at a local scale. A global assessment of the ensemble simulated wet depositions in the Task Force on Hemispheric Transport of Atmospheric Pollutants (TF HTAP) by Vet et al. (2014) also indicated the underprediction of the models in a number of sites in north America, Europe, Central Africa and part of East Asia. The underprediction in Europe was found due to the large underpredictions of precipitation depth, while the reason for East Asia is still unknown. However, most of the models overestimated the wet deposition of N_{ox} in NE. Several models including M1, M5 and M11 simulated more than 10 kg N ha^{-1} a^{-1} N_{ox} wet deposition, almost double higher than the observed value of 5 kg N ha^{-1} a^{-1}. Both the multi-models and the observations showed very low values of 3-4 kg N ha^{-1} a^{-1} N_{ox} wet deposition in NW, where the precipitation depth was very low compared to the other regions of China (Figure S1).

Further evaluations in temporal variations both in urban and rural sites of NC and NE have been displayed in Figure 3. Generally, all of the models and observations performed high level of depositions in spring and summer and low value in winter in the two regions. High depositions were due to large precipitation depth in rainy season. However, this was not always true in some urban sites. For example, higher depositions of N_{ox} were observed in May and June with lower rainfall volume than in July and August with higher rainfall in the urban sites of NC. Similar cases were found at urban sites in NE. Previous studies confirmed there is a decreasing trend in the variations of chemical components in precipitation as the rainfall evolution (Aikawa and Hiraki, 2009;Aikawa et al., 2014;Xu et al., 2017). If the rainfall lasted long enough, or rainfall volume was large enough, the concentrations of chemical components in precipitation remained at low levels and were attributed to the effects of the in-cloud scavenging process. That is, the large rainfall in an event may not cause the high level of monthly wet depositions due to the low level of in-cloud deposition compared to the wet deposited by several different precipitation events, especially in polluted regions in urban sites. Unfortunately, only monthly data of wet deposition as well as precipitation have been compared in this MICS-Asia III.

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Detailed comparison with the rainfall event should be considered in the future.

3.1.2 Yearly comparison and monthly variation of reduced N

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

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

3.2 Map of wet deposition among participant models

3.2.1 Wet deposition of oxidized N

Figure 5 showed the map distribution of the yearly N_{ox} wet deposition simulated by each participant model, the ENM results and the observed results over China. Most models performed the similar spatial pattern with high level of deposition in central to eastern China and low level in western China. However, the threshold value in the Hotspot areas (from light yellow color to orange and red colors) varied significantly among the models and the average is much higher than the Nr deposition threshold value of 10 kg N ha^{-1} to the temperate ecosystems suggested by Bleeker et al.(2011). For example, M1, M5 and M11 simulated very high wet deposition of N_{ox} (almost reach at 20 kg N ha^{-1}) in the middle Yangze River and Yangze River Delta (YRD), basin of Sichuan Province, south of NC and Liaoning Province located in NE. In

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contrast, M2 and M14 failed to perform the relative hotspot N_{wox} in such areas, and M4, M6, M12 and M13 showed the obscure hotspot with small value of 10 kg N ha^{-1}. The significant differences do not only exist between different models but also in the same model CMAQ, i.e., M1, M2, M4, M5 and M6. Since most models were driven by the meteoroidal field and standard emission input except M13 (Geos-Chem) and M14 (RAMS-CMAQ), the differences in simulated N_{ox} wet deposition should come from the CTMs themselves, such as the diffusion and convection process, the oxidation and chemical transformation as well as the wet scavenging and deposition processes. The comparison of the long lifetime specie CO (Kong et al., 2019) and weak chemical activity specie BC (Chen et al., 2019b) revealed that the model uncertainties are less than other species, i.e., O_3 (Li et al., 2019) and NO_3^- (Chen et al., 2019b) which are strong chemical activity and short lifetime in the atmosphere. These results indicated that the chemical reaction process rather than the diffusion and convection process is one of the most important factors affecting the model uncertainties in MICS-Asia III.

3.2.2 Wet deposition of reduced N

Figure 6 showed the map distribution of the reduced N (N_{rd}) wet deposition over China. All of the models performed similar spatial pattern with high values in central and eastern China but low level of deposition in NW and northwestern of NE. Compared with the N_{ox}, little differences of the simulated N_{rd} wet deposition were found among 9 models except M11, which predicted significant lower values. N_{ox} wet deposition of five agricultural dominant provinces including Shandong, Henan, Hubei, Hunan and Anhui is higher than the threshold value of 10 kg N ha^{-1} according to the simulated results by most models. Unfortunately, little observations in these areas made it harder to validate their truthfulness. Evidence showed the high level of N_{rd} wet deposition over the threshold based on the observations in Hebei, YRD and Pearl River delta (PRD). Almost all of the models were under predicted in these areas. Liu et al. (2013) reported the important contribution of N_{rd} to the total N deposition in China based on the long-term national scale of observed nitrogen deposition data. In the agricultural predominant areas, the ammonia emission is the main contribution to the N_{rd} deposition (Liu et al., 2011 AE review; Kang et al., 2016). Thus, although the rarely observation cannot support the simulated high level of N_{rd} wet deposition in agricultural predominant regions, i.e., Shandong, Henan, Hubei, Hunan and Anhui, where the simulated N_{rd} wet depositions are higher than 10 kg N ha^{-1}, it may be more convincing that the N_{rd} wet deposition is higher than the threshold value according to the confirmed underestimation both in agricultural areas (i.e., Hebei) and in non-agricultural areas (i.e., YRD, PRD).

3.3 Comparisons among participant models for reactive N depositions

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

Besides the wet deposition of oxidized and reduced N, dry deposition was also an important process for the total deposition part in China (Liu et al., 2013;Pan et al., 2012). Coefficient of Variation (hereinafter, CV), defined as the standard deviation divided by mean value of all selected model results, with large value denoting lower consistency among the models, is applied for model comparison of simulated reactive

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N depositions both for dry and wet deposition process in MICS-Asia III. Figure 7 shows the distribution of CV for each type of simulated reactive N depositions. Since the low level of mean values of deposition are more likely to associate with higher CV, the gridded CV was only calculated in the area with the simulated depositions higher than 0.5 kg N ha^{-1} (hereafter, analyzed value) in this study. As it is shown in Figure 7, the spatial distribution of CV only covered Eastern China, Southern China and Northeast China, which indicated that the quarterly and yearly fluxes of reactive N deposition in these regions was higher than the analyzed value. For annual case, the CV value of N_{rd} was lower compared with N_{ox} both for dry and wet depositions. This means the multi-model simulations were more consistent in N_{rd} depositions than in N_{ox} depositions. More specifically, the N_{rd} in wet deposition has lowest CV values followed by N_{rd} in dry deposition and then the N_{ox} in wet and dry deposition, which suggested the simulated wet deposition of N_{rd} had less uncertainties than the other types of reactive N depositions.

More complicated patterns were shown in seasonal variations of each type of deposition. The simulated N_{ox} for dry deposition in Figure 7 (a) showed larger uncertainties in southern China (south of 30 ℃N, with the CV > 0.4) than that in northern China (north of 30 ℃N, with the CV <0.3) in all seasons except summer. Similar spatial and temporal patterns of the CV values were found in N_{rd} dry deposition. It is worth noting that the large CV values with the range of 0.4-0.6 were exhibited in Central China (i.e., Henan, Hebei and Shandong provinces) during summer and autumn in spite of the high flux of N_{rd} dry deposition in these regions (map distribution of N_{ox} and N_{rd} dry deposition simulated in 9 participant models was displayed in Figure S3 and Figure S4 of the supplementary documents). This delivered an important message that the uncertainties of the physical and chemical processes in the participant models, including gas-particle equilibrium (Ge et al., 2019), dry deposition parameter scheme (Zhang et al., 2003), transportation as well as the chemical reaction with other acidifying substances (Liu et al., 2019), in the regions of high emission originated from agricultural activities in growing seasons may lead to significant deviation of simulated N_{rd} dry depositions.

For wet deposition of N_{ox}, large uncertainties were located in southern China in summer and autumn with the CV values higher than 0.6 compared with the CV values lower than 0.4 in other regions (Figure 7c). Anyway, this high value of CV was not found in the summer time of simulated N_{rd} wet deposition (Figure 7d). Due to the high portion of summer time flux to the total annual wet deposition, high CV value in N_{ox} contributed to the most important part of the significantly larger annual CV value than that shown in the N_{rd} case. Due to the same rainfall input for the wet deposition in the framework of MICS-Asia III except model 13 and 14, the different CV values for N_{rd} and N_{ox} in same region (i.e., lower CV values of N_{rd} wet depositions in NC, SE and Central China) would attribute to their precursors concentration in the air mass as well as the different wet scavenging processes (Seinfeld and Pandis, 2006). This has been discussed in the following section.

3.3.2 Comparison of precursors in the air mass

As we all know, depositions both from dry and wet part of a certain substance were

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originated from its precursor in the air mass. The uncertainties of the nitrogen related species in the air mass simulated during MICS-Asia III were therefore an important index for estimating the performance of deposition simulations. Figure 8 showed the distribution of CV for NO_x, particulate NO_3^-, gaseous NH_3 and particulate NH_4^+ in the air mass simulated by the 9 participant models during four seasons as well as the annual mean values. There were significant seasonal variations among the spatial patterns of the CV for each type of the N related air pollutants. It is interesting to note that not only the seasonal variations but also the spatial patterns of the simulated precursors' CV were reasonably consistent with those previously shown in the deposition part (Figure 7). For example, high CV values were found in the simulation of particulate NO_3^- in Southern China during summer, reaching to or even higher than 0.8 in SE China (Figure 8b). The high CV values were also found in the summertime of N_{ox} wet deposition (Figure 7c). As the most important precursor of N_{ox} wet deposition (Pan et al., 2012), the correlated consistence between the precursor and the deposition is reasonable. This has also been proved in the distribution of CV values in NO_x (Figure 8a) and N_{ox} dry depositions (Figure 7a) during autumn and winter. However, only uncertainties in precursors cannot explain everything, for example, the high CV values of N_{ox} wet deposition in south China was corresponding to the low CV values of NO_3^- in autumn. Some other factors, such as the scavenging process might be responsible for the unknown-uncertainties. Xu et al. (2017;2019) first compared the below-cloud wet scavenging coefficients based on the different estimation methods and found the magnitude difference between each type of methods. Thus, more detailed comparison such as in-cloud and below-cloud wet scavenging coefficients in each participant model should be carried out in the next phase of MICS-Asia.

As the most important precursor of N_{rd} dry deposition, gaseous NH_3 also showed large CV values in central China during summer time (> 0.6). There were also significant high CV values in south of Yangtze River during autumn and winter period (0.7-0.8 in south of Yangtze River vs 0.3-0.5 in north of Yangtze River). The similar pattern but not significant uncertainty was found in the simulated N_{rd} dry deposition (0.3-0.4 vs 0.2-0.3 in Figure 7b). Different from the particulate NO_3^-, very low CV values were shown in particulate NH_4^+ during summer leading to the less deviation of simulated N_{rd} wet deposition than the N_{ox}. Therefore, the performance of the precursors' simulation was highly correlated with their depositions, while other factors such as wet scavenging process might lead to the unknown uncertainties but this need to be verified in the future.

4 Discussion

4.1 Ensemble results of reactive N deposition and comparison with satellite

Wang et al. (2008) first presented the ENM depositions of acidify species over East Asia based on MICS-Asia II simulations and found that the ENM afford better skill in simulating wet depositions than each single model. In the phase III of MICS-Asia, the ENM value of wet depositions both for N_{ox} and N_{rd} has also been validated by observations and shown in Figure 5l and Figure 6l. The simulated N_{ox} wet deposition and VWA concentration in rainfall exhibited larger dispersion around 1:1 line with the

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correlation coefficients R were 0.23 and 0.54 in 83 sites over China than that found in N_{rd}, which is concentrated around 1:2 line with the correlation coefficients R were 0.32 and 0.8. This implicated the ensemble-mean value of simulated N_{ox} wet deposition has large uncertainties, while N_{rd} wet deposition was under predicted by a factor of two in MICS-Asia III. Compared to each single model, the ensemble-mean showed higher R value than most of single models. However, due to lack of direct observation of dry deposition, the validation for dry and total deposition of reactive N cannot be achieved. Instead, the column densities from satellite and emissions spatial distribution were employed to address the reasonability of the ensemble-mean of four types of reactive N depositions simulated in nine models. As displayed in Figure 9, dry depositions of N_{ox} and N_{rd} were concentrated in NC, YRD and Henan province, which is correspondence to the distribution of their emissions and VCDs, respectively. Meanwhile, wet depositions of N_{ox} and N_{rd} were centered at central China such as Hubei and Hunan province as well as Chengyu regions. Especially, there were high wet depositions of N_{rd} in south west of Hubei province and north east of Chengdu city, where high values of emissions and the VCDs for NH_3 were absent. These regions loading with high wet depositions were mainly due to the high volume of rainfall (for more details at Figure S5) and the long-range transport of acidic substances (Ge et al., 2011).

Another interesting phenomenon was the different allocation of high values between depositions and VCD for N_{ox} and N_{rd}. For example, low depositions were loading in East China with high value of VCD for N_{ox} as the left panel of Figure 9 shown. While as it was displayed in the right panel of Figure 9, large depositions of N_{rd} in East China were corresponding to low level of VCD on the contrary. The whole emissions of NO_x and NH_3 were similar at 8 kg N•ha^{-1} (8.3 kg N•ha^{-1} and 8.7 kg N•ha^{-1} for NO_x and NH_3, respectively) in China statistically from MICS-Asia III emission inventory. This implicated the allocation of both NO_x and NH_3 between the deposition to the surface ground and staying in the atmosphere were conserved to their emission into the air. This conservation data from different sources also implicated the reasonable simulation of depositions for N_{ox} and N_{rd} in MICS-Asia III.

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

Total inorganic N deposition (TIN), which includes the reduced and oxidized forms of inorganic N deposition both from wet and dry processes, has been calculated for estimating its ecosystem effects in this study as they were measured in most cases before (Pan et al., 2012;Liu et al., 2013). Figure 10 and Figure 11 showed the pathway of each type of N deposition to the TIN from spatial distribution view and 6 regions statistical results, respectively. The ENM dry depositions of gaseous HNO_3 and NH_3 were the two major contributors to the TIN, both of which took part in 18% of TIN over the whole country; while the wet deposition of NO_3^- and NH_4^+ were another two main components with the percentage of 23% and 28% (Table 4), respectively. Consistent with that reported in the global assessment under HTAP (Vet et al., 2014) and in nationwide monitoring network (NNDMN) estimation (Xu et al., 2015), the N_{rd} in China dominated the TIN deposition with the averaged percentage reached at 52% for the ensemble results, although slightly lower compared with 60% and 58% in the

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two previous works. The overall contribution of wet and dry deposition to TIN was almost half by half, which is consistent with that reported in NNDMN by Xu et al. (Xu et al., 2015). Considering the total emission, the depositions in whole China took about 67%, 65% and 66% in the 2010 emission of NH_3, NO_x and total N (NH_3-N+NO_x-N), respectively. It is interesting to show that the relationship of the gridded averaged N_{rd} deposition as well as the N_{ox} deposition with their relevant emissions in six regions (shown in Figure 12 with the slope: 0.56, r^2:0.97 for N_{rd} and the slope: 0.47, r^2:0.88 for N_{ox}) were consistent with that reported by Xu et al. (Xu et al., 2015) (slope: 0.51, r^2:0.89 for N_{rd} and slope: 0.48, r^2:0.81 for N_{ox}). Even the increasing trend of the regions from lowest in TP to highest in NC was the same as the previously measurement study. This implicated the spatial distribution as well as the relationships of deposition and emission were comparable with that measured in the NNDMN. Pan et al. (2013) also compared the correlations of the observed depositions vs emissions and attributed the inconsistent distribution between them in NCP to the uncertainties of the emission. However, the patterns of depositions were also influenced by the regional transport besides the emissions. In this study, significant positive correlations of the simulated N_{ox}(N_{rd}) depositions with the correspondingly NO_x(NH_3) emission reflected the control role of the relative emission to the spatial distribution of the depositions. Although most regions were located below 1:1 line of deposition to emission (Figure 12), few regions, such as TP and NE, were close to or above 1:1 line implied the impacts of transport on deposition among the regions.

For regions, the area-averaged deposition of TIN was highest as 29.2 kg N•ha^{-1} 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 and NE, respectively. The TIN in NW and TP were lowest as 3.1 and 2.7 kg N•ha^{-1}. In top brand of two highest regions NC and SE, the deposition of TIN was similar but the pathways to them were different. The N_{rd} deposition (53%) and the dry deposition (54%) contributed more than half portion of TIN in NC, while the N_{ox} deposition (55%) dominated the TIN in SE. Considering the lower ratio of NO_x/NH_3 emission in SE (21.4/21.6, 0.99) than NC (30.4/24.4, 1.25), higher contribution of N_{ox} to TIN in SE indicated more oxidant ratio of the precursors than NC. For more oxidant N species, i.e., HNO_3 and NO_3^-, both dry and wet depositions were higher in SE than that shown in NC (5.8 vs. 4.9 for dry deposition of gaseous HNO_3 and 6.9 vs. 6.3 for wet deposition of particulate NO_3^-). While for less oxidant N and the reduced N, all type of depositions, such as dry deposition of gaseous NO_x, gaseous NH_3 as well as the particulate NH_4^+, were less in SE than NC, except the wet deposition of particulate NH_4^+, which would due to the much higher volume of rainfall in SE (Figure S5). Overall, the oxidant N made the emitted NO_x more easily to be scavenged in SE with the ratio of N_{ox}-deposition/NO_x-emission reaching at 70%, while the reduced N is more likely to be scavenged from its emission with the ratio as 64% in NC. The total ratio of TIN/emission in NC and SE were 53% and 63%, respectively. Compared to the Critical Load (Duan et al., 2001;Zhao et al., 2009;Liu et al., 2011), which is a judgement of the deposited N effects to the ecosystem, the two regions were almost reaching and even exceeding to the CL value (Table 4), indicating serious ecological impacts of the N deposition in NC and SE and should be paid more attention to the

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controlling of the N related species, especially the N_{rd} in NC and N_{ox} in SE.

In the less developed economic and social area of SW, Due to the high emission of NH_3, 60% of the TIN was dominated by N_{rd} deposition. The ratio of NO_x/NH_3 emission reached 0.49 as the more NH_3 emitted from agricultural activity than the NO_x from the fossil fuel consuming. The ratio of wet deposition/TIN was 55%, which was lower than the HTAP comparison during 2000 (60-70%) (Vet et al., 2014) but higher than the results of NNDMN (45%)(Xu et al., 2015). Although the undeveloped society, the TIN deposition was almost reaching at the CL value according to Zhao et al. (Zhao et al., 2009). Besides the high emission of NH_3, high ratio of N_{ox}-deposition/NO_x-emission reaching at 94% reflecting the import of N_{ox} from high emitted area, such as SE and NC, should be attracted our attention in this region. Although the N deposition in TP was not reaching at CL value, which was the lowest in all regions of China with the value of 2.7 kg N•ha^{-1}, the N ecological impacts cannot be neglected since the sensitive ecosystem (Shen et al., 2019) as well as the important climatically influence to whole China. Considering the high ratio of TIN/emission, which were larger than 1 with 3 for TIN, 2.71 for N_{rd} and 4 for N_{ox}, the import from outside was responsible to the N deposition in TP.

5 Conclusion

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

Large deposition of ensemble simulation of N_{rd} deposition in eastern China was corresponding to low level of VCD from satellite measurement, while the case of N_{ox} was just on the contrary. The total emission of NO_x and NH_3 was similar at 8 kg N•ha^{-1} in China. This indicated the allocation of both NO_x and NH_3 between the deposition to the surface ground and staying in the atmosphere were conserved to their emission into the air, which also implicated the reasonable simulation of depositions for N_{ox} and N_{rd} in MICS-Asia III.

Wet deposition of nitrate and ammonium as well as the dry deposition of Gaseous NH_3 and HNO_3 were the important pathway to TIN deposition with the percentage as 18%, 18%, 23% and 28% for ensemble results, respectively. The gridded averaged N_{rd} in China dominated the TIN deposition with the averaged percentage reached at 52%, which was slightly lower than the reported 60% and 58% in HTAP and NNDMN measurements. The contribution of wet and dry deposition to TIN was almost half by half and consistent with that reported in NNDMN. Even the ratio of TIN/emission was also similar with the NNDMN, indicating that the spatial distribution as well as

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the relationships of deposition and emission were comparable with that measured in the NNDMN.

For different regions of China, the simulated N_{ox} wet deposition was overestimated in NE but underestimated in SE and SW, while large uncertainties were shown in NC. Two models simulated 30-50% higher of N_{ox} wet deposition, and four models were 20~40% lower compared with observations in NC. The large divergences do not only exist between different models but also in the same CMAQ model, i.e., M1-M6. For the simulation of N_{rd} wet deposition, all the models were underpredicted in all regions, with the largest underestimation in NC and SE. Different from N_{ox}, almost similar behavior prediction of the less oxidative species such as the N_{rd} wet deposition has been found in CMAQ models, indicating the chemical reaction process is the one of the most importance factors affecting the model uncertainties in MICS-Asia III. Compared to CL value, the reactive N deposition in NC, SE and SW reached or exceeded the reported CL value and indicated serious ecological impacts. The control of N_{rd} in NC and SW and N_{ox} in SE would be effective to mitigate the TIN deposition in these regions. For the lowest reactive N deposition in TP, however, the N ecological impacts cannot be neglected since the sensitive ecosystem as well as the important climatically influence to whole China, especially considering the high ratio of TIN/emission, which was mainly caused by the import from outside. The joint prevention and control of air pollution in China should be carefully considered and implemented in the future.

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Tables

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	NAQPMS	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 ISORROPIA (v2.1)	AERO6 ISORROPIA (v2.1)	AERO5 ISORROPIA (v1.7)	AERO5 ISORROPIA (v1.7)	AERO5 ISORROPIA (v1.7)	ISORROPIA (v1.7)	ISORROPIA (v2.1)	ISORROPIA (2.1)	ISORROPIA (v1.7)
Cloud & Aqueous[e]	ACM-AE6	ACM-AE6	ACM-AE5	ACM_AE5	ACM_AE5	Ge	WC	Jacob	ACM
Dry dep[f]	M3DRY	M3DRY	M3DRY	M3DRY	M3DRY	Wesely	Kajino	Wesely & Wang	M3DRY
Wet dep[g]	Foley	Foley	Foley	Foley	ACM	Ge	Kajino	Liu	Foley
Met	WRF	WRF	WRF	WRF	WRF	WRF	WRF	GEOS-5	RAMS
Emission[h]	standard	standard	standard	standard	standard	standard	standard	standard	standard

[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.

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

[c]: References for the gas phase chemistry are as follows: Bey: Bey et al., 2001; CBMZ: Zaveri and Peters, 1999; SAPRC-99: Carter, 2000.

[d]: References for the aerosol chemistry are as follows: ISORROPIA version 2.1: Fountoukis and Nenes, 2007; version 1.7: Nenes et al., 1998.

[e]: References for the Cloud & Aqueous are as follows: Ge: Ge et al., 2014; WC: Walcek,1986 and Carlton, 2007; Jacob: Jacob, 2000;

[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; Wesely: Wesely, 1989.

[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 al., 2001.

[h]: "standard" indicates the basic emission inventories in Phase III.

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990 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	8.3	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	1.1	0.5	0.8	1.3	0.8	1.5	0.9	1.0	0.6
Rural (N=34)					Oxidized N deposition (kg N ha^{-1})					
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	8.5	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	0.17	0.21	0.21	0.19	0.24	0.37	0.33	0.23
NMB%		30.3%	-38.7%	-10.7%	55.4%	-6.8%	67.2%	0.6%	0.1%	-47.4%
NME%		66.0%	59.5%	54.3%	77.8%	54.3%	82.2%	49.5%	48.3%	56.0%
FAC2%		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	0.51	0.54	0.56	0.53	0.48	0.56	0.46	0.46
NMB%		24.2%	-40.0%	-13.7%	47.6%	-9.5%	70.7%	-2.3%	11.3%	-36.2%
NME%		60.8%	53.1%	49.4%	72.0%	50.1%	92.5%	50.0%	64.4%	57.3%
FAC2%		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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993 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	0.34	0.32	0.41	0.33	0.49	0.05
NMB%		-38.2%	-43.0%	-45.6%	-43.9%	-37.3%	-73.5%	-38.8%	-38.8%	-60.2%
NME%		50.7%	52.2%	52.9%	51.4%	49.8%	73.5%	50.0%	46.3%	64.1%
FAC2%		53.1%	44.9%	42.9%	46.9%	51.0%	16.3%	51.0%	55.1%	34.7%
Mean	10.9	6.7	6.2	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	0.83	0.77	0.86	0.75	0.56
NMB%		-38.0%	-42.6%	-42.2%	-40.6%	-36.1%	-73.8%	-41.1%	-22.1%	-48.8%
NME%		44.5%	47.5%	47.4%	46.0%	43.7%	73.8%	43.7%	46.0%	62.1%
FAC2%		57.1%	51.0%	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
Rural (N=34)	Reduced N deposition (kg N ha^{-1})									
R		0.29	0.29	0.28	0.30	0.32	0.27	0.28	0.52	0.44
NMB%		-14.4%	-22.0%	-21.1%	-18.3%	-13.6%	-62.5%	-23.2%	-19.0%	-46.2%
NME%		48.0%	47.7%	48.1%	46.5%	47.2%	68.5%	45.8%	40.8%	49.4%
FAC2%		70.6%	55.9%	58.8%	61.8%	67.6%	23.5%	61.8%	73.5%	52.9%
Mean	9.0	7.7	7.0	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	0.82	0.80	0.74	0.82	0.69	0.55
NMB%		-27.5%	-34.2%	-31.9%	-29.7%	-26.4%	-69.0%	-33.8%	-20.2%	-47.4%
NME%		37.7%	40.6%	39.0%	36.6%	37.1%	69.6%	37.9%	40.5%	56.9%
FAC2%		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
NMB%		-29.6%	-35.3%	-36.7%	-34.5%	-28.6%	-69.5%	-33.1%	-31.6%	-55.1%
NME%		49.7%	50.6%	51.2%	49.6%	48.9%	71.7%	48.5%	44.3%	58.7%
FAC2%		60.2%	49.4%	49.4%	53.0%	57.8%	19.3%	55.4%	62.7%	42.2%
Mean	10.1	7.1	6.5	6.4	6.6	7.2	3.1	6.8	6.9	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
NMB%		-34.0%	-39.4%	-38.2%	-36.4%	-32.3%	-71.9%	-38.3%	-21.3%	-48.3%
NME%		41.9%	44.9%	44.2%	42.3%	41.2%	72.2%	41.5%	43.9%	60.1%
FAC2%		55.4%	51.8%	47.0%	51.8%	55.4%	13.3%	55.4%	53.0%	32.5%
Mean	1.4	0.9	0.9	0.9	0.9	1.0	0.4	0.9	1.1	0.7

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

Regions		NC	NE	NW	SE	SW	TP	China
Types of deposition	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}/TIN %	This study	53	49	52	45	60	70	52
	NNDMN							58
	HTAP							>60
Wet/TIN %	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	
Emission	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/Emi %	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	

997 1, Duan et al.,2, Zhao et al.,* Liu et al.,,

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Figures and captions

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

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

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

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

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

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

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

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

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

Figure 10: ENM results of each process of N deposition flux (a) and the fraction in TIN (b) in MICS-Asia III. The icons w_N, w_A, d_NO2, d_NH3, d_HNO3, d_ammonium and d_nitrate represented wet deposition of nitrate, wet deposition of ammonium, dry deposition of NH_3, dry deposition of HNO_3, dry deposition of ammonium and dry deposition of nitrate, respectively.

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

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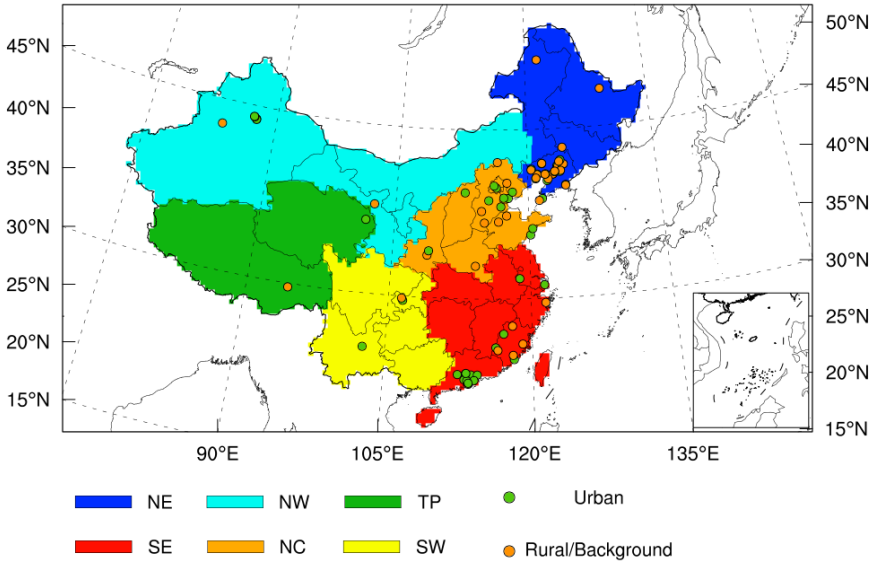
Figure 12: Relationship of N_{rd} deposition vs. NH_3 emission (a) and relationship of N_{ox} deposition vs. NO_x emission (b) in each region of China.



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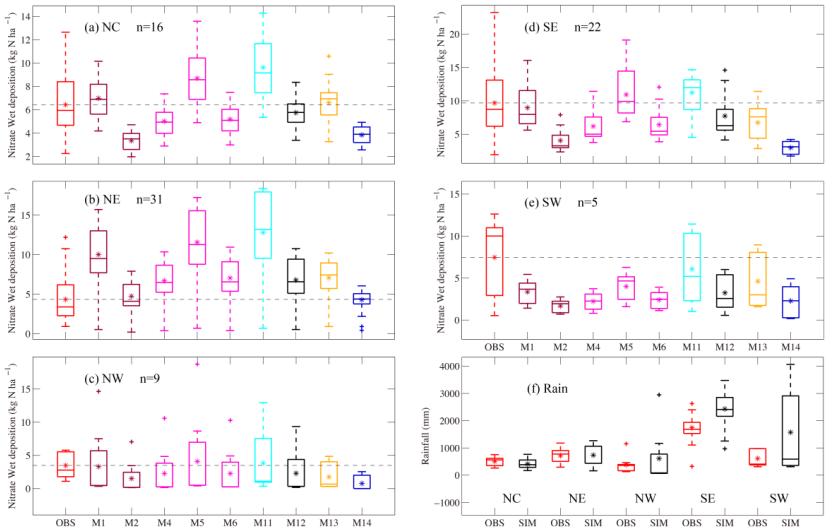

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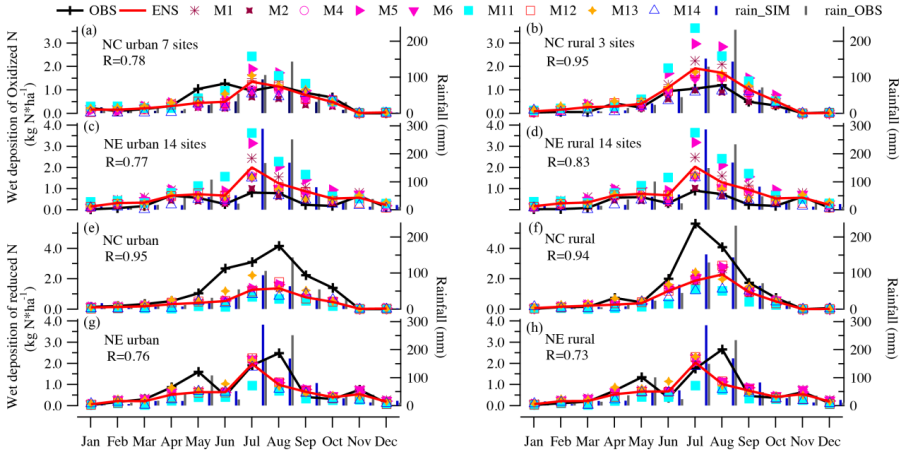

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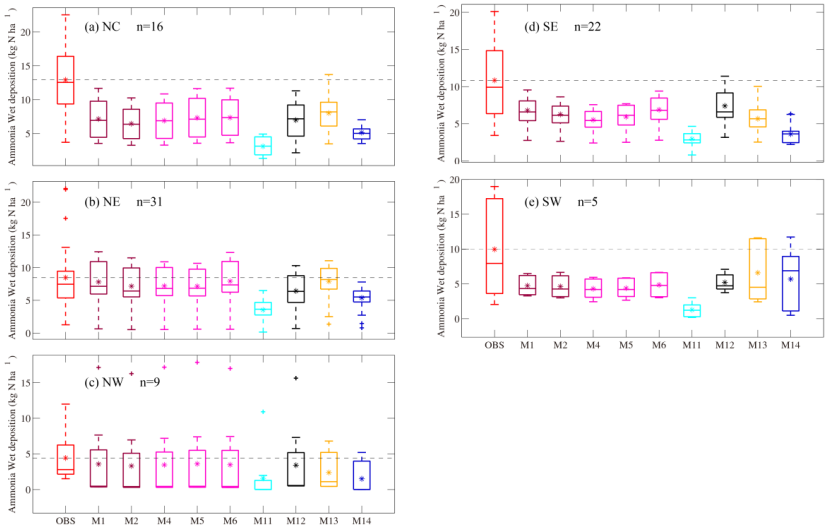

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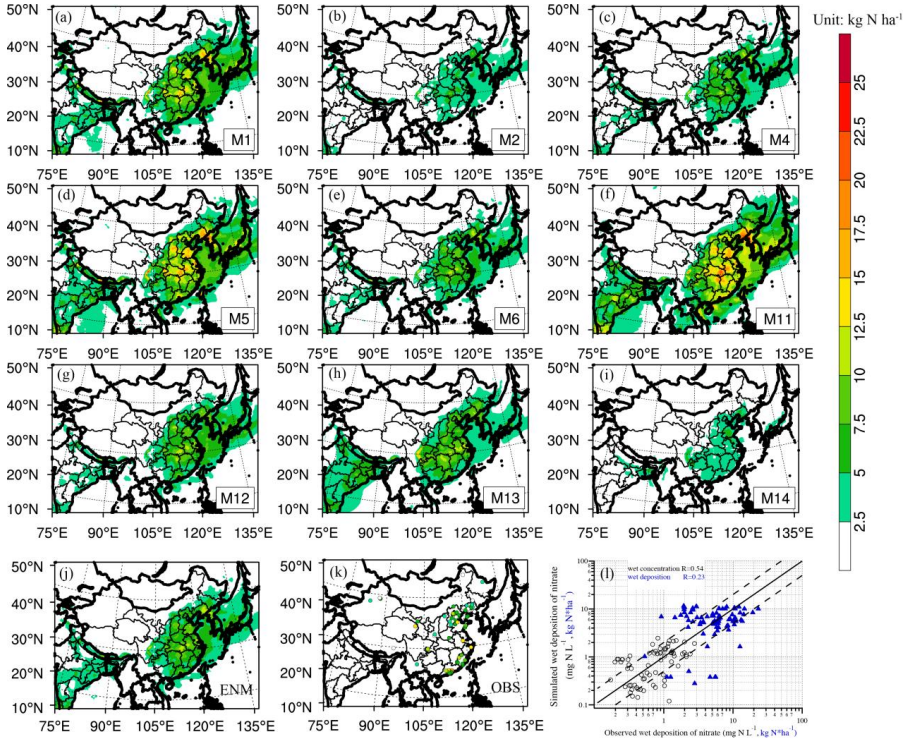

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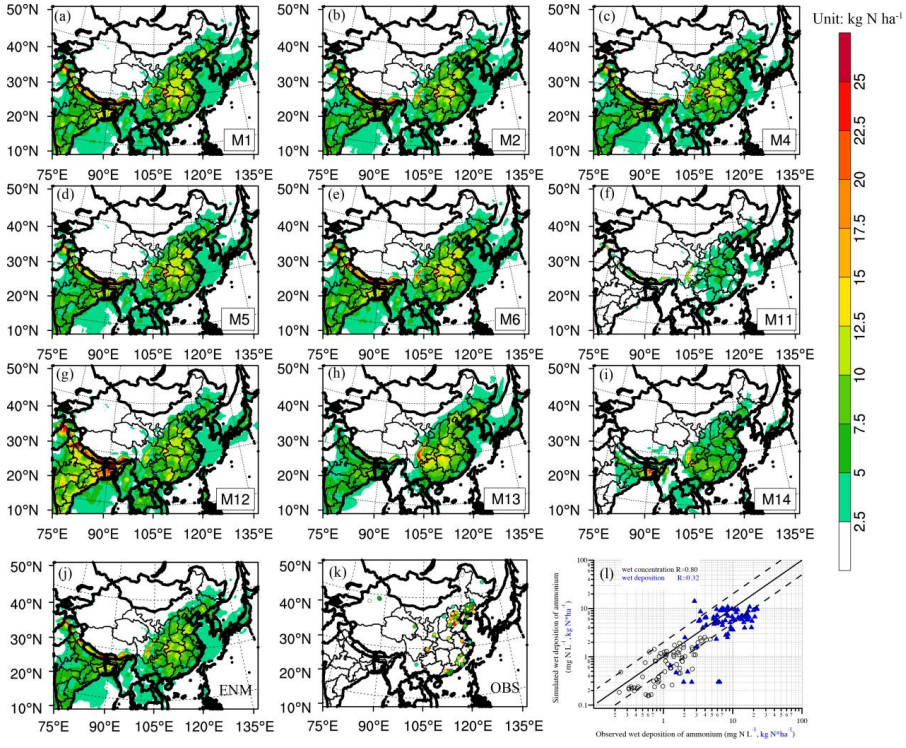

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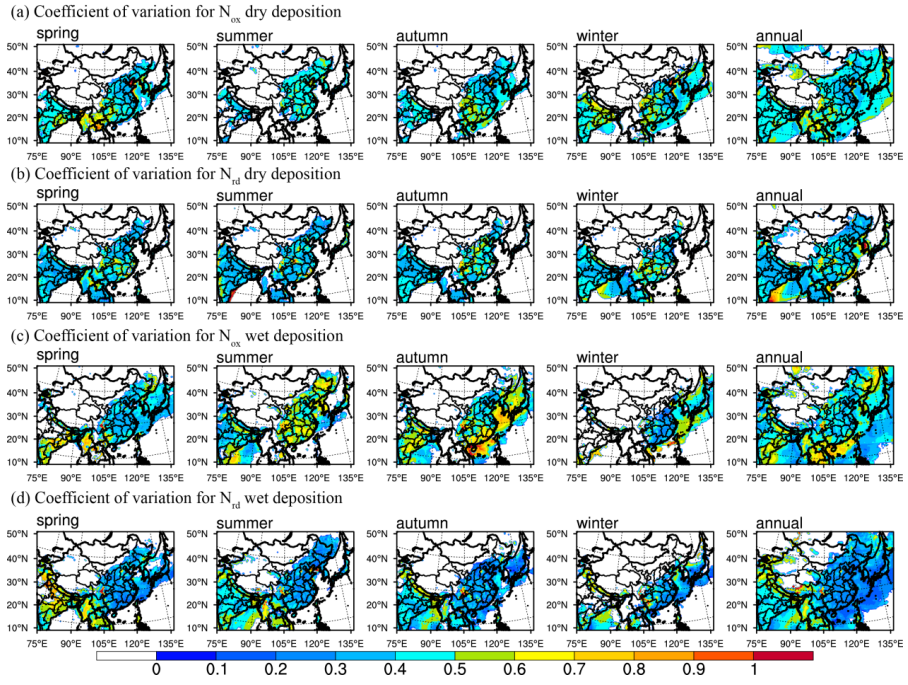

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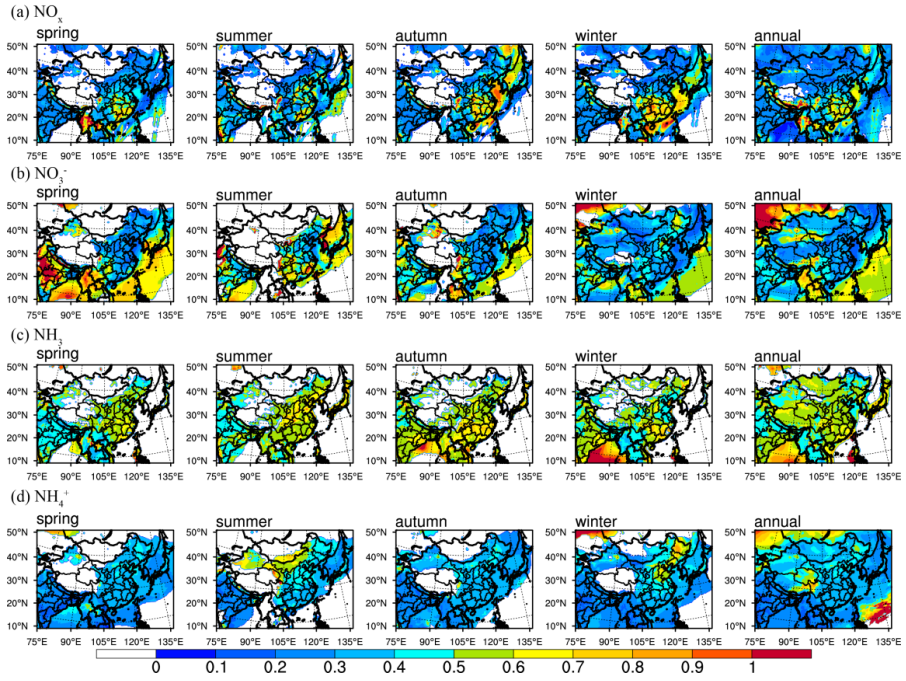

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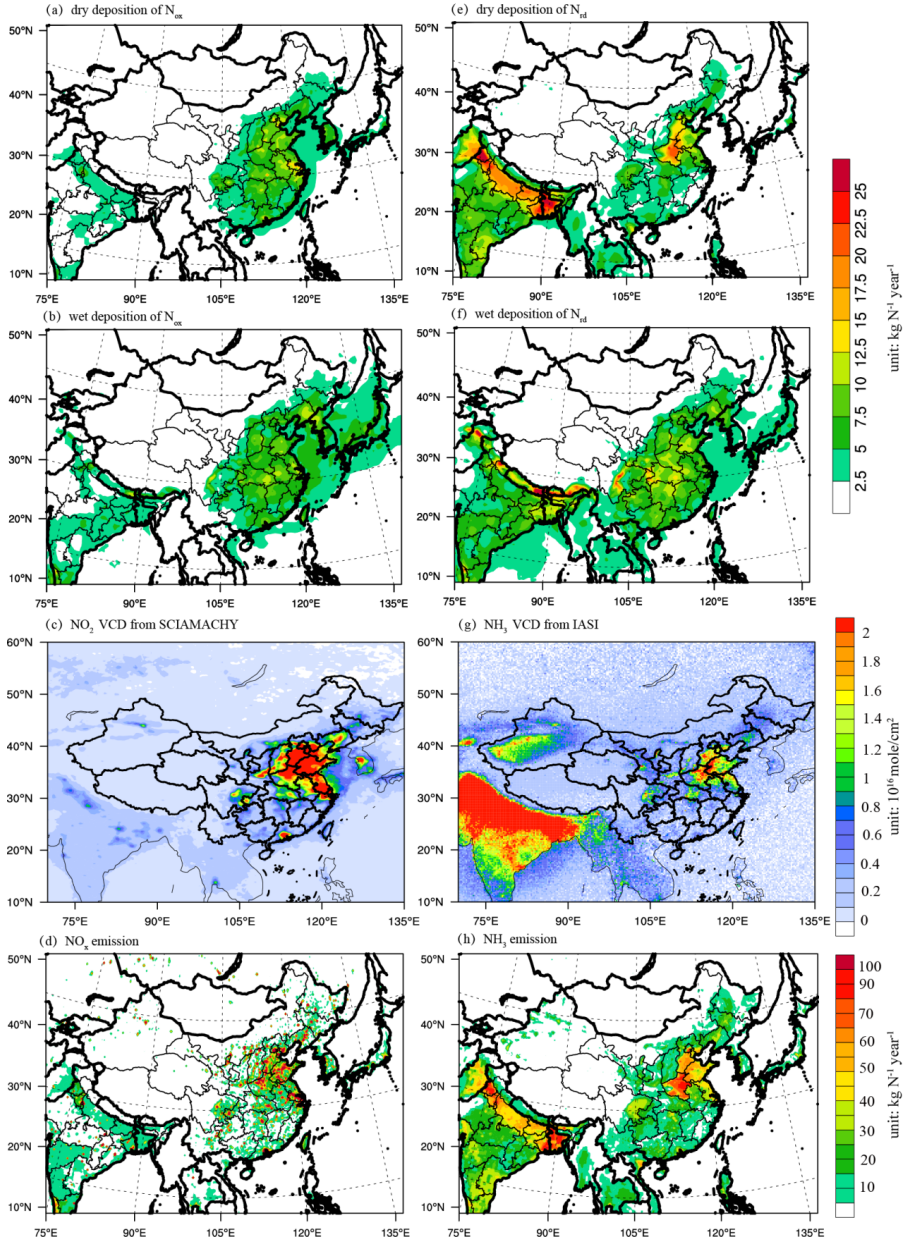

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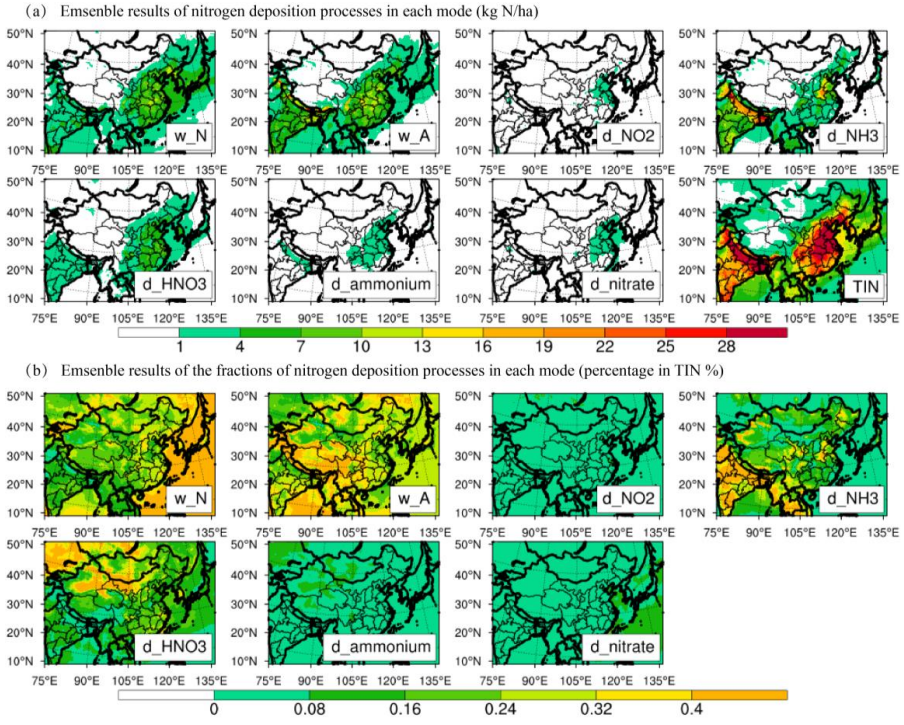

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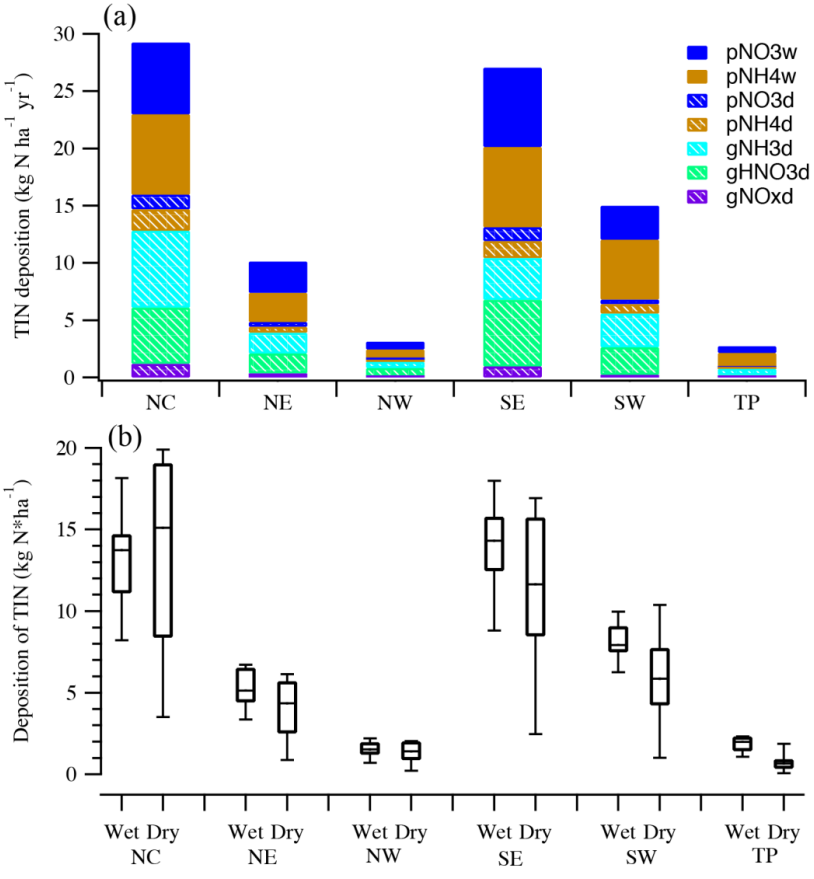

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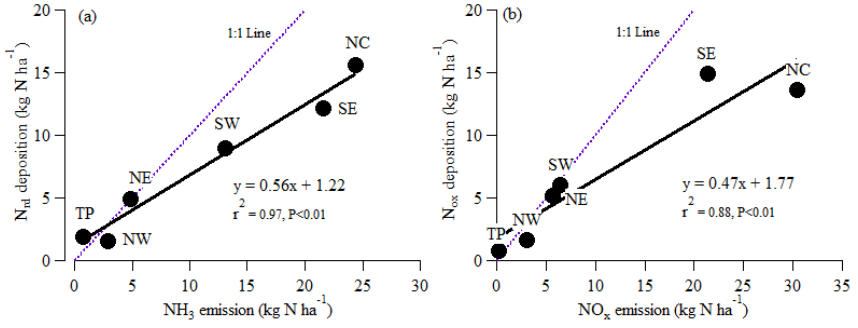

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