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3	Estimating nitrogen and sulfur deposition across China
4	during 2005-2020 based on multiple statistical models
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#### 23 Abstract

24 Due to the rapid development of industrialization and substantial economy, China 25 has become one of the global hotspots of nitrogen (N) and sulfur (S) deposition 26 following Europe and the USA. Here, we developed a dataset with full coverage of N 27 and S deposition from 2005 to 2020, with multiple statistical models that combine 28 ground-level observations, chemistry transport simulations, satellite-derived vertical 29 columns, and meteorological and geographic variables. Based on the newly developed 30 random forest method, the multi-year averages of dry deposition of oxidized nitrogen 31 (OXN), reduced nitrogen (RDN) and S in China were estimated at 10.4, 14.4 and 16.7 kg N/S  $ha^{-1}$  yr<sup>-1</sup>, and the analogous numbers for total deposition were respectively 32 15.2, 20.2 and 25.9 kg N/S ha<sup>-1</sup> yr<sup>-1</sup> when wet deposition estimated previously with a a 33 34 generalized additive model (GAM) was included. The dry to wet deposition ratio 35  $(R_{drv/wet})$  of N stabilized in earlier years and then gradually increased especially for 36 RDN, while that of S declined for over ten years and then slightly increased. The RDN 37 to OXN deposition ratio ( $R_{RDN/OXN}$ ) was estimated to be larger than 1 for the whole 38 research period and clearly larger than that of the USA and Europe, with a continuous 39 decline from 2005 to 2011 and a more prominent rebound afterwards. Compared with 40 the USA and Europe, a more prominent lagging response of OXN and S deposition to 41 precursor emission abatement was found in China. The OXN dry deposition presented 42 a descending gradient from east to west, while the S dry deposition a descending 43 gradient from north to south. After 2012, the OXN and S deposition in eastern China 44 declined faster than the west, attributable to stricter emission controls. Positive 45 correlation was found between regional deposition and emissions, while smaller 46 deposition to emission ratios (D/E) existed in developed eastern China with more 47 intensive human activities.

# 48 **1. Introduction**

49

Atmospheric deposition of nitrogen (N) and sulfur (S) is considered as a serious

50 environmental problem, leading to widespread ecosystem acidification and 51 eutrophication, as well as human health damages (Baker et al., 1991; Burns et al., 2016; 52 Payne et al., 2011; Reuss et al., 1987; Zhang et al., 2018a). In order to understand the 53 spatial distribution and temporal variability of deposition, long-term observation 54 networks have been established globally particularly in developed countries or regions, 55 such as Clean Air Status and Trends Network/the National Atmospheric Deposition 56 Program (CASTNET/NADP) in the USA (Beachley et al., 2016), Canadian Air and 57 Precipitation Monitoring Network (CAPMoN) in Canada (Cheng et al., 2022), 58 European Monitoring and Evaluation Program (EMEP) in Europe (Simpson et al., 59 2012), and Acid Deposit Monitoring Network in East Asia (EANET; Tørseth et al., 60 2012; Totsuka et al., 2005; Yamaga et al., 2021). Reductions of anthropogenic NO<sub>X</sub> and 61 SO<sub>2</sub> emissions in North America have been very effective in reducing the oxidized 62 nitrogen (OXN) and wet S deposition (Cheng and Zhang, 2017; Feng et al., 2021; 63 Likens et al., 2021). In the USA, for example, OXN decreased significantly in most 64 areas, while reduced nitrogen (RDN) increased gradually in agricultural areas (Holland 65 et al., 2005; Li et al., 2016). Similarly, the long-term observation in Europe shows a 66 downward trend for N and S deposition over the last two decades (Keresztesi et al., 67 2019; Theobald et al., 2019).

68 China has become one of global hotspots of atmospheric deposition due mainly to 69 the large anthropogenic emissions from increased industrial economy and energy 70 consumption for the past two decades (Vet et al., 2014). To reduce acid rain and later 71 improve air quality, the Chinese government has enacted a series of policies to cut the 72 emissions of atmospheric deposition precursors since 2005 (Li et al., 2017; Liu et al., 73 2015; Zheng et al., 2018a), including the policy of limiting national total emission levels of SO<sub>2</sub> and NO<sub>X</sub> within the 11<sup>th</sup> Five-year Plan (FYP, 2006-2010) and 12<sup>th</sup> FYP 74 75 period (2011-2015) respectively, the National Action Plan on the Prevention and 76 Control of Air Pollution (NAPPCAP, 2013-2017), and the Three-Year Action Plan to 77 fight air pollution (TYAPFAP, 2018-2020). Estimated by the Multiple-resolution

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78 Emission Inventory for China (MEIC, http://www.meicmodel.org), those policies have 79 reduced annual SO<sub>2</sub> and NO<sub>x</sub> emissions from 2007 and 2012, respectively (Li, 2020; 80 Wang et al., 2022; Zhang et al., 2019), while the change in NH<sub>3</sub> was relatively small. 81 The SO<sub>2</sub> and NO<sub>X</sub> vertical column densities (VCDs) measured from satellite remote 82 sensing have also declined to varying degrees across the country (Krotkov et al., 2016; 83 Xia et al., 2016). Besides emissions and ambient columns, accurate estimation on the 84 changing N and S deposition is crucial for evaluating the effectiveness of national 85 policies on decreasing the ecological risk. Limited by data and methods (explained 86 below), however, few studies have been conducted to link the long-term trend of 87 deposition to the regulations of air pollution prevention.

88 Similar to developed countries, the direct knowledge of deposition in China came 89 first from ground observation. Since 1990s, atmospheric deposition monitoring 90 networks in China have been gradually established and improved, such as the Chinese 91 Nationwide Nitrogen Deposition Monitoring Network (NNDMN; Xu et al., 2019) and 92 the Chinese Ecosystem Research Network (CERN; Fu et al., 2010). They provide 93 essential information for quantifying dry and wet deposition and revealing its long-94 term variability at site level. For example, Liu et al. (2013) found a significant growth 95 in bulk nitrogen deposition in China between 1980 and 2010 based on meta-analyses of 96 historical observation data. Due to insufficient spatial and temporal coverage, however, 97 data obtained at individual sites could not fully support the analysis of widespread and 98 long-term evolution of deposition and might miss diverse patterns of changing 99 deposition by region (Hou et al., 2019; Lye and Tian, 2007). Statistical methods, which 100 incorporated meteorological and environmental variables with higher temporal and 101 horizontal resolutions and wide coverage in time and space (e.g., satellite-derived 102 VCDs), have been increasingly applied to fill the observation gap. Linear or nonlinear 103 relationship between those variables and observed deposition have been developed and 104 applied for periods and regions without observation (Jia et al., 2016; Xu et al., 2018; 105 Yu et al., 2019). For example, Liu et al. (2017a) and Zhang et al. (2018b) obtained the

106 removal rate of  $SO_2$  and  $NO_X$  by precipitation in the whole atmospheric boundary layer 107 through linear regression method, and estimated the wet S deposition in 2005-2016 and 108 nitrogen in 2010-2012 in China. Relatively high uncertainty existed in the simple linear 109 assumption, given the complicated effects of multiple variables (e.g., meteorological 110 conditions and underlying surface types) on deposition. Although advanced statistical 111 methods such as k-Nearest Neighbor (KNN), Gradient Boosting Machine (GBM) and 112 neural networks have been developed to predict the air pollutant concentrations, they 113 are much rarely used in the estimation of deposition (Li et al., 2020b; Li et al., 2019; 114 Qin et al., 2020; Wu et al., 2021). Out of the limited studies, Li et al. (2020a) 115 developed machine learning prediction methods based on multi-sites observation data 116 and integrated meteorological and land use type information, which improved the 117 prediction accuracy of temporal and spatial distribution of ammonium (NH4<sup>+</sup>) wet 118 deposition.

119 Besides spatiotemporal coverage, integrated estimation for multiple species is 120 another great challenge, particularly for dry deposition. Compared with wet or bulk 121 deposition, there are very few data available for direct observation of dry deposition 122 and an "inferential method" that incorporates numerical-simulated dry deposition 123 velocity ( $V_d$ ) and surface concentration has been commonly applied (Cheng et al., 2012; 124 Luo et al., 2016; Wesely, 1989; Xu et al., 2015; Wen et al., 2020). Notably, there are 125 even fewer studies on the dry deposition of secondary-formation species with neither 126 surface nor satellite observation data available at the regional scale (e.g., nitrate ( $NO_3^{-}$ ), NH4<sup>+</sup>, and sulfate (SO4<sup>2-</sup>)). Chemistry transport modeling (CTM), which takes 127 128 mechanisms of secondary formation of atmospheric species into account, is able to 129 provide the temporal and spatial distribution of ambient concentration of those species, 130 thus can potentially be incorporated into the machine learning framework to improve 131 the deposition estimation and complete the information for individual species. Such 132 application (combination of CTM and machine learning in deposition estimation) has 133 been seldom reported to our knowledge.

134 In response to the above limitations, this study aims to develop a machine 135 learning framework for estimating the historical long-term deposition of multiple N and S species at relatively high horizontal  $(0.25^{\circ} \times 0.25^{\circ})$  and temporal resolution 136 137 (monthly) for China, and to explore the comprehensive impact of the national air 138 pollution controls on the deposition. We select the period 2005-2020, which covers three national FYP periods (11<sup>th</sup>-13<sup>th</sup>), NAPPCAP and TYAPFAP. We applied a random 139 140 forest (RF) method and a generalized additive model (GAM) combining different 141 datasets, including ground-level deposition observation, satellite-derived VCDs, meteorological and geographic variables, and CTM simulation, and explore the 142 143 spatiotemporal variability of dry and wet deposition for the country. The ratios of 144 deposition to emissions (D/E) were then calculated by region and species to illustrate 145 the source-sink relationships of atmospheric pollutants. The outcomes provide 146 scientific basis for further formulating emission control strategies, combining potential 147 ecological risks of deposition.

148 **2** 

# 2. Materials and methods

### 149 **2.1 Study domain**

150 We selected Chinese mainland as the research area including 31 provincial-level 151 administrative regions (excluding Hong Kong, Macao and Taiwan). As shown in 152 Figure 1, the 31 provinces are geographically classified into 6 parts, i.e., North Central 153 (NC), North East (NE), North West (NW), South East (SE), South West (SW), and the Tibetan Plateau (TP), representing the diverse social-economical and geo-climatic 154 155 conditions. The details in climate, population and GDP are provided by region in Table 156 S1 in the Supplement. Basically, NC (with Inner Mongolia excluded) and SE belong to 157 the relatively developed regions in eastern China, NW, SW and NE belong to less 158 developed regions, while TP represents the background region. Bounded by the 159 Qinling Mountain-Huaihe River Line (Figure 1), the climate in the south (SE and SW) 160 is humid with more precipitation than the north (e.g., NC).

161 **2.2 Dry deposition flux estimation** 

# 162 2.2.1 Random forest (RF) model description

163 Figure 2 shows the methodology framework of dry and wet deposition simulation. 164 We applied a multisource-fusion RF model to estimate the spatiotemporal pattern of dry deposition for individual N and S species including NO<sub>3</sub><sup>-</sup>, HNO<sub>3</sub>, NO<sub>2</sub>, NH<sub>4</sub><sup>+</sup>, NH<sub>3</sub>, 165  $SO_2$ , and  $SO_4^{2-}$  (H<sub>2</sub>SO<sub>4</sub> is not included due to its tiny amount and unavailability of 166 relevant data), at 0.25°×0.25° horizontal resolution and monthly level for 2005-2020. 167 168 RF model is a state-of-art statistical method to deal with the complicated nonlinear 169 relationship between response variable and interpretation variables. Briefly, with the 170 ensemble learning, the RF regression predictions are determined as the average of the 171 multiple regression trees based on the bootstrap sampling method (Breiman, 2001). 172 The model performance strongly depends on two crucial parameters, *ntree* (number of 173 the regression trees) and *mtry* (number of interpretation variables sampled for splitting 174 at each node), and they were respectively determined at 1000 and 3 to train our model. 175 Not all interpretation variables participate in the process of node splitting (Li et al., 176 2020b), thus significant correlations of regression trees can be avoided. Besides, the 177 backward variable selection was performed on the RF model to achieve the better 178 performance. Please refer to SI Text Section for the detailed algorithm of the model.

We ran the RF modeling program by using the "caret" package in R software (version 4.1.2; Kuhn, 2021). As shown in Figure 2, we firstly applied the inferential method to calculate the dry deposition flux ( $F_d$ ) at ground observation sites as response variable:

183  $F_{\rm d} = C \times V_{\rm d} \tag{1}$ 

184 where C is the estimated (for  $SO_4^{2-}$ ) or observed concentration (for other species)

described in Section 2.2.2, and  $V_d$  is the modeled dry deposition velocity ( $V_d$ ) with the Goddard Earth Observation System-Chemistry (GEOS-Chem) 3-D global transport model described in Section 2.2.4.

Secondly, we selected satellite-derived tropospheric VCDs of SO<sub>2</sub>, NO<sub>2</sub> and NH<sub>3</sub>, 188 surface concentrations of NO3<sup>-</sup>, HNO3, NH4<sup>+</sup>, and SO4<sup>2-</sup> simulated from CTM, 189 190 meteorological factors, geographic covariates, and emission data as interpretation 191 variables. We used the "nearZeroVar" function in "caret" package to eliminate the zero 192 variance variables, to delete highly correlated variables, and to prevent the 193 multicollinearity. Based on the Recursive Feature Elimination (RFE), we then input the 194 final variables to the model as summarized in Table S2 in the supplement. The RFE 195 algorithm is a backward selection of variables based on the relative importance of 196 interpretation variables (RIV). In order to eliminate the different distributions/ranges 197 caused by the magnitudes of various variables, we mapped them to the same interval 198 through standardization and normalization. Before modeling, the interpretation 199 variables were sorted, and the less important factors were eliminated in turn. The RF 200 model captured the nonlinear relationship between the dry deposition  $(F_d)$  and 201 interpretation variables:

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$$F_{\rm d} = \frac{1}{N} \sum_{n=1}^{N} \{\theta(VCDs) + \theta(Mete) + \theta(Emi) + \theta(Geo) + \theta(CTM)\}$$
(2)

where N is the number of samples;  $\theta$  is the random vector; *Mete*, *Emi*, *Geo* and *CTM* represents the meteorology factors, emission data, geography and surface concentrations simulated from CTM, respectively.

Finally, we split the entire model fitting dataset into 10 groups to test the robustness of RF model (10-fold cross validation). In each round of cross validation, the samples in 9 groups were used as the training data, and the remaining group was applied for prediction. This process repeated 10 times and every group was tested. The consistency between the calculated  $F_d$  (as an observation) and predictions was evaluated using statistical indicators, including coefficient of determination ( $R^2$ ), root mean squared prediction error (RMSE), mean prediction error (MPE) and relative prediction error (RPE).

### 214 **2.2.2** Ground-level concentration observations and prediction

215 The daily ground-level concentrations of NO<sub>2</sub> and SO<sub>2</sub> during 2013-2020 were 216 obtained from the real-time data publishing system of the China National 217 Environmental Monitoring (CNEMC, Centre 218 http://datacenter.mee.gov.cn/websjzx/queryIndex.vm), with the abnormal values 219 eliminated. The total number of observation sites reached 1532 in 2020, mainly located 220 eastern China with dense industrial economic and population (e.g., 600 and 408 sites in 221 SE and NC, respectively), as shown in Figure 1. Monthly-level concentrations were 222 then calculated for RF model prediction. The Nationwide Nitrogen Deposition 223 Monitoring Network (NNDMN) established by China Agricultural University contains 224 43 monitoring sites in China (as shown in Figure 1) and measured monthly 225 concentrations gaseous NH<sub>3</sub>, NO<sub>2</sub>, and HNO<sub>3</sub> and particulate NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> in air from 2010 to 2014. The NH<sub>3</sub>, HNO<sub>3</sub>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> concentrations were measured 226 227 using the DELTA active sampling systems (DEnuder for Long-Term Atmospheric sampling), while NO<sub>2</sub> samples were collected with Gradko passive diffusion tubes 228 229 deployed in duplicate or triplicate. The empirically determined effective size cut-off for 230 aerosol sampling was of the order of 4.5 µm (Flechard et al., 2011). The complete 231 datasets of NNDMN were published in previous work (Xu et al., 2019).

Due to the lack of large-scale ground observation data,  $SO_4^{2-}$  concentrations were obtained with an indirect method, according to the strong association between  $SO_2$  and  $SO_4^{2-}$  (Luo et al., 2016). We simulated  $SO_2$  and  $SO_4^{2-}$  concentrations for 2013-2020 with CTM, and developed the relationships between the two with GAM for each year. The  $SO_4^{2-}$  concentrations were then calculated based on the observed  $SO_2$ concentrations from CNEMC and the relationships between  $SO_2$  and  $SO_4^{2-}$ :

238 
$$C_{SO_4^{2-}} = C_{SO_2} \times f(C_{CTM - SO_4^{2-}}, C_{CTM - SO_2})$$
(3)

where  $C_{SO_2}$  is the monthly ground-level concentration at CNEMC for each year of 2013-2020;  $C_{CTM-SO_4^{2-}}$  and  $C_{CTM-SO_2}$  are the SO<sub>4</sub><sup>2-</sup> and SO<sub>2</sub> concentrations simulated by CTM for each year of 2013-2020 (see Section 2.2.4 for CTM description), respectively; and *f* is the relationship between SO<sub>4</sub><sup>2-</sup> and SO<sub>2</sub> obtained from GAM. As shown in Figure S1 in the Supplement, significant positive correlations were found for SO<sub>4</sub><sup>2-</sup> and SO<sub>2</sub> concentrations, with the total correlation coefficient (R) estimated at 0.86 (p<0.001) for 2013-2020.

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### 2.2.3 Satellite-derived VCDs

The tropospheric VCDs of NO2 from 2005 to 2020 were taken from Peking 247 University OMI NO2 tropospheric product version2 (POMINO v2; Liu et al., 2019), 248 249 based on the observation of Ozone Monitoring Instrument (OMI). The VCDs with 250 cloud coverage over 25% were eliminated as high cloudiness would distort satellite 251 detection and increase inversion error. The daily SO2 VCDs were obtained from Level-252 3e OMSO2 2005 Data Products from to 2020 253 (https://disc.gsfc.nasa.gov/datasets/OMSO2e\_003/summary). All the OMI SO2 data 254 were generated by an algorithm based on principal component analysis (PCA), which 255 was considerably sensitive to anthropogenic emissions (Krotkov et al., 2016). The total 256 VCDs of NH<sub>3</sub> were derived from the Infrared Atmospheric Sounding Interferometer 257 (IASI), board on MetOp-A platform. The standard daily IASI/Metop-A ULB-LATMOS 258 total column Level-2 product v2.2.0 is available from 2008 to 2020 (https://iasi.aeris-259 data.fr/nh3 iasi a arch/). The daily total column was excluded when the cloud coverage was >25%, the relative error was >100%, or the absolute error was >5×10<sup>15</sup> 260 molecules cm<sup>-2</sup> (Whitburn et al., 2016). The NH<sub>3</sub> VCDs from 2005 to 2008 were 261 262 estimated based on the linear correlations between NH<sub>3</sub> emission and VCDs during 263 2008-2020.

We used the Kriging interpolation method to fill the missing values, and obtained the spatial pattern of VCDs at the horizontal resolution of 0.25°×0.25°. Monthly-level VCDs were calculated based on the daily products from 2005 to 2020.

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# 2.2.4 CTM model description

We used GEOS-Chem v12.1.1 (http://geos-ch em.org) to simulate the  $V_d$  and the 268 ground-level concentrations of individual species. The GEOS-Chem model is a global 269 270 3-D model of atmospheric composition driven by assimilated meteorological 271 observations from the GEOS of the NASA Global Modeling and Assimilation Office. 272 It is a state-of-the-art, comprehensive, easily accessible global atmospheric 273 composition model that has been widely applied around the world to advance the 274 understanding of human and natural impacts on the atmospheric environment (Bey et al., 2001; Park, 2004; Eastham et al., 2018). A nested version was applied with the 275 276 native horizontal resolution of 0.5°×0.625° over East Asia (70-150°E, 11°S-55°N) and  $4^{\circ} \times 5^{\circ}$  for rest of the world, and the simulated  $V_{d}$  and concentrations within China were 277 spatially interpolated at the resolution of  $0.25^{\circ} \times 0.25^{\circ}$ . The V<sub>d</sub> for 2013-2020 was 278 279 calculated based on a standard big-leaf resistance-in-series parameterization as 280 described by Wesely (1989) for gases and Zhang et al. (2001) for total particles, and 281 applied in estimation of the response variable dry deposition flux. The detailed 282 calculation process is described in the Text Section in the supplement, and the annual averages of V<sub>d</sub> for different species are presented by land use type in Table S3 in the 283 supplement. The simulated concentrations of individual species (NO3<sup>-</sup>, HNO3, NH4<sup>+</sup>, 284 and SO<sub>4</sub><sup>2-</sup>) during 2005-2020 were used as the interpretation variable in RF. The 285 286 simulated concentrations were in good agreement with the ground measurements, with 287 the correlation coefficients ranging between 0.51 and 0.82 and the normalized mean 288 biases within 30% (Chen et al., 2021).

The model was driven by the MERRA-2 assimilated meteorological data provided by the Global Modeling and Assimilation Office (GMAO) at the National Aeronautics and Space Administration (NASA). Meteorology fields such as vertical pressure
velocity, temperature, surface pressure, relative and specific humidity had a temporal
resolution of 3 h, and surface variables (such as sea level pressure, tropopause pressure)
and mixing depths were at 1 h resolution. The model had 47 vertical layers from
surface to 0.01 hPa, and the lowest layer is centered at 58 m above sea level.

296 Emissions in GEOS-Chem were processed through Harvard-NASA Emission 297 Component (HEMCO; Keller et al., 2014). We used the Community Emissions Data 298 System for global anthropogenic emissions, overwritten by the regional emissions 299 inventories in the USA, Europe, Canada and Asia, involving the National Emissions 300 Inventory from EPA (NEI; https://www.epa.gov/air-emissions-inventories/air-pollutant-301 emissionstrends-data), European Monitoring and Evaluation Programme emissions 302 (EMEP; European Monitoring and Evaluation Programme; www.emep.int/index.html) 303 and the MIX inventory that included MEIC over China. Natural NO<sub>X</sub> sources from soil 304 and lightning were also included (Lu et al., 2021).

# 305 **2.2.5 Other data**

The meteorological parameters for 2005-2020, including precipitation, boundary layer height, temperature at two meters, wind speed, wind direction, surface pressure, total column, total column ozone, were downloaded from the European Centre for Medium-Range Weather Forecasts (ECMWF, <u>https://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/</u>) at the resolution of  $0.25^{\circ} \times 0.25^{\circ}$ .

Land-Use and Land-Cover Change (LUCC), Digital Elevation Model (DEM), population density data (POP) and Gross Domestic Product (GDP) were obtained from Chinese Resource and Environment Data Cloud Platform (<u>http://www.resdc.cn/</u>). Except for the DEM, other data were compiled at a five-year interval (2005, 2010 and 2015 for this study). LUCC was generated by manual visual interpretation of Landsat

317 TM/ETM remote sensing image. We calculated the area fractions of different land use 318 in the buffer zone (60 km in diameter around each site). The elevation spatial 319 distribution data (DEM) were extracted from the Shuttle Radar Topography Mission at 320 the 1-km resolution, assuming no variability during the study period. For GDP and 321 POP, datasets with 1-km resolution were developed through spatial interpolation, 322 taking their spatial interactions with land use type and night light brightness into 323 account (Xu, 2017). Linear interpolation was applied to complete the information for 324 all the years within the research period, and all the above-mentioned interpretation 325 variables were resampled to a uniform horizontal resolution of 0.25°×0.25°.

### 326 **2.3 Wet deposition flux estimation**

327 As shown in Figure 2, we applied a nonlinear Generalized Additive Model (GAM) 328 developed in our previous work (Zhao et al., 2022) to estimate the monthly wet deposition of  $SO_4^{2-}$ ,  $NO_3^{-}$  and  $NH_4^{+}$  in China at a horizontal resolution of  $0.25^{\circ} \times 0.25^{\circ}$ . 329 This model considered the linear and nonlinear correlations between the response 330 331 variable (referred to as wet deposition in this study) and the selected interpretation 332 variables (satellite-derived VCDs, meteorological factors and geographic covariates, 333 etc.). If there is no strict linear assumption, the likelihood estimate of the wet 334 deposition was the sum of the smooth function of the interpretation variables:

335 
$$g(\mu_m) = \sum_{i=1}^n f_i(x_{i,m}) + \sum_{p,q} f_{pq}(x_{p,m}, x_{q,m}) + X_m \theta + \varepsilon_m$$
(4)

where g is the "link" function, which specifies the relationship between the response variable  $\mu$  and the linear formulation on the right side of equation;  $f_i(x_i)$  is the nonlinear smooth function that explores the single effect of individual interpretation variable  $x_i$ ; *m* indicates the month; *n* represents the total number of interpretation variables for which single effect was considered in the model;  $f_{pq}(x_p, x_q)$  is nonlinear smooth function that explores the interaction effect of interpretation variable  $x_p$  and  $x_q$ ;  $X\theta$ represents an ordinary linear model component for interpretation variables (elements of

343 the vector *X*) not subject to nonlinear transformations; and  $\varepsilon$  represents the residuals of 344 models. The smooth functions  $f_i(x_i)$  and  $f_{pq}(x_p, x_q)$  are fitted by thin-plate regression 345 splines and tensor product smoothing, respectively. With an assumption of normal 346 distribution, Gaussian distribution and the log link function are applied for the model 347 residuals.

For SO<sub>4</sub><sup>2-</sup>, the observation data of monthly wet deposition were collected from the 348 349 East Asia Acid Deposition Monitoring Network (EANET) as response variables. For 350  $NO_3^-$  and  $NH_4^+$ , the observed monthly bulk deposition collected by the rain gauges at 351 NNDMN served as the response variables. For all the three species, the interpretation 352 variables contained the precipitation, satellite-derived VCDs, PM<sub>2.5</sub> concentrations, 353 total column liquid water, temperature, boundary layer height, forest-cover and urban-354 cover. The data sources and model performance evaluation were described in Zhao et al. (2022). Overall, the 10-fold cross validation R<sup>2</sup> for NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> reached 355 356 0.51, 0.60, and 0.71, respectively, implying the model well captured the spatiotemporal 357 patterns of wet deposition. Although bulk deposition includes a small amount of dry 358 deposition, the deposition in precipitation obtained through GAM was uniformly 359 defined as wet deposition in this work.

# 360 **3. Results and discussions**

### 361 **3.1 RF model prediction performance**

The RF model performances for dry deposition estimation evaluated with 10-fold cross validation are shown in Figures S2 and S3 in the supplement based on CNEMC and NNDMN, respectively. The multi-year average  $R^2$  of N and S species over China were all above 0.7 and the RMSE of all models were less than 1 kg N/S ha<sup>-1</sup> yr<sup>-1</sup> except for NO<sub>2</sub> (1.09 kg N ha<sup>-1</sup> yr<sup>-1</sup>) and SO<sub>2</sub> (6.46 kg S ha<sup>-1</sup> yr<sup>-1</sup>), indicating the satisfying consistency between observation and prediction. However, the model tended to underestimate the high deposition and overestimate the low one possibly because the 369 model algorithm based on the average of all regression trees resulted in relatively weak 370 estimation of the extreme values. The modeling prediction performance of OXN ( $NO_3^{-1}$ , HNO<sub>3</sub> and NO<sub>2</sub>) was better than that of RDN (NH<sub>4</sub><sup>+</sup> and NH<sub>3</sub>) and S (SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>). 371 For example, the R<sup>2</sup> of NO<sub>2</sub>, NO<sub>3</sub>-and HNO<sub>3</sub> were 0.87, 0.73 and 0.78, while those of 372  $NH_3$  and  $NH_4^+$  were 0.71 and 0.65. POMINO, which reduced the bias of the default 373 374 product by the OMI Nitrogen Dioxide Algorithm Team (Krotkov et al., 2019; Liu et al., 375 2019), was demonstrated to be satisfyingly applicable in OXN deposition prediction 376 for China. In addition, the prediction performances of CNEMC were better than those 377 of NNDMN (except for SO<sub>2</sub>), attributed partly to much more monitoring stations for 378 the former. As indicated in our previous work, improved model performance could be 379 expected along with the increased abundance of observation data (Zhou et al., 2021).

380 To evaluate the long-term average deposition from RF modeling, we collected 34 381 studies that quantified the deposition of different species and forms (dry or wet) for 382 China using observational, geostatistical or modal methods (Table S4 in the supplement). As shown in Figure 3, gaseous NH3 and SO2 were identified as the 383 species with largest dry deposition, while  $SO_4^{2-}$  as the species with the largest wet 384 385 deposition. The multi-year averages (2005-2020) of dry deposition for different species estimated in this study were within the range between 25<sup>th</sup> Quantile (Q1) and 75<sup>th</sup> 386 Quantile (Q3) of selected studies except for NH<sub>3</sub> (Figure 3a), but that of SO<sub>4</sub><sup>2-</sup> wet 387 388 deposition closing to Q1 was basically lower compared to existing studies (Figure 3b). Most of the existing studies reported  $SO_4^{2-}$  wet deposition in China for 2001-2005 389 390 when the national control of SO<sub>2</sub> emissions and acid rain was still in its initial stage, 391 while limited data was available for more recent years when sharp declines were found 392 for SO<sub>2</sub> emissions. Therefore, the average of existing studies might potentially 393 overestimate the actual average level of S deposition across the country. Overall, the 394 total deposition of N and S from RF modeling was satisfyingly closed to the median 395 level of the existing studies (Figure 3c), indicating the robustness of deposition

396 estimation.

397 We calculated the shares of different forms and species to the average of national 398 total deposition in 2005-2020 (Figure 4). The dry deposition of N followed an order of 399  $NH_3 > HNO_3 > NO_2 > NH_4^+ > NO_3^-$ , while the wet  $NH_4^+$  deposition was larger than  $NO_3^-$ . 400 As a whole, RDN (58%) was found to contribute more than OXN (42%) to the total N deposition. For S species, the dry deposition of SO<sub>2</sub> was over ten times of SO<sub>4</sub><sup>2-</sup>, while 401 the latter was only species of wet deposition. Dry deposition was estimated to be 402 403 higher than wet for both N and S, with its fraction reaching 70% and 65% within the 404 research period, respectively. The more specific interannual variability and spatial 405 distribution for different forms will be described in Sections 3.2 and 3.3.

### 406 **3.2 Temporal variability in N and S deposition**

407 Based on the newly developed RF method, the average dry deposition of OXN, 408 RDN, total N and S in China were estimated at 10.4, 14.4, 24.9 and 16.7 kg N/S ha<sup>-1</sup>  $vr^{-1}$  from 2005 to 2020, respectively. The total deposition reached 15.2, 20.2, 35.4 and 409 25.9 kg N/S  $ha^{-1}$  yr<sup>-1</sup>, respectively, when the average wet deposition estimated with 410 411 GAM (Zhao et al., 2022) was included. Figure 5a-d illustrates the long-term 412 interannual variability of dry and wet deposition for OXN, RDN, total N and S, 413 respectively. Different temporal trends are found for N and S, due partly to the diverse 414 of their precursor emissions. As indicated by MEIC, China's NO<sub>X</sub> emission control was 415 limited till 2010, allowing annual national emissions to grow 49% from 2005 to 2012 (Figure 5f). The country required installation of selective catalyst reduction (SCR) 416 417 systems from 2011, and NAPPCAP drove fast growing penetration of SCR in the 418 power and cement production sectors, resulting in a 28.6% reduction in the annual total 419 emissions of NO<sub>X</sub> from 2013 to 2020 (Karplus et al., 2018; Li et al., 2018). Similar 420 temporal variability was found for OXN deposition: it was increasing slightly from 14.7 in 2005 to 15.7 kg N ha<sup>-1</sup> yr<sup>-1</sup> in 2012, and then declining to 14.5 kg N ha<sup>-1</sup> yr<sup>-1</sup> 421

422 in 2020 (Figure 5a). The interannual variation in NH<sub>3</sub> emissions has been much smaller 423 than NO<sub>X</sub>, with a slight reduction by 9% from 2005 to 2020 (Figure 5f), attributed to 424 the changes in Chinese agricultural practices, e.g., improved waste management in 425 livestock farming and replacement of highly volatile ammonium bicarbonate with urea 426 in fertilizer types (Liu et al., 2017b; Zheng et al., 2018b). However, the big emission 427 abatement of acidic gases like SO<sub>2</sub> after 2013 was recognized to reduce the sink of NH<sub>3</sub> 428 in the atmosphere and to increase of gas-phase NH<sub>3</sub> concentrations (Liu et al., 2018), 429 resulting in more dry NH<sub>3</sub> deposition (Figure 5b). After 2015, China's RDN deposition 430 became relatively stable, which could be partly explained by the implementation of 431 Zero Increase Action Plan for N fertilizer after 2015 (Liu et al., 2022). As a combined effect of changing emissions and atmospheric conditions, the RDN deposition was 432 estimated to grow from 19.5 in 2005 to 20.6 kg N ha<sup>-1</sup> yr<sup>-1</sup> in 2020. China has widely 433 434 applied flue gas sulfurization (FGD) in the power sector since 2005, and has expanded 435 its application to other industries (such as sintering furnaces and non-electric coal-fired 436 boilers) since 2013, as a part of NAPPCAP (Zheng et al., 2018a). As a result, the 437 annual national SO<sub>2</sub> emissions were estimated to decline by 76% from 2005 to 2020 438 (Figure 5f), and the dry deposition of S by 31% (Figure 5d). The wet deposition was 439 less responsive to emissions than dry deposition, and the growth in precipitation was 440 likely offsetting part of the benefit of emission control on wet deposition (Zhao et al., 441 2022). The total S deposition was calculated to decline 26%, from 28.8 in 2005 to 21.3 kg S ha<sup>-1</sup> yr<sup>-1</sup> in 2020. 442

Shown in Figure 5a-d as well is the long-term interannual variability of the dry to wet deposition ratio ( $R_{dry/wet}$ ) during 2005-2020. Mann-Kendall test (Ahmad et al., 2015; Comero et al., 2014) was applied to evaluate the significance of  $R_{dry/wet}$  trend for N and S, as shown in Table S5 in the supplement. The  $R_{dry/wet}$  of N species kept relatively stable for earlier years and then slightly increased since 2015, with the multiyear average ratios estimated at 2.2, 2.5 and 2.4 for OXN, RDN and total N,

449 respectively. The  $R_{dry/wet}$  of S declined significantly before 2015 and then slightly 450 increased afterwards, with the average ratio estimated at 1.8 for 2005-2020. The 451 growth of  $R_{dry/wet}$  of RDN could be partly attributed to the improved control of acid 452 precursor emissions for recent years. Since 2013, as mentioned above, implementation 453 of NAPPCAP and abatement of SO<sub>2</sub> emissions has reduced the sink of NH<sub>3</sub> in the 454 atmosphere, elevating the free ammonia in the air and thereby  $R_{\rm drv/wet}$  of RDN. 455 Significant negative correlation coefficient between precipitation and R<sub>dry/wet</sub> was found 456 for both OXN (-0.63) and S (-0.64), indicating the influence of precipitation. Notably, precipitation increased at a rate of 6.3 mm yr<sup>-1</sup> in China during 2005-2015 (Figure S4 457 458 in the supplement), motivating the formation of wet deposition of SO<sub>2</sub> that is easily soluble in water. Besides, the general growth of air pollutant emissions (excluding SO<sub>2</sub>) 459 elevated the atmospheric oxidizing capacity, thereby promoting SO4<sup>2-</sup> formation for 460 461 wet deposition. The declining precipitation after 2015 resulted in the reduced wet 462 deposition and thereby enhanced  $R_{dry/wet}$  for OXN and S.

463 Figure 5e shows the long-term interannual variability of the ratio of N to S 464 deposition ( $R_{N/S}$ ) and the ratio of RDN to OXN deposition ( $R_{RDN/OXN}$ ) for different 465 forms during 2005-2020. Growing  $R_{N/S}$  was found for most time within the research 466 period, as China started SO<sub>2</sub> emission control earlier than NO<sub>X</sub> and NH<sub>3</sub>. R<sub>RDN/OXN</sub> 467 indicates the relative contributions of industrial and agricultural activities to N 468 deposition, as the major anthropogenic sources of RDN are animal excrement and 469 fertilizer use in agriculture while those of OXN are fossil fuel combustion in power, 470 industrial and transportation sectors (Pan et al., 2012; Zhan et al., 2015; Zhu et al., 471 2015). R<sub>RDN/OXN</sub> is estimated to be larger than 1 for the whole research period, with a 472 continuous decline from 2005 to 2011 and more prominent rebound afterwards, and it 473 reached 1.5 for total N in 2020. The ratio for dry deposition was larger than the wet 474 one. The declining  $R_{\text{RDN/OXN}}$  resulted mainly from the growth of NO<sub>X</sub> emissions and 475 thereby OXN deposition, driven by the fast development of industrial economy and 476 increasing fossil fuel combustion. The growing  $R_{\text{RDN/OXN}}$  since 2012 was expected to 477 be largely driven by the continuous efforts of NO<sub>X</sub> emission controls, and highlighted 478 the benefit of those efforts on limiting OXN pollution. Regulation on NH<sub>3</sub> emission 479 controls, mainly in agricultural activities, became increasingly important for further 480 alleviating the N pollution.

481 As summarized in Table S6 in the supplement, the annual average deposition of N 482 and S in China was much larger than that for USA estimated by Clean Air Status and 483 Trends Network (CASTNET, https://www.epa.gov/castnet) and National Atmospheric 484 Deposition Programme (NADP, https://nadp.slh.wisc.edu/networks/national-trends-485 network/) and Europe by European Monitoring and Evaluation Programme (EMEP, 486 https://projects.nilu.no/ccc/index.html). According to Vet et al. (2014), the ensemble-487 mean results of 21 global CTMs indicated that eastern China was the region with the highest nitrogen deposition in the world, with a value of 38.6 kg N ha<sup>-1</sup> yr<sup>-1</sup>. Compared 488 489 with USA and Europe, China has not only experienced high deposition of N and S but 490 also featured the greatest increase over the past decade (Du and Liu, 2014; Fu et al., 491 2022; Jia et al., 2016). Figure 6 illustrates the interannual variations of emissions, 492 deposition and  $R_{\text{RDN/OXN}}$  for China as well as the more developed USA and Europe (28) 493 countries). The emission data for the three regions were respectively taken from MEIC, 494 the U.S. Environmental Protection Agency (EPA, https://www.epa.gov/air-emissions-495 inventories/air-pollutant-emissionstrends-data), and European Environment Agency 496 (EEA, https://www.eea.europa.eu/themes/air). As shown in Figure 6a and 6c, the 497 interannual trends in estimated deposition were basically consistent with those in 498 emissions, with observed reduction for both OXN and S deposition over the USA and 499 Europe. With the slowdown in economic growth and the implementation of air 500 pollution control actions for decades (e.g., Clean Air Act (CAA) in the USA and 501 Convention on Long-range Transboundary Air Pollution (CLRTAP) in Europe), the 502 emissions of NO<sub>X</sub> and SO<sub>2</sub> have been reduced by more than 60% and 90% between 503 1980 and 2020, respectively (Constantin et al., 2020; Fowler et al., 2013; Skyllakou et 504 al., 2021; Zhao and Qiao, 2022). However, as a result of the rapidly growing demand 505 for economic development and energy, the fossil fuel consumption and fertilizer 506 utilization increased by 3.2 and 2.0 times during 1980-2010 for China, which ultimately 507 led to an increase in the OXN and RDN deposition from 2005 to 2010 (An et al., 2019; 508 Li, 2020; Liu et al., 2020). Following developed countries, gradually tightened 509 measures of reducing SO<sub>2</sub> and NO<sub>X</sub> have been launched since 2005 and 2011 510 respectively, and the deposition began to decline afterwards.

511 We selected the periods with fast declines in deposition of OXN and S for the 512 three regions and compared them in Table 1. The relative changes in deposition were 513 smaller than those of emissions for all the regions, and greater declines were found for 514 S for both emissions and deposition than OXN. Compared with Europe and the USA, 515 China had the smallest benefit of precursor emission abatement on deposition. For 516 example, the SO<sub>2</sub> emissions in the USA, Europe and China had been cut by 78.4% 517 (2003-2016), 57.6% (2000-2013) and 75.5% (2007-2020) respectively, while S 518 deposition had declined by 72.5%, 49.9% and 27.0%. This may be caused by a lagging 519 response of deposition to emission abatement, which is more prominent in China. 520 Europe and the USA started emission controls earlier than the selected periods, resulted 521 in a smaller gap between the changes in emissions and deposition afterwards. The 522 comparison implies that the effect of short-term emission reduction in China would not 523 immediately be fully reflected in the deposition. As reported by Yamaga et al. (2021), the trend of  $NO_3^-$  to non-sea-salt  $SO_4^{2-}$  concentration ratio in precipitation in Japan 524 525 clearly corresponded to that of the NO<sub>X</sub> to SO<sub>2</sub> emission ratio in China. Therefore, the 526 short-term emission reduction in China was likely to reduce the transboundary 527 deposition to downwind areas (such as Japan) sooner. Under this condition, continuous 528 efforts on emission abatement should be made to achieve substantial reduction in 529 domestic deposition and to further mitigate ecological risks. Along with continuous controls of anthropogenic emissions, moreover, the variation of natural sources (e.g.,
NO<sub>X</sub> from soils) may play a more important role on the changing deposition and
deserves more attentions in the future.

533 Figure 6d presents the interannual changes of R<sub>RDN/OXN</sub> for China, USA, and 534 Europe (28 countries). The  $R_{RDN/OXN}$  in China was higher than those in the other two, 535 with an average of 1.3 in 2005-2020 (0.9 and 1.0 for the USA and Europe during the 536 same period). As a developing country, China is an important food producing country 537 in the world, with a long history of agricultural production and planting. Large 538 agricultural production and relatively weak policy management made China the largest 539 NH<sub>3</sub> emissions in the world, leading to a high proportion of RDN deposition to the 540 total N deposition (Kang et al., 2016; Liu et al., 2022). In contrast, in developed USA 541 and Europe with high level of agricultural mechanization and abundant industry and 542 transportation, the relatively high NO<sub>X</sub> emissions compared to NH<sub>3</sub> resulted in smaller 543  $R_{\text{RDN/OXN}}$  than China.

544 Similar temporal changes in  $R_{\text{RDN/OXN}}$  can be found for USA and China, i.e., 545 decline in earlier years and growth afterwards. For USA, the turning point of R<sub>RDN/OXN</sub> 546 occurred in 1999, 13 years earlier than that of China in 2012. The turning points were 547 closely associated with the introduction and implementation of NO<sub>X</sub> emission controls 548 for the two countries (CAA Amendments since 1990 for the USA and NAPPCAP since 549 2013 for China). While RDN in China has been the major species since 2005, the OXN 550 in the USA was larger than RDN for over 20 years. The  $R_{RDN/OXN}$  kept growing since 551 2000 and exceeded 1 in 2014, indicating a transition of major N species in the 552 deposition. Different from China and the USA, R<sub>RDN/OXN</sub> in Europe kept declining 553 since 2000, and being smaller than 1 after 2013. In many European countries with 554 abundant agricultural activities (such as Netherlands, Germany, Switzerland and 555 France), the chemical fertilizer and livestock breeding release a large amount of NH<sub>3</sub>. 556 Europe attached great importance to the source control of agricultural pollution 557 (though not as strong as for  $NO_X$ ), adopted the economic guidance method for 558 agricultural environmental subsidies, and member states actively assumed the 559 responsibility for governance for decades (i.e., Common Agriculture Policy, CAP; 560 Zhang et al., 2020). Therefore, the control of NH<sub>3</sub> in Europe was ahead of China, 561 resulting in continuous reduction in NH<sub>3</sub> emissions and thereby  $R_{RDN/OXN}$ .

#### 562 **3.3 Spatial variability in N and S deposition**

563 Figure 7 shows the spatial distributions of N and S deposition fluxes during 2005-564 2020. In general, relatively large deposition was found in eastern China with more 565 population and developed industrial economy (e.g., SE and part of NC in Figure 1). Hotspots of dry deposition were commonly located in the north while wet in the south. 566 567 As a joint effect of concentrations and  $V_d$ , high level of OXN dry deposition was 568 estimated in areas with high vegetation cover, such as Yunnan and Fujian province. For 569 S dry deposition, coal-fired boilers for power and heating were intensively distributed 570 in the north, leading to abundant SO<sub>2</sub> emissions and thereby dry deposition. 571 Furthermore, the relatively stable weather conditions with less convection in the north 572 was unfavorable to the dispersion and dilution of pollutants. The emissions were thus 573 liable to be deposited locally. For RDN, the agricultural production, animal husbandry 574 and biomass burning in NC and the northern part of SE led to relatively NH<sub>3</sub> emissions 575 and thereby high dry deposition. The more acidic and humid soils in the south made 576 NH<sub>3</sub> more difficult to release, resulting in lower dry deposition compared to the north. 577 Large wet deposition was mainly found in the south of China associated with the 578 uneven distribution of precipitation. In summer, the air masses in the western Pacific 579 Ocean and the South China Sea were affected by the southeast and southwest monsoon, 580 significantly increasing the rainfall in southeast China. For the total deposition (wet 581 plus dry), the high deposition of OXN and S were located in SE, while RDN and total 582 N were mainly concentrated in NC and the north of SE.

583 As shown in Table S7 in the Supplement, the  $R_{dry/wet}$  of N and S in the eastern 584 China (SE+NC with Inner Mongolia excluded) was smaller than that in western China 585 (NW+TP), attributed mainly to the large precipitation in the former. Given the dry 586 climate and less anthropogenic activities, the pollution was mainly removed from the 587 atmosphere by dry deposition in western country. The  $R_{dry/wet}$  of TP was the highest out 588 of the six regions, with 2.6 and 3.7 for total N and S, respectively. The  $R_{dry/wet}$  in NE, 589 NW and NC was generally higher than that in the south (SE and SW), resulting also 590 from the abundant precipitation in the south. Higher  $R_{RDN/OXN}$  was found in the west 591 (e.g., NW and TP) and lower in the east (Table S7), as more developed industry in the 592 east resulted in relatively large NO<sub>X</sub> emissions and thereby OXN deposition, while 593 farming and animal husbandry shared more in the economy in the west, leading to 594 substantial NH<sub>3</sub> emissions.

595 Figure 8 and Table 2 compare the relative changes of total deposition (wet plus 596 dry) of different species for eastern, western and whole country. The interannual 597 changes of deposition for all species were smaller than that of emissions (Table 2), 598 reconfirming lagging response of deposition to changing emissions as mentioned in 599 Section 3.2. During the period when emissions declined rapidly, the change of 600 deposition has not yet occurred. It should be noted that the emission reduction might be 601 overestimated by MEIC particularly for SO<sub>2</sub> for recent years. Through a "top-down" 602 methodology based on satellite observation, China's SO<sub>2</sub> emissions were estimated to 603 decline 26% from 2011 to 2015 (Qu et al., 2019), slower than the estimation by MEIC 604 at 42%. The relative changes for N and S deposition in eastern China were generally 605 larger than the whole country, indicating the effectiveness of extremely stringent 606 emission controls on those regions with abundant emissions from industrial and traffic 607 sources. The OXN deposition for all the concerned regions shows an invert "V" pattern 608 over time, consistent with the progress of NO<sub>X</sub> emissions control (Figure 8a). The 609 relative annual changes in eastern China (9% in 2005-2012 and -12% in 2012-2020)

610 were generally greater than in western (4% in 2005-2012 and -5% in 2012-2020). 611 More specifically, the turning point for western China was later than the East, likely 612 resulting from later implementation of emission control policies. Most measures were 613 first implemented in the highly developed key regions in east and then applied more 614 widely afterwards. As shown in the Figure 8b and Table 2, RDN deposition was 615 relatively stable before 2012, and the temporal changes in eastern and western China 616 were generally consistent with each other. The lack of comparable control policies for 617 NH<sub>3</sub> and strict policy of acid precursors likely explained the increasing trend in RDN 618 afterwards, with 9% in eastern and 10% in western China between 2012 and 2020. The 619 biggest reduction was achieved for S deposition, and the decline in eastern China was 620 faster than that in the western (Figure 8c). Attributable to the earlier and broader use of 621 FGD at coal combustion sources, greater abatement of SO<sub>2</sub> emissions was achieved 622 than NO<sub>X</sub> or NH<sub>3</sub> over the past decade, leading to the faster reduction in S deposition 623 than in OXN or RDN (Table 2). In addition, the reduction during 2012-2020 (28%, 18% 624 and 21% for the eastern, western and the whole country, respectively) was clearly 625 larger than that during 2005-2012 (3%, 9% and 7%, respectively), indicating the 626 greatly improved SO<sub>2</sub> controls compared to earlier years.

627 The ratio of deposition to emissions (D/E) is used to analyze the interactions 628 between the pollutant sources and sinks. Figure 9a shows the annual mean D/E ratios 629 during 2005-2020 by species and region. The nationwide D/E of OXN, RDN, and S 630 were 1.4, 2.4, and 2.3, respectively. The D/E in eastern China (e.g., NC and SE) was 631 generally smaller than in western China (NW, SW and TP). The low D/E identified 632 those regions as the major sources of air pollutants due mainly to their intensive 633 emissions, likely influencing air pollution levels in surrounding regions. With less 634 industry, energy consumption and population, by contrast, western China received 635 relatively high deposition compared to local emissions, resulting in large D/E. The very 636 high ratio of D/E indicated that TP was strongly influenced by regional pollution 637 transport. The D/Es of RDN in the six regions were higher than that of OXN and S 638 (except for TP). Due to its relatively short life time, most of NH<sub>3</sub> deposits near the 639 source area, while stronger transport and chemical reaction may occur for NO<sub>X</sub> and 640 SO<sub>2</sub> given their longer life time. Significantly positive correlations were found between regional deposition and emissions for all the concern species, with  $R^2$  estimated at 0.81, 641 642 0.92, and 0.78 for OXN (Figure 9b), RDN (Figure 9c), and S (Figure 9d), respectively. 643 The result implies that the N and S deposition to the six regions were strongly 644 dependent on the spatial pattern of anthropogenic emissions.

The annual emissions, deposition and D/E by land use type were displayed in Table S8 in the supplement. High deposition was commonly found in areas with high energy consumption and large emissions, such as urban and construction sites. Associated with different human activities, moreover, the D/E for S and OXN were smaller in urban regions than those in rural ones, whereas that for RDN was slightly larger in urban areas. Transportation and industries resulted in larger NO<sub>X</sub> and SO<sub>2</sub> emissions in urban locales and agricultural activities enhanced NH<sub>3</sub> in rural ones.

652 Figure 10 shows the spatial distribution of multi-year average deposition by 653 season, which was influenced jointly by varying meteorology and emissions. Basically, 654 larger deposition was found in summer than that in winter, and the seasonal difference 655 was particular bigger for N. The deposition in summer was estimated to be 1.9 and 1.6 656 times in winter for OXN and RDN, respectively, while the ratio was much smaller at 657 1.1 for S. As shown Figure S5 in the Supplement, the  $V_d$  of HNO<sub>3</sub> in summer was 4.4 658 times in winter, leading to larger OXN deposition in summer. Moreover, warm weather 659 elevated the volatility of NH<sub>3</sub> in croplands, resulting in greater emissions and thereby 660 deposition in summer. The hotspot of deposition was commonly found in NC and 661 northern SE in summer, while it moved to central SE in winter attributed partly to the 662 prevailing northwesterly wind.

### 663 **3.4 Uncertainties**

664 Uncertainties existed in current analysis. First, the estimated dry deposition or  $V_{\rm d}$ 665 could not be fully examined with sufficient data from direct observation, attributed 666 mainly to the lack of field measurements. Micrometeorological methods can be used 667 for direct observation of dry deposition, including eddy correlation method, gradient 668 method and relaxation vortex accumulation method. Due to the need for extremely fast 669 response instruments and uniform underlying surfaces, those methods have not yet 670 been widely applied in a long-term and extensive manner. We compared the simulated 671  $V_{\rm d}$  in this work with other simulation studies in Table S9 in the Supplement. The values 672 from various CTMs are commonly of the same orders for most cases, while big 673 differences exist in some species/land use types, e.g., NO<sub>2</sub> for coastal regions. The bias could be better quantified when more direct observations of  $V_d$  get available. 674

Second, error may come from ground-level monitoring data. We collected available data from different monitoring networks, and ignored the difference in observed deposition from diverse methods of sample collection and measurement. Moreover, current RF model relied on the data from observation sites, most of which are located in the eastern China with dense population and developed economy. The model accuracy for remote areas (such as NW and TP) should be further evaluated when more observation data get available for those areas.

Third, there was additional uncertainty in the estimation of  $SO_4^{2-}$  dry deposition, 682 as there were limited observed ambient concentrations of  $SO_4^{2-}$  available for estimation 683 of dry deposition, and CTM had to be applied. The conversion of SO<sub>2</sub> to SO<sub>4</sub><sup>2-</sup> is 684 685 influenced by the atmospheric oxidizing capacity, and thereby the NO<sub>X</sub> concentration 686 (He et al., 2014; Ye et al., 2023). Along with economic development and 687 implementation of air pollution controls, the changing emissions of NO<sub>X</sub> as well as 688 some other species (e.g., volatile organic compounds) have altered atmospheric 689 oxidizing capacity within the research period. Using the relationship between 2013 and 26 690 2020 to extrapolate the  $SO_4^{2-}$  deposition for 2005-2020 would potentially bring some 691 uncertainty.

692 Furthermore, bulk deposition obtained from the open precipitation gauge contains 693 part of dry deposition and therefore likely overestimate actual wet deposition. The bias 694 varied by region and was hard to be quantified at the national level. For example, dry 695 deposition was observed to account for around 20% of the bulk at three rural stations in 696 the North China Plain, and the contribution could reach 39% in some urban areas 697 (Zhang et al., 2015; Zhang et al., 2008). In contrast, the difference between bulk and 698 wet deposition of dissolved inorganic nitrogen (DIN) was equal to 12% of the bulk in a 699 rural site in SW (Kuang et al., 2016; Song et al., 2017). Basically, the uncertainty was 700 greater in areas with a higher proportion of dry to total deposition (such as NW and NE 701 areas with less precipitation), and smaller in areas with a lower proportion (such as SE 702 with more precipitation). As SE is the most developed region in China, with relatively 703 high emissions and deposition across the country, the uncertainty from bulk deposition 704 measurement and application is likely of limited impact on the national level or the 705 overall spatial pattern of deposition. Along with continuous development of monitoring 706 networks and increasing availability of deposition data for diverse species, those 707 uncertainties can be further reduced and more accurate deposition estimation can be 708 expected.

### 709 **4. Conclusions**

We developed a full N and S deposition dataset for mainland China at the horizontal resolution of 0.25° for 2005-2020, combining the ground-level observations, satellite-derived VCDs, meteorological and geographic information, and CTM. Based on the newly developed RF method, the annual average dry deposition of OXN, RDN and S in China was estimated at 10.4, 14.4 and 16.7 kg N/S ha<sup>-1</sup> yr<sup>-1</sup>, while the total deposition reached 15.2, 20.2 and 25.9 kg N/S ha<sup>-1</sup> yr<sup>-1</sup>, respectively, with the wet deposition estimated with a GAM model included. The  $R_{dry/wet}$  of N kept relatively

717 stable at the beginning and then gradually increased, especially for RDN, while that of 718 S declined for over 10 years and then slightly increased. Within the whole study period, 719  $R_{\text{RDN/OXN}}$  was estimated to be greater than 1 and clearly larger than that of the USA and 720 Europe, with a continuous decline from 2005 to 2011 and a growth afterwards. The 721 frequent agricultural activities and relatively weak management of manure have 722 resulted in abundant NH<sub>3</sub> emissions and thereby a high proportion of RDN deposition. 723 Improved NO<sub>X</sub> emission control was the main reason for the elevated  $R_{\text{RDN/OXN}}$  for 724 recent years. Compared with Europe and the USA, China had the smallest benefit of 725 precursor emission reduction on deposition. The prominent lagging response of 726 deposition to emission abatement requires a continuous long-term emission control 727 efforts to substantially reduce atmospheric deposition. As a joint effect of emissions 728 and individual meteorological factors, a downward gradient from east to west was 729 found for dry deposition of OXN while from north to south for S. The wet deposition 730 frequently occurred in the south of China, associated with the spatial distribution of 731 rainfall. The deposition of OXN and S declined faster in eastern China than that in the 732 west after 2012, indicating the effectiveness of extremely strict emission control in 733 developed areas with abundant emissions from industry and transportation. The D/E in 734 eastern China was generally smaller than that in west, as the former was the major 735 sources of air pollutants and the latter received relatively high deposition through 736 regional transport. At the national scale, the deposition strongly depended on the 737 spatial pattern of anthropogenic emissions within the regions. The current study 738 broadens the scientific understanding of China's long-term changes in deposition of 739 typical atmospheric species, as well as the influences of human activities and emission 740 controls. More observation and modeling work is recommended for in-depth analyses 741 on the complicated and changing relationship between emissions and deposition for 742 specific species, as well as the consequent varying effects on ecosystem.

#### **Data availability** 743

744 The multiyear deposition data by species at the horizontal resolution of 0.25° will be

745 available at http://www.airqualitynju.com/En/Data/List/Datadownload once the paper

746 is published.

#### **Author contributions** 747

748 KZhou developed the methodology, conducted the research, performed the analyses 749 and wrote the draft. YZhao developed the strategy, designed the research and revised 750 the manuscript. LZhang and MMa provided the support of air quality modeling. WXu 751 and XLiu provided the support of NNDMN data.

#### 752 **Competing interests**

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

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### 1123 **Figure captions**

Figure 1 The research domain of this study. The pink points represent China National Environmental Monitoring Centre (CNEMC) and the green points represent Nationwide Nitrogen Deposition Monitoring Network (NNDMN). The Qinling-Huaihe Line is the boundary between the north and the south of the country.

- 1128 Figure 2 Methodology framework to estimate dry and wet deposition of this study. The
- blue process shows the four steps to establish the RF model. The orange process shows
- 1130 the three steps in establishing a GAM model. See Sections 2.2 to 2.3 of the method
- 1131 section in the text for the acquisition of the preliminary data set.
- 1132 Figure 3 Comparison of deposition between this study and other literatures for dry (a), 1133 wet (b) and total deposition (c). The black cross and the pentagram are the average of 1134 literature-reported results and the multi-year average of this study, respectively. The 1135 boxplots represent the dispersion of deposition collected from literatures. The central 1136 horizontal line, the upper side line, and the lower side line of the box represent the median value, the upper quartile (75<sup>th</sup> Quantile, Q3) and the lower quartile (25<sup>th</sup> 1137 1138 Quantile, Q1). The vertical line extending out of the box represents 1.5 times the 1139 interquartile interval (IQR, i.e., Q3-Q1), and the horizontal lines represent the upper 1140 limit (Q3+1.5IQR) and the lower limit (Q1-1.5 IQR).
- Figure 4 Contribution of different forms and species to the estimated total N and Sdeposition in China.
- Figure 5 The interannual variability of N and S deposition, emissions and component
  proportion in China from 2005 to 2020. The emission data over China were taken from
  MEIC.
- 1146 Figure 6 The interannual variations of emissions, deposition and RDN/OXN for China, 1147 28 Europe countries (EU) and the USA. All the data are relative to the 2005 levels. The 1148 grey dotted lines are a visual guidance for 1.0 on each of the y axes. (a) NO<sub>X</sub> emissions 1149 and OXN deposition; (b) NH<sub>3</sub> emissions and RDN deposition; (c) SO<sub>2</sub> emissions and 1150 sulfur deposition; and (d) RDN/OXN. The emission data were respectively taken from 1151 MEIC, the European Environment (EEA, Agency 1152 https://www.eea.europa.eu/themes/air), and U.S. Environmental Protection Agency 1153 https://www.epa.gov/air-emissions-inventories/air-pollutant-emissionstrends-(EPA,

- 1154 data, while deposition data from European Monitoring and Evaluation Programme
- 1155 (EMEP, https://projects.nilu.no/ccc/index.html) for Europe and Clean Air Status and
- 1156 Trends Network (CASTNET, https://www.epa.gov/castnet) and National Atmospheric
- 1157 Deposition Program (NADP, https://nadp.slh.wisc.edu/networks/national-trends-
- 1158 network/) for the USA.
- 1159 Figure 7 The spatial distributions of N and S deposition flux in 2005-2020.
- 1160 Figure 8 The interannual variations and relative changes of deposition of OXN (a),
- 1161 RDN (b) and sulfur (c) by region. All the data are relative to the 2005 levels. The
- 1162 orange line represents eastern China (SE+NC with Inner Mongolia excluded, see
- 1163 Figure 1 for the region definitions), the blue line represents western China (NW+TP),
- and the red line represents the average level of whole China.
- Figure 9 Annual mean D/E ratio of OXN, RDN and sulfur from 2005 to 2020 in different regions (a) and linear relationship between regional deposition and emissions (b-d).
- Figure 10 The spatial distribution of multi-year seasonal variation of the totaldeposition across 2005-2020.
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# 1171 **Tables**

1172 Table 1 Comparison of relative change rates of emissions and deposition in the 1173 process of pollution control in China, Europe and the USA. The starting and ending 1174 time was selected according to the period of the fastest decline of deposition in China, 1175 and the time period of emission decline was selected according to the reference 1176 deposition. The emission data were respectively taken from MEIC, the European 1177 Environment Agency (EEA, https://www.eea.europa.eu/themes/air), and U.S. 1178 (EPA, Environmental Protection Agency https://www.epa.gov/air-emissions-1179 inventories/air-pollutant-emissionstrends-data, while deposition data from European 1180 Monitoring and Evaluation Programme (EMEP, https://projects.nilu.no/ccc/index.html) 1181 Status for Europe and Clean Air and Trends Network (CASTNET, 1182 https://www.epa.gov/castnet) and National Atmospheric Deposition Program (NADP, 1183 https://nadp.slh.wisc.edu/networks/national-trends-network/) for the USA.

Deletine aleman		Emi	ssions		
Relative change		$NO_X$		$SO_2$	
The USA	-35.9%	(2003-2011)	-78.4%	(2003-2016)	
Europe	-17.3%	(2000-2008)	-57.6%	(2000-2013)	
China	-32.2%	(2012-2020)	-75.5%	(2007-2020)	
		osition			
		OXN	S		
The USA	-26.0%	(2003-2011)	-72.5%	(2003-2016)	
Europe	-11.1%	(2000-2008)	-49.9%	(2000-2013)	
China	-7.1%	(2012-2020)	-27.0%	(2007-2020)	

- 1185 Table 2 The interannual changes in deposition and emissions of N and S by
- 1186 regions for 2005–2020. Eastern China includes NC (Inner Mongolia excluded) and SE,
- and western China includes TP and NW (see Figure 1 for the region definitions). P1

Interannu	Interannual change (units: kg N/S ha <sup>-1</sup> yr <sup>-1</sup> )		Whole China		Eastern China		Western China	
(units: kg l			P2	P1	P2	P1	P2	
	NO <sub>X</sub>	0.60	-0.42	1.12	-1.33	0.63	-0.24	
Emissions	NH <sub>3</sub>	0.08	-0.21	0.08	-0.83	0.09	-0.02	
	$SO_2$	-0.39	-1.24	-2.98	-4.62	0.01	-0.89	
	Total OXN	0.09	-0.15	0.22	-0.41	0.07	-0.08	
Denosition	Total RDN	0.05	0.06	0.06	0.28	0.05	0.22	
Deposition	Total N	0.14	-0.09	0.28	-0.14	0.13	0.14	
	Total S	-0.29	-0.82	-0.34	-1.55	-0.29	-0.60	
Relative ann	Relative annual change to							
2005 (P1)	or 2012 (P2)	P1	P2	P1	P2	P1	P2	
	NO <sub>X</sub>	49%	-31%	17%	-25%	110%	-29%	
Emissions	NH <sub>3</sub>	7%	-15%	2%	-22%	17%	-3%	
_	$SO_2$	-13%	-72%	-25%	-73%	10%	-74%	
	Total OXN	5%	-7%	9%	-12%	4%	-5%	
Denosition	Total RDN	3%	3%	5%	9%	3%	10%	
Deposition	Total N	4%	-2%	7%	-2%	3%	1%	
	Total S	-7%	-21%	-3%	-28%	-9%	-18%	

and P2 indicate 2005–2012 and 2012–2020, respectively.

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#### Figure 1

























