Response to reviewers' comments and main revisions

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Title: Estimating nitrogen and sulfur deposition across China during 2005-2020 based on multiple statistical models

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We thank very much for the valuable comments and suggestions from the editor and reviewers, which help us improve our manuscript. The comments have been carefully considered and revisions have been made in response to suggestions. Following are our point-by-point responses to the comments and corresponding revisions. **Please note that the line/table/figure numbers mentioned following refer to the clean version of the revised manuscript, unless specifically noted.**

Comments from Reviewer #1

Q1. This study aims to develop a machine learning framework for estimating spatial distribution and long-term trend of N and S deposition across China. Estimated dataset during the period from 2005 to 2020 is valuable to understand effects of emission reductions on deposition and N and S input to ecosystems in China. On the other hand, the dataset has considerable uncertainties (as the authors mentioned in section 3.4). The authors should also take the uncertainties into account in other sections. In addition, there are some parts where discussion is insufficient.

Response and main revisions:

We appreciate the reviewer's positive remarks and important comments of this work. Briefly, in the revised manuscript, we have expanded uncertainty analysis and discussion on the overestimation of bulk deposition (please see our response to

Question 3 of reviewer, as well as that to Question 2 of Reviewer #3), and transformation of SO_2 to sulfate (please also see our response to Questions 7 and 28 of Reviewer#2). We have also added necessary discussions in "Materials and methods" and "Results and discussions" sections, based on the reviewer's valuable comments. Please find the point-by-point response and corresponding revisions below.

Q2. To estimate dry deposition flux, deposition velocity (V_d) was calculated by CTM (GEOS-Chem). Current V_d models (resistant models) have large uncertainties, especially for gaseous and particulate Nr components. Therefore, the authors should open V_d calculation in detail. Although the authors indicate gaseous V_d parameterization in L255 (Wesely, 1989) used in this study, aerosol V_d parameterization should be indicated too. General aerosol models output V_d by size. On the other hand, monitored particulate NH_4^+, NO_3^- do not have detailed size information (only the information of cutoff size: $PM_{2.5}$, PM_{10}, or TPM). It is necessary to explain how to treat the aerosol size to calculate the dry deposition based on equation (1). Moreover, calculated V_d values should be indicated. For example, average values of V_d for each land use are very informative for relevant researchers. This will be important information when comparing the dataset with the results of other studies.

Response and main revisions:

We thank and agree the reviewer's important comment. As summarized below, we have included the calculation process for the dry deposition velocities (V_d), average V_d values by land use type, and the consistency between modeling and monitoring in particle size in the revised manuscript and supplement.

Firstly, we have expanded the calculation process of V_d. The process follows a standard big-leaf resistance-in-series model as described by Wesely (1989) for gases and Zhang et al. (2001) for aerosol. The equations and principles have been added **in the Text Section in the revised supplement**. In particular, the aerodynamic resistance to turbulent transfer from the measurement heights (~3 m) to the roughness

height is estimated using the MERRA-2 data. The surface resistance is calculated based on the Global Land Cover Characteristics Data Base Version 2.0 (http://edc2.usgs.gov/glcc/globdoc2_0.php), which defines land types (e.g., urban, forest, etc.) at 1 km × 1 km resolution and is then binned to the model resolution as fraction of the grid cell covered by each land type. Bi-directional NH_3 exchange is not considered in the model.

Secondly, monthly V_d was obtained as the average of hourly values for further estimation of dry deposition flux of N and S species. The annual averages of V_d have been provided by land use type **in a newly added Table S3 in the revised supplement**.

Finally, the consistency in particle size between monitored concentration and modeled V_d has been discussed. In this study, the concentrations of NH_4^+ and NO_3^- aerosols were measured using the DELTA active sampling system (DEnuder for Long-Term Atmospheric sampling; described in detail in Flechard et al., 2011). Briefly, the sampling train consists of four denuders and two filters to collect gaseous and particulate N species, respectively. This series does not apply staged cut-off instruments for aerosol sampling, and the empirically determined effective size cut-off is of the order of 4.5 μm, without particle size distribution (**Lines 226-230 in the revised manuscript**). As NH_4^+ and NO_3^- are mainly distributed in the fine particle mode, the samples collected by the DELTA system are considered to represent the content of total particulate matter. Correspondingly, the V_d simulated with GEOS-Chem indicates the total particles as well. The dry deposition flux of particulate NH_4^+ and NO_3^- was then calculated by multiplying the measured concentrations with simulated V_d.

Table S3 in the revised supplement: The modeled V_d for different land use categories (cm s^{-1}).

Land use categories	HNO_3	NH_3	NH_4	NO_2	NO_3	SO_2	SO_4
Paddy fields	1.63	0.47	0.14	0.20	0.14	0.45	0.14
Dry land	1.42	0.42	0.16	0.17	0.16	0.41	0.16
Forestland	2.55	0.49	0.14	0.24	0.14	0.46	0.14
Shrub forest	1.83	0.45	0.16	0.21	0.16	0.43	0.16
Sparse forestland	1.96	0.47	0.15	0.22	0.15	0.44	0.15
Other forestland	2.17	0.53	0.14	0.22	0.14	0.52	0.14
High coverage grassland	1.29	0.36	0.18	0.12	0.18	0.36	0.18
Medium coverage grassland	1.05	0.34	0.18	0.09	0.18	0.34	0.18
Low coverage grassland	0.88	0.31	0.17	0.04	0.17	0.31	0.17
River channel	1.17	0.38	0.15	0.13	0.15	0.37	0.15
Lakes	0.93	0.32	0.19	0.07	0.19	0.32	0.19
Reservoir pond	1.37	0.43	0.14	0.14	0.14	0.43	0.14
Permanent glacial snow	0.59	0.27	0.16	0.03	0.16	0.28	0.16
Tidal-flat	1.06	1.01	0.07	0.02	0.07	1.02	0.07
Beach land	1.30	0.37	0.16	0.12	0.16	0.36	0.16
Urban land use	1.37	0.44	0.15	0.14	0.15	0.44	0.15
Rural settlements	1.27	0.40	0.15	0.14	0.15	0.40	0.15
Other construction land	1.40	0.47	0.14	0.16	0.14	0.47	0.14
Sand	0.87	0.30	0.10	0.02	0.10	0.30	0.10
Gobi	0.98	0.30	0.10	0.02	0.10	0.30	0.10
Saline alkali soil	0.87	0.31	0.15	0.04	0.15	0.31	0.15
Swamp land	1.61	0.38	0.15	0.14	0.15	0.37	0.15
Bare land	1.10	0.30	0.10	0.03	0.10	0.30	0.10
Bare rock	0.88	0.30	0.14	0.03	0.14	0.30	0.14
Other unused land	0.72	0.29	0.18	0.04	0.18	0.30	0.18

Note: Land-Use and Land-Cover Change (LUCC) data were obtained from Resource and Environment Data Cloud Platform (http://www.resdc.cn/), generated by manual visual interpretation of Landsat TM/ETM remote sensing images.

Q3. This study uses wet deposition of SO_4^{2-} (EANET) and wet or bulk deposition of NO_3^-, NH_4^+ (NNDMN). There is a need to discuss which regions the overestimation of NO_3^-, NH_4^+ by bulk sampling may affect in "3 Results and discussion".

Response and main revisions:

We thank and agree the reviewer's important comment. We have expanded the discussion on the uncertainty from bulk deposition sampling **in Lines 697-705 in the revised manuscript**. The uncertainty was greater in areas with a higher proportion of dry to total deposition (such as NW and NE areas with less precipitation), and smaller in areas with a lower proportion (such as SE and SW with more precipitation). For example, we compared the results from Kuang et al. (2016) and Song et al. (2017), and found that the difference between 5-year in situ measurements of bulk and wet dissolved inorganic nitrogen (DIN) deposition was only about 2.5 kg N ha^{-1} yr^{-1} at a rural site in SW, equaling to 12% of annual bulk deposition. As SE is the most developed region in China, with relatively high emissions and deposition across the country, the uncertainty from bulk deposition measurement and application is of limited impact on the national level or the overall spatial pattern of deposition.

Q4. L180: Does "chemical transport mode (CTM) results" means emission inventory? If so, "emission inventory" should be used as shown in Fig. 2. "mode" may be mistake for "model".

Response and main revisions:

We appreciate the reviewer's reminder. "**Chemical transport model (CTM) results**" means surface concentrations of NO_3^-, HNO_3, NH_4^+, and SO_4^{2-} simulated from CTM simulation. Emission inventories were also included in RF. We have accordingly modified **Figure 2 in the revised manuscript**.

Q5. L184: "dry deposition rate" means dry deposition amount. "dry deposition velocity" is correct.

Response and main revisions:

We thank the reviewer's reminder and "dry deposition rate" has been modified as "dry deposition velocity" **in Lines 123 and 185 in the revised manuscript**.

Q6. L215: Regarding particulate NH_4, NO_3, particle size information should be indicated (TPM, PM_{10}, $PM_{2.5}$ etc.). In the case of $PM_{2.5}$, dry deposition of NO_3 in coarse aerosols, which contributes considerable part of total NO_3 dry deposition, is ignored. If so, it should be discussed in section 3.4.

Response and main revisions:

We thank the reviewer's important comment. The dry deposition flux of NH_4^+ and NO_3^- was calculated by multiplying measured concentrations with simulated V_d from the GEOS-Chem. Both the observed concentrations and simulated V_d represented (or approximated) the values for total particles (Please also see our response to Question 2).

Q7. L406-410: Trend analysis (e.g. Mann-Kendall test) is effective to mention statistically that $R_{dry/wet}$ is stable for N, decline for S before 2015. It is also available for the increases after 2015.

Response and revisions:

We thank the reviewer's important comment. Following his/her suggestion, we conducted Mann-Kendall test for the trend of $R_{dry/wet}$ for N and S, and the results are presented **in a newly added Table S5 in the revised supplement**. According to the test, the $R_{dry/wet}$ of N species kept relatively stable for earlier years and then slightly increased since 2015 ($p>0.01$). The $R_{dry/wet}$ of sulfur declined significantly before 2015 ($p<0.01$) and then slightly increased afterwards without statistical significance ($p>0.01$). Therefore, our statement was supported by the test.

Table S5 in the revised supplement: The Mann-Kendall test for the trend of $R_{dry/wet}$ of N and S in 2005-2020. The z-value represents the standard normal statistic, and the p-value represents the generalization. The former indicates the trend, while the latter indicates statistical significance. P1 and P2 indicate 2005–2015 and 2015–2020, respectively.

Species	OXN		RDN		N		S	
Period	P1	P2	P1	P2	P1	P2	P1	P2
z	-2.024	2.254	-2.024	1.879	-2.336	1.879	-3.270	1.127
p	0.043	0.024	0.043	0.060	0.020	0.060	0.001	0.260

Note: Negative and positive z-value indicate a downward and upward trend in the time series, respectively; $p<0.01$ indicates a significance level of 99%.

Q8. L420-423: In Figure S3, the range of temperature variation during the period is within 1 K. I think it is too small to enhance dry deposition by stomatal uptake. Moreover, dry deposition of these N and S component (aerosols and reactive gases) to stomata is small compared to deposition to cuticle.

Response and revisions:

We thank and agree the reviewer's comment. The temperature variation is too small to enhance dry deposition, thus we have deleted the sentence and modified **Figure S4 in the revised supplement (Figure S3 in the original submission)**.

Q9. L440-445: Please check the amount of N deposition in USA and Europe in Table S4. They are too small compared with the global distribution of total N deposition (Fig. 4.8a) by Vet et al (2014) in L883. It shows the range of 1-20 kg N and 2-40 kg N in USA and Europe, respectively. Schwede and Lear (2014) also shows same range of N deposition in USA. (http://dx.doi.org/10.1016/j.atmosenv.2014.04.008)

Response and revisions:

We thank the reviewer's reminder and we are sorry for the error in the calculation in our original submission. We have checked and corrected the results **in Table S6 in the revised supplement (Table S4 in the original submission)**. The corrected results are within the range provided by Vet et al (2014), as shown in Table R1.

Table S6 in the revised supplement: Comparisons of total deposition fluxes of different species between our study in China and two networks in other countries (kg N/S ha^{-1} yr^{-1}).

	Period	RDN	OXN	N	S
USA	1990-2020	2.7	13.3	16.0	16.1
Europe	2000-2019	1.4	4.7	6.1	4.1
China	2005-2020	20.2	15.2	35.4	25.9

Table R1 Comparisons of total N deposition fluxes between our study and Vet et al (2014) (kg N/S ha^{-1} yr^{-1}).

	Reference	N	S
USA	This study	16.0	16.1
	Vet et al (2014)	0.4-20.0	4.0-23.4
Europe	This study	6.1	4.1
	Vet et al (2014)	2.0-28.1	4.0-32.0

Q10. L471-485: The authors should discuss why the short-term emission reduction was not well reflected in the deposition. For example, Yamaga et al. (2021) in L935 mentioned that recent decrease of total S deposition in Japan was associated with recent reduction in SO_2 in China. Therefore, the short-term emission reduction might be reflected in decrease of transboundary air pollution at first, because the reduction started from the east side in China.

Response and revisions:

We thank and agree the reviewer's very important comment. It is really an impressive thought (also raised by Reviewer #2, see his Questions 19 and 25). As reported by Yamaga et al. (2021), the trend of NO_3^- to nss-SO_4^{2-} concentration ratio in precipitation in Japan clearly corresponded to that of the NO_X to SO_2 emission ratio in China, indicating that the OXN and S deposition in Japan might be influenced by the NO_X and SO_2 emissions in China, respectively. Therefore, the short-term emission reduction in China was likely to reduce the transboundary deposition to downwind areas (such as Japan). Such transboundary impact might be sooner than the local one. We have stated **in Lines 523-527 in the revised manuscript**.

Q11. L600-607: The authors are requested to discuss why the larger N deposition was found in summer.

Response and revisions:

We thank the reviewer's reminder and have added the reasons why the larger N deposition was found in summer **in Lines 657-660 in the revised manuscript**. The seasonal trend of N deposition was largely influenced by dry deposition, given its large proportion to the total. As shown **in a newly added Figure S5 in the revised supplement**, the V_d of HNO_3 in summer was 4.4 times in winter, leading to larger OXN deposition in summer. Moreover, warm weather elevated the volatility of NH_3 in croplands, resulting in greater emissions and thereby deposition in summer.

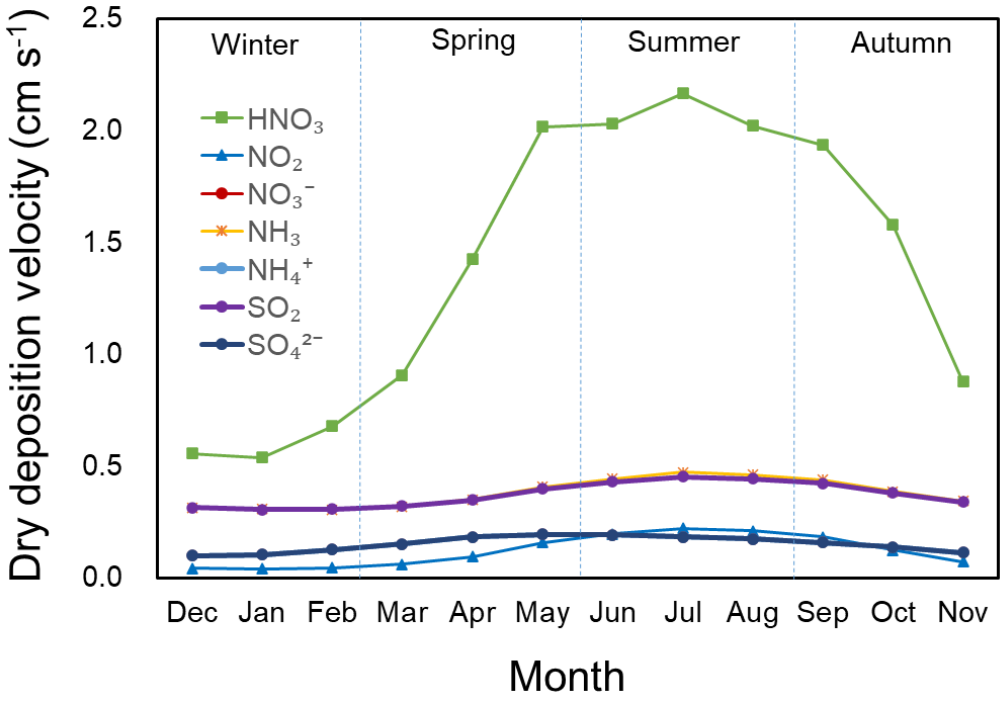

Figure S5 in the revised supplement: The monthly means of the modeled dry deposition velocity of N and S during 2013-2020.

Reference

Flechard, C. R., Nemitz, E., Smith, R. I., Fowler, D., Vermeulen, A. T., Bleeker, A., Erisman, J. W., Simpson, D., Zhang, L., Tang, Y. S., and Sutton, M. A.: Dry deposition of reactive nitrogen to European ecosystems: a comparison of inferential models across the NitroEurope network, Atmos. Chem. Phys., 11, 2703-2728, https://doi.org/10.5194/acp-11-2703-2011, 2011.

Kuang, F., Liu, X., Zhu, B., Shen, J., Pan, Y., Su, M., and Goulding, K.: Wet and dry nitrogen deposition in the central Sichuan Basin of China, Atmos. Environ., 143, 39-50, https://doi.org/10.1016/j.atmosenv.2016.08.032, 2016.

Song, L., Kuang, F., Skiba, U., Zhu, B., Liu, X., Levy, P., Dore, A., and Fowler, D.: Bulk deposition of organic and inorganic nitrogen in southwest China from 2008 to 2013, Environ. Pollut., 227, 157-166, https://doi.org/10.1016/j.envpol.2017.04.031, 2017.

Vet, R., Artz, R. S., Carou, S., Shaw, M., Ro, C.-U., Aas, W., Baker, A., Bowersox, V. C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar, J. J., Gillett, R., Forti, M. C., Gromov, S., Hara, H., Khodzher, T., Mahowald, N. M., Nickovic, S., Rao, P. S. P., and Reid, N. W.: A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmos. Environ., 93, 3-100, https://doi.org/10.1016/j.atmosenv.2013.10.060, 2014.

Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 23, 1293-1304, https://doi.org/https://doi.org/10.1016/0004-6981(89)90153-4, 1989.

Yamaga, S., Ban, S., Xu, M., Sakurai, T., Itahashi, S., and Matsuda, K.: Trends of sulfur and nitrogen deposition from 2003 to 2017 in Japanese remote areas, Environ. Pollut., 289, 117842, https://doi.org/10.1016/j.envpol.2021.117842, 2021.

Zhang, L., Gong, S., Padro, J., and Barrie, L.: A size-segregated particle dry deposition scheme for an atmospheric aerosol module, Atmos. Environ., 35, 549-560, https://doi.org/10.1016/S1352-2310(00)00326-5, 2001.

Comments from Reviewer #2

Q1. It is of great interest and importance to modeling the historical S/N deposition in China, one of the hotspots in the world, for supporting policy-making. Based on various databases and applying machine learning method, the authors estimate the deposition of different species of N and S at relatively high resolution during 2005-2020. A delayed response of N/S deposition to NO_X/SO_2 emission abatement was found in China. In general, the manuscript was well written, and worth publishing after some minor revision. The reason of delay need be further discussion, e.g., the transport of N/S out of land of China and the changes of atmospheric oxidizing capacity.

Response and main revisions:

We appreciate the reviewer's positive comment, and have made point-by-point response and revisions as summarized below. In particular, the delayed response of deposition to emission control is of great interest to research community and more careful studies are needed to answer the question. In this work, we have added discussions on the transport out of China and changing atmospheric oxidizing capacity. Please find our response to Questions 19 and 25, and to Questions 7 and 28, respectively.

Q2. Line 30: The full name of OXN or RDN need be given when first occurs.
Line 34 & 35: Same as above for $R_{dry/wet}$ and $R_{RDN/OXN}$.

Response and main revisions:

We thank the reviewer's reminder and have added the full name of OXN, RDN, $R_{dry/wet}$ and $R_{RDN/OXN}$ **in Lines 30-31, 34, and 36-37 in the revised manuscript.**

Q3. Line 68: to reduce acid rain and later improve …

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as suggested **in Lines 70-72 in the revised manuscript.**

Q4. Line 72: Total emission control of NO_X was carried out in the 12th FYP.

Response and main revisions:

We thank the reviewer's reminder and we have corrected this error as "including the policy of limiting national total emission levels of SO_2 and NO_X within the 11^{th} Five-year Plan (FYP, 2006-2010) and 12^{th} FYP period (2011-2015) respectively" **in Lines 73-75 in the revised manuscript.**

Q5. Line 77: Which years?

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as "……those policies have reduced annual SO_2 and NO_X emissions from 2007 and 2012, respectively." **in Line 79 in the revised manuscript.**

Q6. Line 138: (GAM)

Response and main revisions:

We thank the reviewer's reminder and have modified as "(GAM)" **in Line 140 in the revised manuscript**.

Q7. Line 223: The transformation of SO_2 to sulfate depends on atmospheric oxidizing capacity, thus on NO_X concentration (or emission). The discussion on the uncertainty need added.

Response and main revisions:

We thank the reviewer's important comment and have added the discussion on the uncertainty **in Lines 684-691 in the revised manuscript**. SO_2 can be oxidized to form sulfate in the troposphere, which may occur in the gas phase, clouds or fog

droplets, or on aerosol particles (Lee et al., 2019; Sarwar et al., 2013). Those processes are influenced by the atmospheric oxidizing capacity, and thereby the NO_X concentration (He et al., 2014; Ye et al., 2023). Along with economic development and implementation of air pollution controls, the changing emissions of NO_X as well as some other species (e.g., volatile organic compounds) have altered atmospheric oxidizing capacity within the research period. Using the relationship between 2013 and 2020 to extrapolate the sulfate deposition for 2005-2020 would potentially result in some uncertainty to the results.

Q8. Line 257: What were the modeled years? How was the performance of the model for China.

Response and main revisions:

We are sorry for the ambiguity and thank the reviewer's reminder. The simulation was conducted for 2005-2020 with good model performance. The correlation coefficients between simulated and observed concentrations ranged 0.51–0.82 and the normalized mean biases were within 30%. We have added the model performance and the relevant reference **in Lines 285-288 in the revised manuscript**.

Q9. Line 299: Although there is reference, the brief introduction on the method is needed. What was the performance of the model?

Response and main revisions:

We thank the reviewer's reminder. We have added brief principle and model performance of GAM in the revised manuscript. The GAM model connects the nonlinear relationship between wet deposition and predicted variables (satellite-derived VCDs, meteorological factors and geographic covariates, etc.) to estimate the monthly wet deposition of SO_4^{2-}, NO_3^- and NH_4^+ in China at a horizontal resolution of 0.25°×0.25° (**Lines 351-355 in the revised manuscript**). The predicted wet deposition was basically consistent with the ground-level observed values, and

the correlation coefficient was greater than 0.7, indicating the good performance of GAM (**Lines 355-359 in the revised manuscript**).

Q10. Line 345: Were these studies carried out for the whole country or just at several sites? Since the monitoring sites are concentrated in the more developed east part of China, how did you consider the uncertainty caused by the bias?

Response and main revisions:

We thank the reviewer's comment and have added the research scale information **in the Table S4 in the revised manuscript (Table S3 in the original submission)**. Indeed the studies conducted in less developed west China were much less than those in east. As those studies were not included in the RF or GAM, the uneven distribution of monitoring sites would not directly influence our prediction results, but might bring bias to the comparison between prediction and observation. The bias is expected to be further evaluated and reduced when more observation data get available in the west.

Q11. Line 353: in its high stage?

Response and main revisions:

We thank the reviewer's reminder. We mean the control of SO_2 started from 2005, thus it should be "in its initial stage" **in Line 390 in the revised manuscript**, not in its high stage.

Q12. Line 378: Limited before 2010. The total emission control of NO_X was carried out in the 12th FYP (2011-2015).

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as "China's NO_X emission control was limited till 2010" **in Lines 414-415 in the revised manuscript.**

Q13. Line 379: The total emission control of NO_X in the 12th FYP required the installation of SCR since 2011, although more and more SCR installation had been finished after 2013.

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as "The country required installation of selective catalyst reduction (SCR) systems from 2011, and NAPPCAP drove fast growing penetration of SCR in the power and cement production sectors, resulting in a 28.6% reduction in the annual total emissions of NO_X from 2013 to 2020 (Karplus et al., 2018; Li et al., 2018)" **in Lines 416-419 in the revised manuscript.**

Q14. Line 418: Another reason that increasing NO_X led to high atmospheric oxidizing capacity, and thus promoting sulfate formation for wet deposition?

Response and main revisions:

We thank and agree the reviewer's important comment. The transformation of SO_2 to sulfate depends on atmospheric oxidizing capacity and thereby the NO_X concentration (Please also see our response to Question 7). The growth of NO_X emissions before 2013 led to high atmospheric oxidizing capacity, thereby promoting sulfate formation for wet deposition. We have added the discussion **in Lines 459-461 in the revised manuscript.**

Q15. Line 421: How about the decrease in atmospheric oxidizing capacity?

Response and main revisions:

We thank the reviewer's comment. Although NO_X was declining for most recent years, the atmospheric oxidizing capacity was still growing with enhanced ground-level ozone concentration in most areas of China (Feng et al., 2021; Wang et al., 2023). The reasons include less effective control on the emissions of volatile organic compounds (Ding et al., 2021; Zheng et al., 2018). Therefore we mean

"decrease in atmospheric oxidizing capacity" is not a strong explanation for the growing $R_{dry/wet}$.

Q16. Line 453: Replacing continents by regions? Same for the whole text.

Response and main revisions:

We thank the reviewer's reminder and the "continents" has been modified as "regions" **in Lines 493, 512-513 in the revised manuscript**.

Q17. Line 463: Delete 'in'. Need more recent literature on the trends.

Response and main revisions:

We thank and agree the reviewer's comment. We have deleted "in" and included more recent studies (Constantin et al., 2020; Fowler et al., 2013; Skyllakou et al., 2021; Zhao and Qiao, 2022) **in Lines 502-504 in the revised manuscript**.

Q18. Line 469: 2005 was only for SO_2. NOx had later year.

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as "Following developed countries, gradually tightened measures of reducing SO_2 and NO_X have been launched since 2005 and 2011 respectively, and the deposition began to decline afterwards" **in Lines 508-510 in the revised manuscript**.

Q19. Line 485: What were the reasons for the delay? How about the contribution of natural sources for NOx, or the transboundary transport of S/N?

Response and main revisions:

We thank the reviewer's important comment and have made response and corresponding revisions regarding the two issues.

(1) The transport of deposition to downwind areas could be part of reasons for the delay, and we have added the discussions **in Lines 523-529 in the revised manuscript**. As reported by Yamaga et al. (2021), the trend of NO_3^- to nss-SO_4^{2-} concentration ratio in precipitation in Japan clearly corresponded to that of the NO_X to SO_2 emission ratio in China, indicating that the OXN and S deposition in Japan might be influenced by the NO_X and SO_2 emissions in China, respectively. Therefore, the short-term emission reduction in China was likely to reduce the transboundary deposition to downwind areas (such as Japan). Such transboundary impact might be sooner than the local one.

(2) The soil NO_X emissions from both the natural nitrogen pool and fertilizer input are conventionally considered as natural sources, and have not been included in the current design of emission control strategies in China yet (Zhang et al., 2019). The nitrogen inputs to soil lead to soil NO_X emissions in China reaching 0.4-1.3 Tg N yr^{-1}, about 12% of the anthropogenic NO_X emissions (Lu et al., 2019; Lu et al., 2021). Given the small fraction of natural NO_X to the total, they could not be the main reason for the delay of deposition to the control of anthropogenic NO_X. With continuous control of anthropogenic emissions in the future, however, the variation of emissions from natural sources might play a more important role on the changing deposition and deserves more attentions. (**Lines 529-532 in the revised manuscript**).

Q20. Line 506: What are the countries for example?

Response and main revisions:

We thank and agree the editor's comment and the sentence has been modified as "In many European countries with abundant agricultural activities (such as Netherlands, Germany, Switzerland and France)" **in Lines 554-555 in the revised manuscript**.

Q21. Line 508: although not as strong as for NO_X.

Response and main revisions:

We thank and agree the reviewer's comment and the phrase has been added **in line 557 in the revised manuscript**.

Q22. Line 538: Delete 'the pollution was mainly transported by atmospheric turbulence and'.

Response and main revisions:

We thank the reviewer's reminder and the phrase has been deleted.

Q23. Line 546: Not dominate, although share more.

Response and main revisions:

We thank the reviewer's reminder and the sentence has been modified as "while farming and animal husbandry shared more in the economy in the west, leading to substantial NH_3 emissions" **in Lines 592-594 in the revised manuscript.**

Q24. Line 551: Did the results of satellite-derived VCDs show so sharp reduction of vertical column densities? How about the possibility of overestimation of the emission reduction?

Response and main revisions:

We thank and agree the reviewer's important comment. While MEIC has been recognized as the best estimation of China's air pollutant emissions and widely applied, the uncertainty of China's emission inventory is always a concern in the research community. Satellite-derived VCDs provide some information on the changing emissions, but cannot provide accurate estimates alone as well. We compared MEIC and estimation with a "top-down" methodology based on satellite observation (Qu et al., 2019), and found that MEIC indeed provided a more optimistic

estimation for China's SO_2 emission reduction. We have added the information **in Lines 600-604 in the revised manuscript**.

Q25. Line 555: I guess higher ratio of emission from the east than west was transported out of land and deposit on the sea. What is the effect on the effectiveness?

Response and main revisions:

We thank and agree the reviewer's important comment. As we respond to Question 19 (and Question 10 of Reviewer #1), the changing transboundary deposition to downwind areas from eastern China could be part of reason for the delayed response of local deposition to changing emissions. We have stressed this **in Lines 523-529 in the revised manuscript**. Regarding the difference in changing deposition between eastern and western China, the controls of precursor emissions should still be the dominating driving factor. Please also see our response to Question 19.

Q26. Line 577: The D/E ratio for the whole China need be added in the figure.

Response and main revisions:

We thank and agree the reviewer's important comment. We have added the D/E ratio for the whole China **in Figure 9** and "The nationwide D/E of OXN, RDN, and S were 1.4, 2.4, and 2.3, respectively." **in Lines 629-630 in the revised manuscript**.

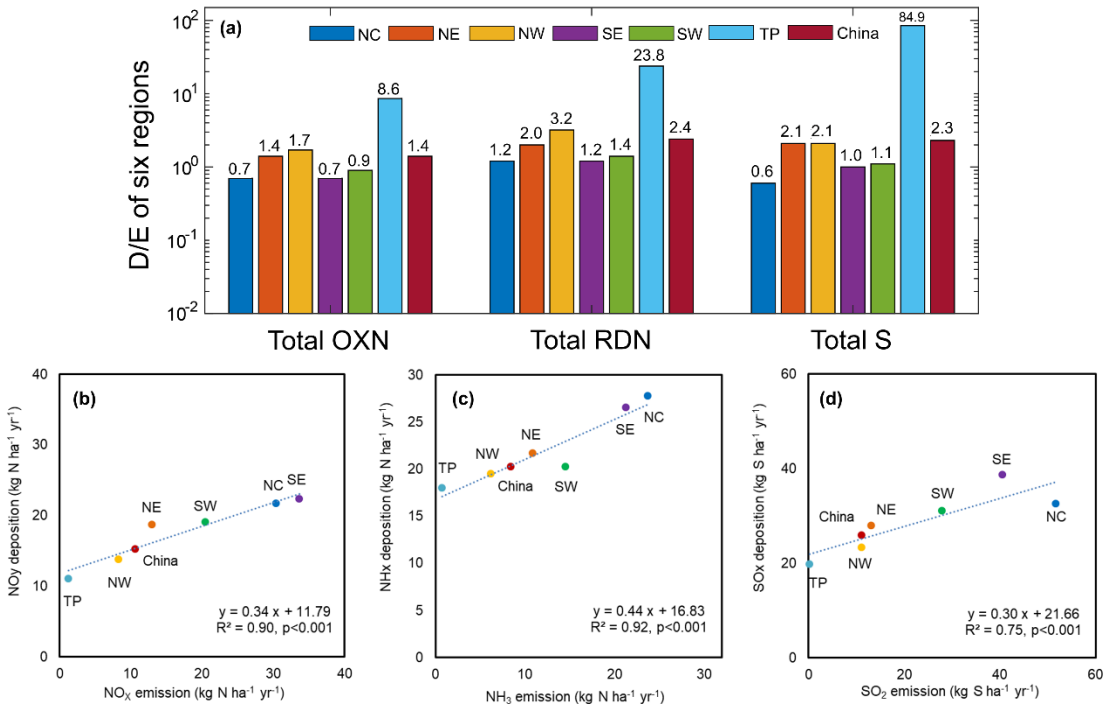

Figure 9 in the revised manuscript: Annual mean D/E ratio of OXN, RDN and sulfur from 2005 to 2020 in different regions (a) and linear relationship between regional deposition and emissions (b-d).

Q27. Line 592: Emphasis need be taken to the region under the line, which might have transboundary deposition.

Response and main revisions:

We thank the reviewer's important comment and understand his concern. Figure 9b-d shows the linear relationship between multiple-year average deposition and emissions for different regions during 2005-2020. Indeed, the regions under the line are more inclined to transmit pollution outward compared with those above the line. However, as the figure focus on the correlation for different regions, it is not an ideal way to measure the source-sink relationship for a certain region. The more direct indicator is D/E, as shown in Figure 9a. The relevant discussions have been given **in Lines 628-640 in the revised manuscript**.

Q28. Line 633: The effects of atmospheric oxidizing capacity on sulfate formation was not considered.

Response and main revisions:

We thank the reviewer's comment and have added the discussion on the uncertainty of the effects of atmospheric oxidizing capacity on sulfate formation **in Lines 684-691 in the revised manuscript**: "The conversion of SO_2 to sulfate is influenced by the atmospheric oxidizing capacity, and thereby the NO_X concentration (He et al., 2014; Ye et al., 2023). Along with economic development and implementation of air pollution controls, the changing emissions of NO_X as well as some other species (e.g., volatile organic compounds) have altered atmospheric oxidizing capacity within the research period. Using the relationship between 2013 and 2020 to extrapolate the sulfate deposition for 2005-2020 would potentially bring some uncertainty." Please also see our response to Question 7.

Q29. Line 993: The full name of CNEMC and NNDMN need be added.

Response and main revisions:

We thank the reviewer's reminder and have added the full name of CNEMC and NNDMN **in Lines 1124-1126 in the revised manuscript**.

Q30. Line 1018: How about the data sources of deposition in USA and EU?

Response and main revisions:

We thank the reviewer's reminder and have added the data sources of deposition in USA and EU **in Lines 1150-1158 in the revised manuscript**.

Q31. Line 1044: Same as above.

Response and main revisions:

We thank the reviewer's comment. The original text specified the source of the data **in Lines 1179-1183 in the revised manuscript**.

Q32. Line 1066: The ratio of N and S deposition need be added in Figure e?

Response and main revisions:

We thank and agree the reviewer's important comment. We have added the ratio of N to S deposition ($R_{N/S}$) **in Figure 5e** and the sentence "Growing $R_{N/S}$ was found for most time within the research period, as China started SO_2 emission control earlier than NO_X and NH_3" **in Lines 465-466 in the revised manuscript**.

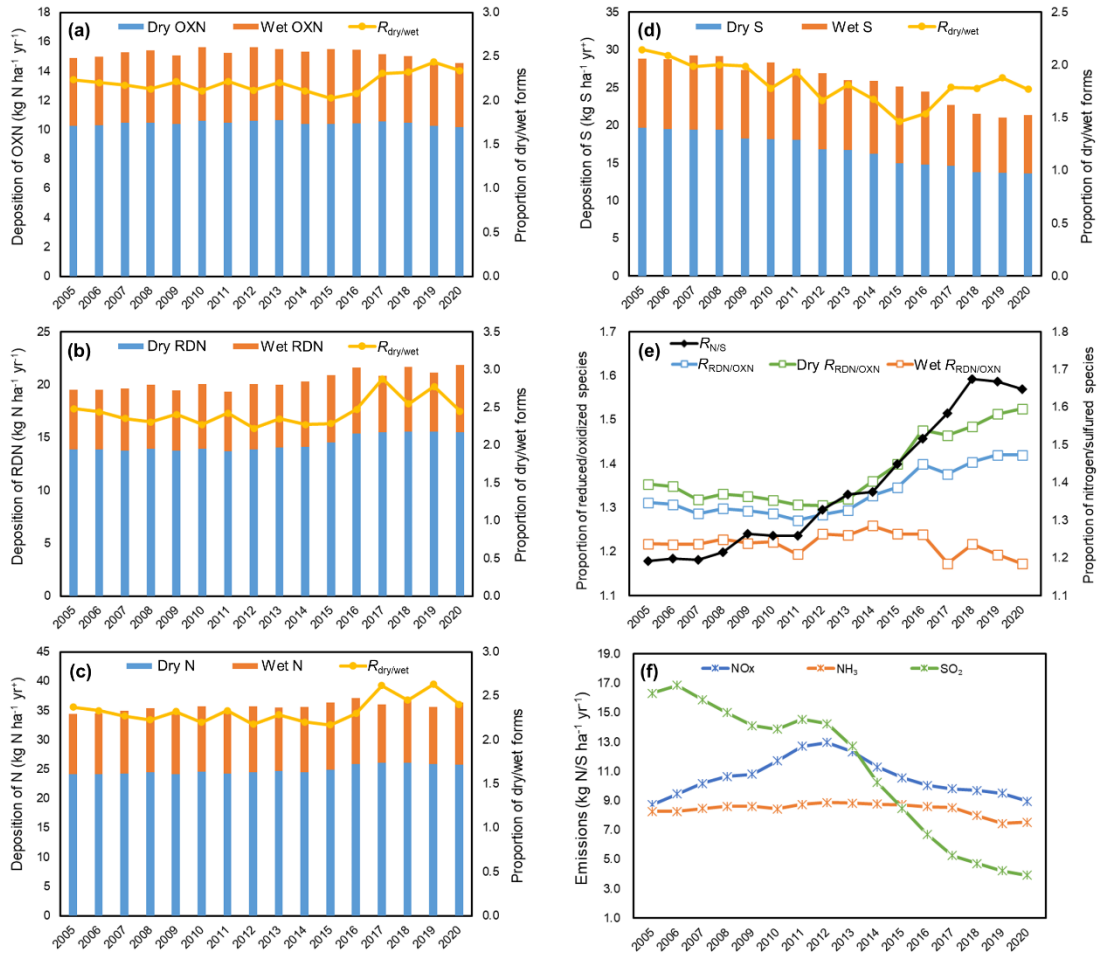

Figure 5 in the revised manuscript: The interannual variability of N and S deposition, emissions and component proportion in China from 2005 to 2020. The emission data over China were taken from MEIC.

Q33. Line 1072: I suggest to use the same scale (in the legend) for dry, wet and total deposition.

Response and main revisions:

We thank the reviewer's reminder and have improved the figure by using the same scale in the legend for three forms of deposition **in Figure 7 in the revised manuscript**.

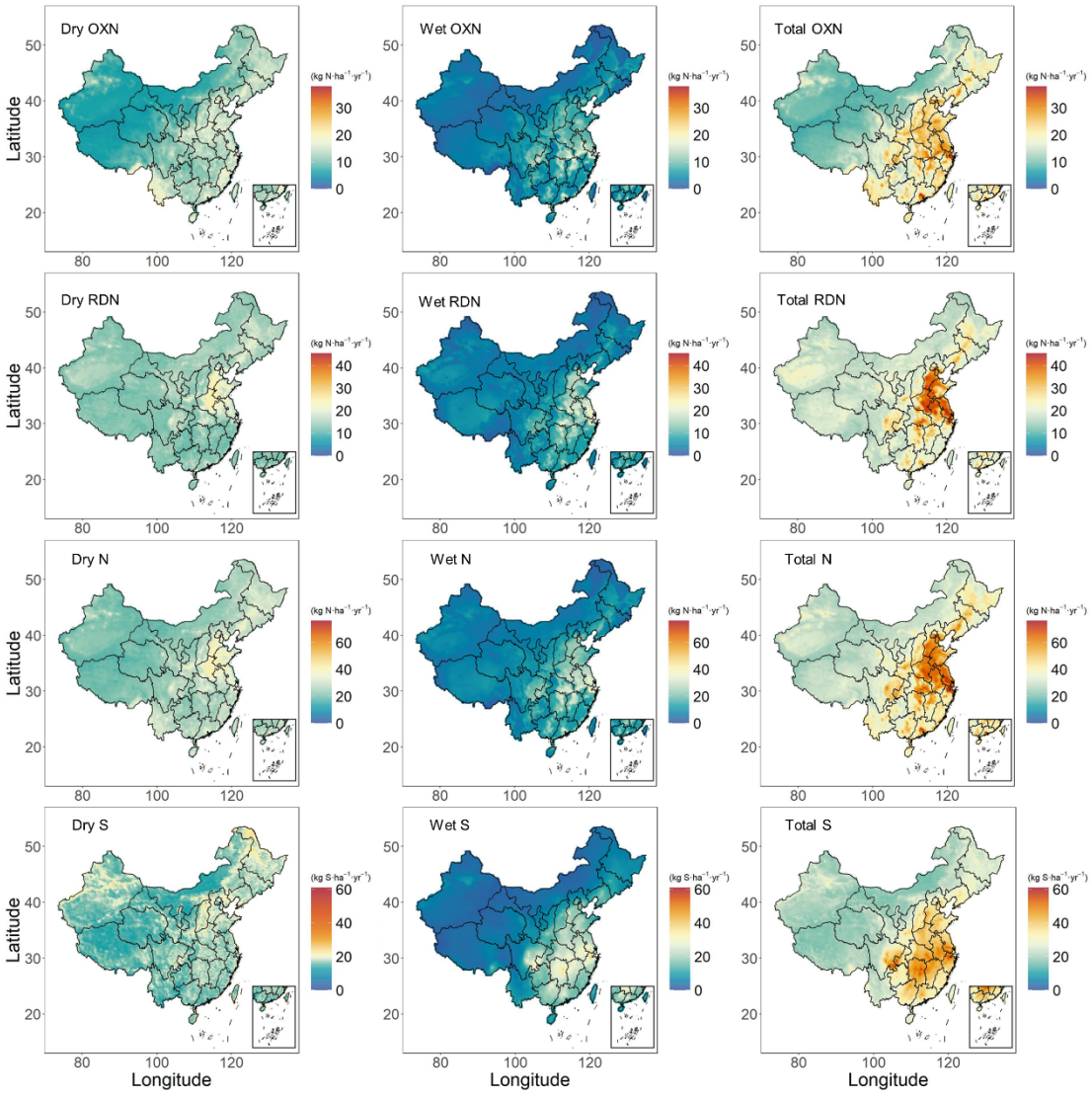

Figure 7 in the revised manuscript: The spatial distributions of N and S deposition flux in 2005-2020.

Reference

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Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18, 14095-14111, https://doi.org/10.5194/acp-18-14095-2018, 2018.

Comments from Reviewer #3

Q1. This study estimated nitrogen and sulfur deposition across China during 2005-2020 based on multiple statistical models and satellite observations. Their estimates were separated by the dry and wet contributions, based on satellite observations. The authors also present a long-term record of speciated nitrogen deposition. The authors motivate their study by (1) indicating the need for long-term measurements of dry deposition (2) calling for more spatially-resolved estimates of nitrogen and sulfur deposition (beyond what surface networks can provide) and (3) the large uncertainty of previous estimates for China.

This study is not suitable for publication because several aspects of their approach appear poorly justified. It claims to do far more than it does. This starts from the title/abstract: the authors use three satellite observations (NO_2/SO_2 column concentrations and NH_3 column concentrations) from which they extract both dry and wet deposition of 7 species (NO_3^-, HNO_3, NO_2, NH_4^+, NH_3, SO_2, and SO_4^{2-}). These transformations rely heavily on a CTM model (GEOS-Chem) and surface observations in a procedure that is not described sufficiently in the main text or Methods, and even not well described in the Supporting Information.

Response and main revisions:

We appreciate the reviewer's valuable and important comments, and respectfully understand his/her concerns. We read all the comments carefully, and feel that part of our research framework and methodology for dry deposition estimation might be misunderstood by the reviewer. We are sorry for the unclear or insufficient statements in the original submission, and have improved the manuscript by reorganizing the random forest (RF) model description, adding essential information on observation and CTM, and expanding the uncertainty analysis. The details can be found in our point-by-point responses and corresponding revisions below. In particular, we would like to briefly describe the RF we used and hopefully it reads clearer to the audience.

As described in Section 2.2 of the revised manuscript, we applied RF model to obtain the long-term (2005-2020) spatiotemporal patterns of dry S and N deposition in China (the estimation of wet deposition is provided in Section 2.3). Given very few direct observation data available, the response variable of RF, i.e., dry deposition flux (F_d), was calculated using a "inferential" method, which has been widely applied in international and domestic monitoring networks (e.g., CASTNET and EANET) and studies (Wen et al., 2020; Xu et al., 2019; Yu et al., 2019; Pan et al., 2012). F_d was calculated as the product of surface concentration (C) and deposition velocity (V_d) for given species:

$$F_d = C \times V_d \quad (1)$$

As we described in Section 2.2.2, surface concentrations (C in equation (1)) were obtained from high-quality monitoring networks for SO_2, NO_2, HNO_3, NO_3^-, NH_3, and NH_4^+, while those of SO_4^{2-} were estimated based on the observed SO_2 and the model-derived correlation between SO_2 and SO_4^{2-}. Dry deposition velocities (V_d in equation (1)) were simulated (not "assumed" as indicated by the reviewer in Question 9) with a state-of-art chemistry transport model (GEOS-Chem).

We then selected interpretation variables for RF, including satellite-derived tropospheric VCDs, meteorological factors, geographic covariates, surface concentrations from CTM, and emission data (**Lines 188-191 in the revised manuscript**). RF captured the nonlinear relationship between response variable (F_d calculated in equation (1)) and those interpretation variables, through the following equation (the newly added equation (2) in the revised manuscript):

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

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

Regarding the reviewer's concern of application of three VCDs to estimate seven species, therefore, it can be seen that the VCDs were not directly applied in

calculating the response variable (F_d), but included in the RF model as an interpretation variable. Such application did not weaken the robustness of dry deposition estimation. Indeed there is lack of direct satellite-derived HNO_3, NO_3^-, SO_4^{2-}, and NH_4^+ information, and we had to use the columns of their precursors, i.e., NO_2, SO_2, and NH_3 as interpretation variable in RF. The uncertainty has been demonstrated to be limited. Please see the detailed evidences in our response to Question 2 of the reviewer.

For observation of surface concentration (C in equation (1)) and simulation of deposition velocity (V_d in equation (1)), we have followed the reviewer's comments and added more technical information in Sections 2.2.2 and 2.2.4, respectively. For example, the measurement procedure has been given in **Lines 226-230 in the revised manuscript**. The principles and calculation procedures of V_d has been provided **in the Text Section of revised supplement**. The uncertainty analysis of the research framework has also been expanded in Section 3.4, and we kindly refer the reviewer to our response to Question 4.

Q2. This approach seems to contradict the motivations for the study: their estimates have large uncertainties in estimating dry HNO_3, NO_3^-, SO_4 due to the lack of the satellite data of HNO_3, NO_3^-, SO_4; wet deposition in fact was the bulk deposition estimated. Dry deposition accounted for around 20% of the bulk deposition based on observation at three rural stations on the North China Plain, and this contribution could reach 39% in urban areas. Thus, the total nitrogen and sulfur could be largely overestimated.

Response and main revisions:

We thank the reviewer's important comment and understand his/her concern.

For estimation of dry deposition of HNO_3, NO_3^-, and SO_4^{2-} (as well as NH_4^+), as we stated in the response to Question 1 of the reviewer, the VCDs from satellite

observation were not directly applied in calculating the response variable (dry deposition flux F_d), but included in the RF model as an interpretation variable. Therefore the application of satellite observation data did not weaken the robustness of dry deposition estimation. For RF estimation, indeed there is lack of direct satellite-derived HNO_3, NO_3^-, and SO_4^{2-} (as well as NH_4^+) information, thus we had to rely on the columns of their precursors, i.e., NO_2, SO_2, as well as NH_3. The bias from such application was expected to be limited, supported by the following two evidences. First, as can be seen in Figure R1, satellite-derived VCDs of the precursors were identified as the most important factors for F_d estimation except for SO_4^{2-}. Therefore, application of precursor VCDs did not only follow the natural logic but was also of statistical significance. Second, as shown in Figures S2 and S3 in the revised supplement (Figures S1 and S2 in the original submission), our RF model successfully captured the non-linear relationship between deposition and interpretation variables, with the annual means of R^2 larger than 0.7 for all species. The good performance of RF thus suggested satisfying estimation of dry deposition across the country.

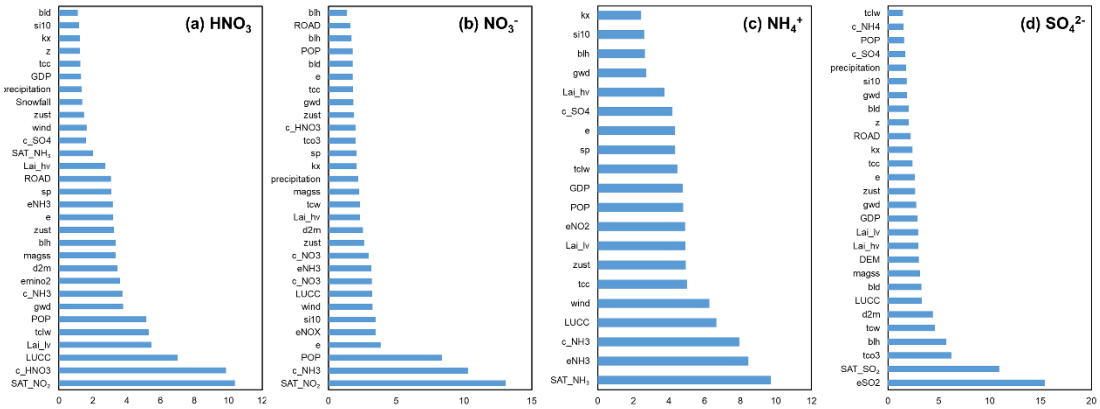

Figure R1. The relative importance of interpretation variables (RIV) to the RF predictions. See Table S2 for the meanings of the abbreviations in the figure.

We have also expanded the discussion on the uncertainty from bulk deposition monitoring and application in **Lines 694-705 in the revised manuscript**. The term 'total deposition' of N is commonly defined as the sum of dry and bulk deposition (Xu et al., 2015; Xu et al., 2019; Zhang et al., 2021), although it is in principle the

sum of dry and wet deposition. The major reason is the big technical and labor cost to quantify the part of dry deposition from the bulk, for such a great number of observation sites across the country. Yu et al. (2019) converted bulk N deposition into wet N deposition using a uniform coefficient of 0.70, but the bias varied by region and was hard to be quantified at the national level. The uncertainty was greater in areas with a higher proportion of dry to total deposition (such as NW and NE areas with less precipitation), and smaller in areas with a lower proportion (such as SE and SW with more precipitation). For example, we compared the results from Kuang et al. (2016) and Song et al. (2017), and found that the difference between 5-year in situ measurements of bulk and wet dissolved inorganic nitrogen (DIN) deposition was only about 2.5 kg N ha^{-1} yr^{-1} at a rural site in SW, equaling to 12% of annual bulk deposition. As SE is the most developed region in China, with relatively high emissions and deposition across the country, the uncertainty from bulk deposition measurement and application is likely of limited impact on the national level or the overall spatial pattern of deposition. It should also be noted that this uncertainty does not apply for S, as wet deposition from EANET was applied in this work.

Q3. I cannot see any improvements compared to previous studies regarding our understanding of China's atmospheric deposition. One acceptable approach is to use the data assimilation methods rather than the rough approach conducted here. The authors can use the satellite NO_2/SO_2 column concentrations and NH_3 column concentrations to better constrain the estimates of CTM modelling.

Response and main revisions:

We thank the reviewer's comment and understand his/her concern.

Regarding the improvements, we combined a newly developed random forest (RF) model and a generalized additive model (GAM), and obtained the long-term (2005-2020) spatiotemporal patterns of dry and wet deposition of multiple N and S species (NO_3^-, HNO_3, NO_2, NH_4^+, NH_3, SO_2, and SO_4^{2-}) for China, at comparatively

high horizontal ($0.25^{o}\times0.25^{o}$) and temporal resolutions (monthly). Due to lack of direct observation and large uncertainty of deposition modeling, in particular, there were very few dry deposition datasets with satisfying completeness in space, time and species. Based on available observation data and modeling techniques, we developed a RF model that well captured the complex nonlinear relationships between dry deposition and multiple variables including satellite-derived VCDs, surface concentrations from CTM, emissions, and meteorological and geographical factors. As shown **in Figures S2 and S3 in the revised supplement (Figures S1 and S2 in the original submission)**, satisfying model performance has been achieved, with the R^2 between observation-based and RF-predicted deposition above 0.7 for all species (except for NH_4^+).

Those methodological and data advances broaden the scientific understanding of China's long-term changes in deposition of N and S species, as well as the influences of human activities and emission controls. Previous studies have rarely linked the changing spatiotemporal pattern of deposition to the regulations of national air pollution prevention and controls. By combining the emission and deposition data, we found a prominent lagging response of deposition to emission abatement in China. Regarding the regional difference, a downward gradient from east to west was detected for dry deposition of OXN while from north to south for S, as a joint effect of anthropogenic activities (which influenced emissions) and meteorological factors. The deposition of OXN and S declined faster in eastern China than that in the west after 2012, indicating the effectiveness of extremely strict emission control in developed east. Compared with Europe and the USA, moreover, China had the smallest benefit of precursor emission reduction on deposition, and relatively weak management of agricultural emissions has resulted in a high proportion of RDN deposition to total N. We elaborated those findings mainly in Section 3 (Results and discussions) and summarized in the Section 4 (Conclusions).

Currently very few assimilation studies were conducted for deposition estimation. Some applied CTMs to estimate the ratio of surface concentration to total troposphere

columns, and then multiplied the ratio by satellite columns to estimate satellite-derived surface concentration (Van der Graaf et al., 2018; Nowlan et al., 2014; Zhang et al., 2017; Liu et al., 2017). We did not use the data assimilation method based on satellite columns because current assimilation techniques could not predict deposition very efficiently and has still to include the V_d from CTMs. Machine learning models offer new options for researchers, as they could fast capture the complex relationships and generally show better predictive accuracy than CTMs and traditional statistical models (Li et al., 2022; Wei et al., 2021; Zhan et al., 2018).

Q4. The study fails to present any uncertainty analysis. Given the error on the satellite retrievals, and the numerous transformations required to estimate the deposition, these uncertainties are likely extremely large and thus a discussion of uncertainties should be central to this analysis.

Response and main revisions:

We thank the reviewer's important comment. We have expanded the uncertainty analysis **in Section 3.4 in the revised manuscript**. Four major uncertainties of this work have been discussed, including (1) the lack of direct evaluation of simulated V_d, (2) uneven distribution of observation sites for surface concentration monitoring, (3) the simulation of conversion of SO_2 to sulfate (please also see our responses to Questions 7 and 28 of Reviewer #2), and (4) application of bulk deposition measurement as wet one (please also see our response to Question 2 of the reviewer).

In particular, we respond to the reviewer's comment on satellite retrievals and "transformations required to estimate the deposition". First, as we explained in responses to Questions 1 and 2 of the reviewer, satellite-derived VCDs were not directly used to calculate the dry deposition flux but included as an interpretation variable in the RF model. The uncertainty from satellite-retrieval was insignificant to the dry deposition prediction, as reconfirmed by the satisfying model performance (R^2

larger than 0.7 for all the species except NH_4^+, as shown **in Figures S2 and S3 in the revised supplement (Figures S1 and S2 in the original submission)**.

Second, as we explained in responses to Question 1 of the reviewer, the inferential method (F_d= C×V_d) is not a "transformation" but a widely accepted method to calculate the dry deposition. The concentrations (C) were obtained from high-quality monitoring networks (see the added monitoring procedures **in Lines 226-230 in the revised manuscript**), while V_d is simulated with the state-of-art GEOS-Chem model (see its principle and calculation procedures **in the Text Section in the revised supplement**). As there are very few direct observations of V_d, we have compared V_d used in this work with other simulation results, and provided the information in **Lines 670-674 in the revised manuscript**. Please also see our response to Question 9 of the reviewer.

Q5. In terms of novelty, multiple studies have reported similar results using remote sensing of SO_2, NO_2 and NH_3 to estimate deposition:

Yu, G., Jia, Y., He, N., Zhu, J., Chen, Z., Wang, Q., Piao, S., Liu, X., He, H., Guo, X., 940 Wen, Z., Li, P., Ding, G., and Goulding, K.: Stabilization of atmospheric nitrogen deposition in China over the past decade, Nature Geosci., 12, 424-431, https://doi.org/10.1038/s41561-019-0352-4, 2019

Zhao, Y., Xi, M., Zhang, Q., Dong, Z., Ma, M., Zhou, K., Xu, W., Xing, J., Zheng, B., Wen, Z., Liu, X., Nielsen, C. P., Liu, Y., Pan, Y., and Zhang, L.: Decline in bulk deposition of air pollutants in China lags behind reductions in emissions, Nature Geosci., 15, 190-195, https://doi.org/10.1038/s41561-022-00899-1, 2022.

Response and main revisions:

We thank the reviewer's important comment. Compared with Zhao et al. (2022) that stressed bulk deposition, this study developed a random forest (RF) model to estimate the long-term spatiotemporal patterns of dry deposition, based on available

observation of surface concentrations, satellite-derived VCDs, emissions, and meteorological and geographical factors. Eventually we obtained a full dataset of China's dry and wet deposition of seven N and S species from 2005 to 2020, with comparatively high horizontal ($0.25^{o}\times0.25^{o}$) and temporal resolutions (monthly). Comparing with Yu et al. (2019), our RF model captured the complex nonlinear relationships between deposition and multiple interpretation variables, instead of applying a linear model. Good model performance has been achieved, with the annual means of R^2 between observation-based and RF-predicted deposition above 0.7 for all species (except for NH_4^+), as shown **in Figures S2 and S3 in the revised supplement (Figures S1 and S2 in the original submission)**. Together, these methodological and data advances broaden the scientific understanding of long-term changes in N and S species deposition in China, in the absence of sufficient direct observation of dry deposition. Please also see our response to Question 3 of the reviewer.

Q6. Lines 218-216: This overview of methodology is completely unclear; more information is needed here and in Methods on how f(so2, so4) are used (over what time horizon) and how they are analysed.

Response and main revisions:

We thank the reviewer's important comment and have added the detailed description of the estimation on the SO_4^{2-} concentrations **in Lines 234-245 in the revised manuscript**.

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-}:

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

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_4^{2-} and SO_2 concentrations simulated by CTM for each year of 2013-2020, respectively; and *f* is the relationship between SO_4^{2-} and SO_2 obtained from GAM. As shown in a newly added Figure S1 in the revised supplement, significant positive correlations were found for SO_4^{2-} and SO_2 concentrations, with the total correlation coefficient (R) estimated at 0.86 ($p<0.001$) for 2013-2020.

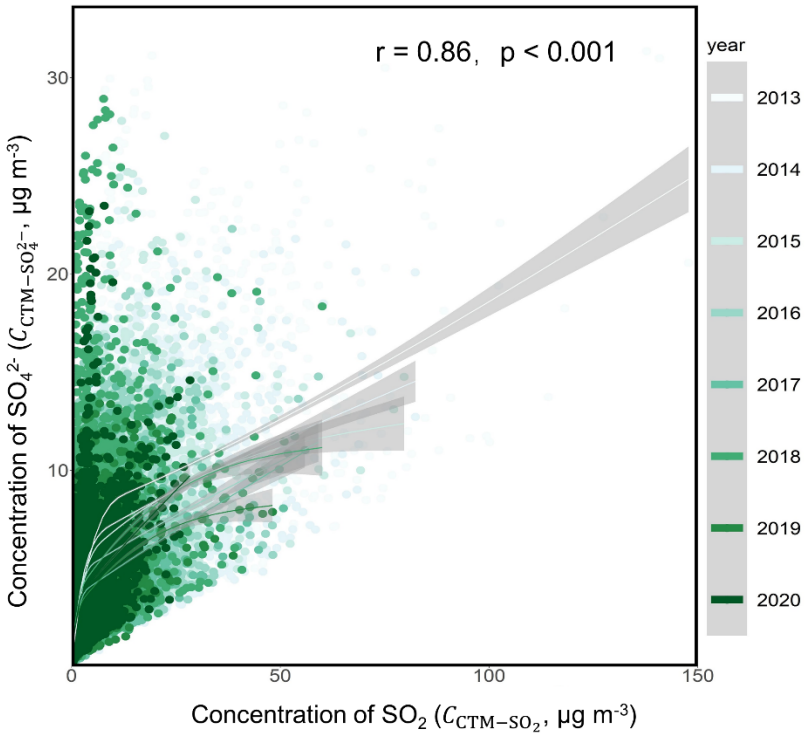

Figure S1 in the revised Supplement: Correlations between simulated SO_4^{2-} and SO_2 concentrations from GAM.

Q7. Line 324-368: Unclear why the deposition/concentrations numbers are not compared to GEOS-Chem with no further context (it would also be useful to compare with the GEOS-Chem values since that model is used to translate columns to surface concentrations). If comparison with a model is featured, the authors should evaluate the model precipitation and concentrations to establish

whether the model is biased in terms of concentrations or the parameterization of the deposition process (assumed dep velocity, precipitation, etc.).

Response and main revisions:

We thank the reviewer's important comment and understand his/her concern on model evaluation. As we respond to Question 1 and 2 of the reviewer, GEOS-Chem was not applied directly to simulate dry deposition or "to translate columns to surface concentrations", thus the RF-predicted deposition should not be compared with GEOS-Chem simulation but observation-based deposition. Note the latter was calculated based on V_d simulated from GEOS-Chem.

GEOS-Chem in this work supported RF model, by (1) simulating V_d for calculation of F_d (the response variable in RF, see equation (1)), and (2) simulating the ground-level concentrations of secondary-formation species (HNO_3, NO_3^-, SO_4^{2-}, and NH_4^+) that were included in RF as an interpretation variable (see the term "CTM" in equation (2)). For (1), as we respond to Question 4 of the reviewer, there are very few direct observations of V_d. We have compared V_d used in this work with other simulation results, and provided the information in **Lines 670-674 in the revised manuscript**. Please also see our response to Question 9 of the reviewer. For (2), good model performance was found for those species, with the correlation coefficients between simulated and measured concentrations ranging 0.51-0.82. We have added the information **in Lines 285-288 in the revised manuscript**. Such CTM bias did not bring big uncertainty to RF prediction, which performed well with the multi-year average R^2 greater than 0.7 for most species.

Q8. Figure 2: this is not a particularly useful figure; a more specific graphic showing how the authors went from column concentrations of 3 species to estimate the 7 species of speciated wet & dry deposition would be more helpful to orient the reader.

Response and main revisions:

We thank the reviewer's comment and have modified **the Figure 2 in the revised manuscript**. The boxes on the left side represent the procedures of dry deposition estimation with the RF model, and those on the right represent the procedures of wet deposition with the GAM. As we respond to Questions 1 and 2 of the reviewer, satellite-derived VCDs were included as interpretation variable in RF for predicting each of the seven species, and the selection of variables in the RF were determined through the Recursive Feature Elimination (RFE) technique (**Lines 193-196 in the revised manuscript**).

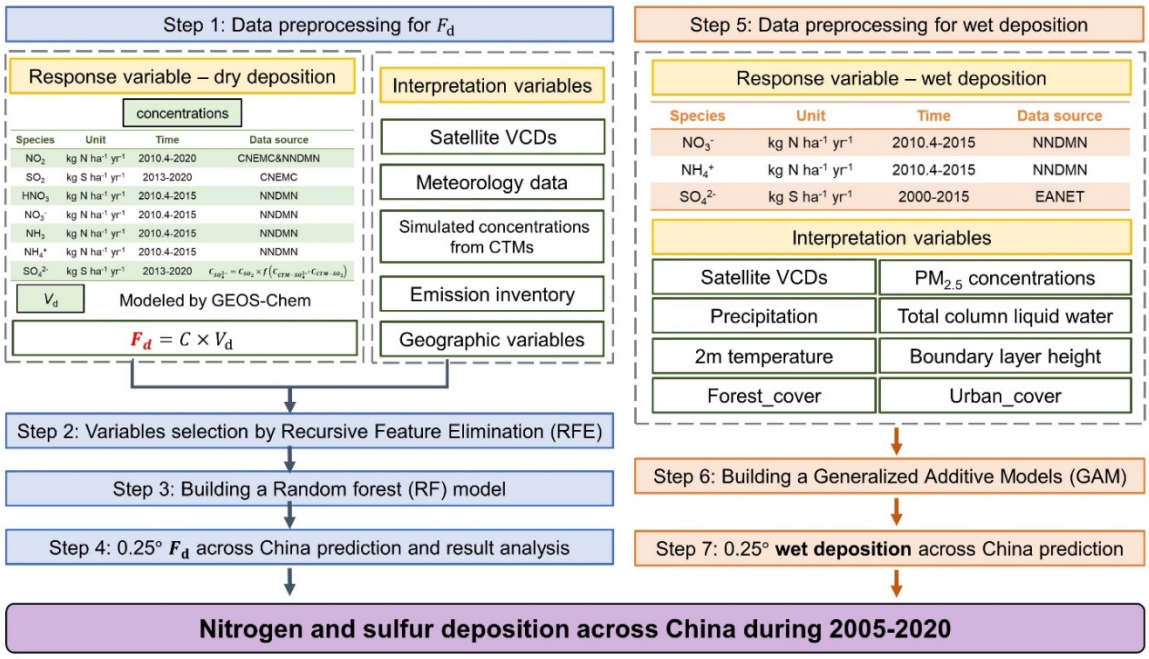

Figure 2 in the revised manuscript: Methodology framework to estimate dry and wet deposition of this study. The blue process shows the four steps to establish the RF model. The orange process shows the three steps in establishing a GAM model. See Sections 2.2 to 2.3 of the method section in the text for the acquisition of the preliminary data set.

Q9. Lines 249: how do the deposition velocities used here compare to those assumed in GEOS-Chem or WRF? Why did the authors choose to use another model (WRF) here rather than GEOS-Chem (which they used to translate from column to surface concentrations/deposition)?

Response and main revisions:

We thank the reviewer's comment. We have only applied GEOS-Chem to simulate (not assume) V_d based on a standard big-leaf resistance-in-series parameterization (**Lines 278-281 in the revised manuscript; Details in Text Section in the revised supplement**). We did not use WRF (could "RF" be misread as "WRF"?)

Currently there are very few direct observations of V_d, and we have compared V_d used in this work with other simulation results, as shown in a newly added Table S9 in the revised supplement. The values from various CTMs are commonly of the same orders for most cases, while big differences exist in some species/land use types, e.g., NO_2 for coastal regions. The bias could be better quantified when more direct observations of V_d get available. We have added the discussion **in Lines 670-674 of Section 3.4 in the revised manuscript**.

Table S9 in the revised supplement: Comparison of the annual V_d of nitrogen compounds by land use type in this and other studies (cm s^{-1}).

Land use type	Deposition velocity (cm s^{-1})						References
	NO_2	HNO_3	NO_3^-	NH_3	NH_4^+	SO_2	
Farmland	0.17	1.45	0.15	0.43	0.15	0.44	This study
	0.18	1.52	0.19	0.40	0.19		Xu et al. (2015)
	0.10	0.76	0.25	0.18		0.25	Zhang et al. (2004)
						0.56	Zhang et al. (2003)
Urban	0.14	1.37	0.15	0.44	0.15	0.44	This study
	0.06			0.78			Pan et al. (2012)
	0.03					0.20	Su et al. (2012)
						0.55	Zhang et al. (2003)
	0.07	1.77	0.44	0.28	0.44		Li et al. (2013)
	0.30	1.10	0.24	0.50	0.24		Luo et al. (2013)
Coastal	0.16	1.56	0.10	0.65	0.13	0.66	This study
	0.01	0.63		0.63			Zhang et al. (2010)
	0.01	0.84	0.27	0.55	0.27	0.63	Zhang et al. (2004)
						0.40	Su et al. (2012)
Forest	0.19	2.23	0.16	0.41	0.16	0.46	This study
	0.10	2.45	0.30	0.20	0.30		Zhang et al. (2004)
	0.19	2.23	0.16	0.41	0.16		Xu et al. (2015)
	0.04					0.16	Su et al. (2012)
Grassland	0.15	1.09	0.19	0.38	0.19	0.33	This study
	0.13	1.16	0.28	0.23	0.28	0.37	Zhang et al. (2004)
	0.15						Xu et al. (2015)
						0.49	Zhang et al. (2003)

Note: Zhang et al. (2004), Su et al. (2012), Xu et al. (2015), Zhang et al. (2010) and Zhang et al. (2003) applied RegADMS, NAQPMS, GEOS-Chem, MM5/CMAQ and AURAMS, respectively. In particular, Zhang et al (2003) focused on the global land use and did not provide specific discussion for China, and was thus excluded when calculating the mean of China.

Q10. Lines 370-513: how do estimated trends compare to previous analyses of satellite observations and ground observations in China?

Response and main revisions:

We thank the reviewer's important comment. The long-term trend of deposition in China is only available for bulk deposition based on 238 observation studies, as investigated and summarized in Wen et al. (2020). We have plotted our estimates with those studies and satellite-derived VCDs in Figure R2. As can be found in Figure R2a-c, our estimation of wet N deposition trend (yellow) was basically consistent with the long-term observation (orange), although generally smaller than the arithmetic mean of observation. Note that observation was more conducted in the developed east with relatively high emission and deposition level, the national average might be overestimated by the arithmetic mean. Similar trends were also found between deposition (this work) and satellite observation, while the latter shows stronger interannual variability particularly for NO_2. In general the long-term trends in both deposition and satellite-derived VCDs reflected the changing emissions resulting from national air pollution control polices.

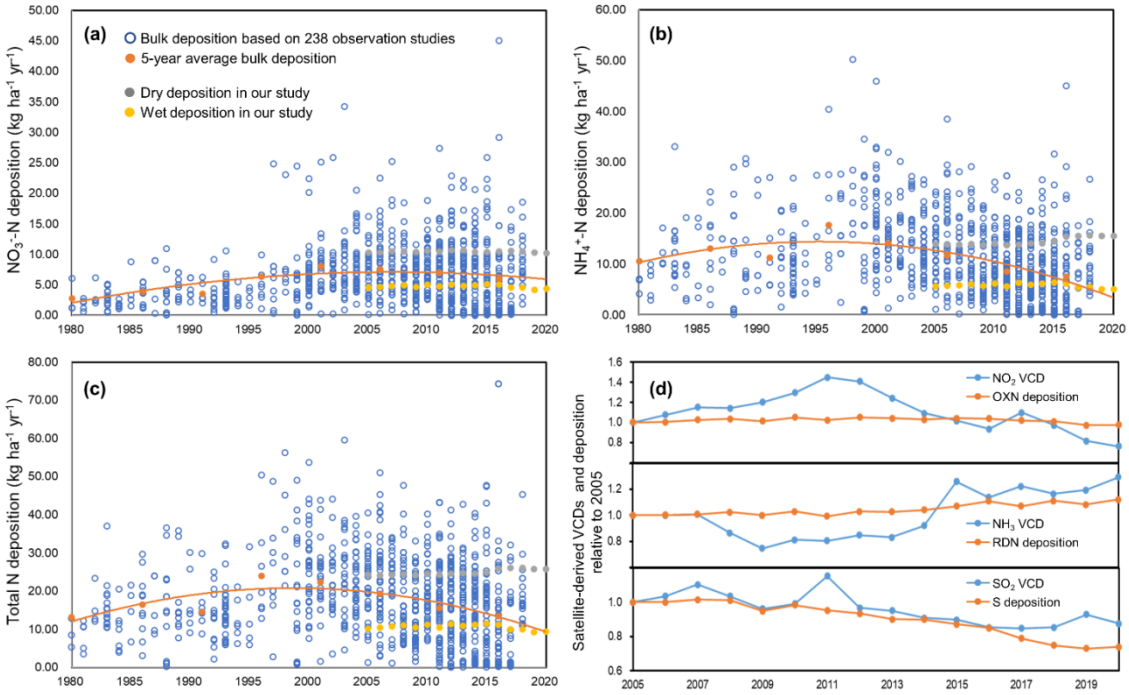

Figure R2 (a-c) Annual mean wet and dry deposition of the N species derived from two complementary databases: (1) published data of bulk N deposition covering 1980–2018 and (2) wet and dry deposition based on the machine learning methods for 2005-2020 in this work. For (1), the blue open circles represent annual average bulk N deposition. The orange curve shows the trends

in inorganic N bulk deposition, while the orange dots represent the 5-year average bulk deposition. For (2), the grey dots represent the dry deposition and the yellow dots represent the bulk deposition. (d) The interannual variations of satellite-derived VCDs and total deposition for China in 2005-2020. All the data are relative to the 2005 levels.

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