

SUPPLEMENT FOR THE RESPONSE TO REVIEWER #2

Number of tables:1 Number of figures: 3

Table list

Table S4 in the revised supplement (Table S3 in the original submission): Comparison of the annual F_d of N and S in this and other studies ($\text{kg N/S ha}^{-1} \text{ yr}^{-1}$).

Figure list

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.

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

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).

Table S4 in the revised supplement (Table S3 in the original submission): Comparison of the annual F_d of N and S in this and other studies (kg N/S ha⁻¹ yr⁻¹).

| Reference | Study region | Research scale | Study period | Dry deposition | | | | | | Wet/bulk deposition | | | Total deposition | | |
|------------------------|-----------------|------------------|--------------|-----------------|------------------|------------------------------|-----------------|------------------------------|-----------------|-------------------------------|------------------------------|------------------------------|-------------------------------|------|------|
| | | | | NO ₂ | HNO ₃ | NO ₃ ⁻ | NH ₃ | NH ₄ ⁺ | SO ₂ | SO ₄ ²⁻ | NH ₄ ⁺ | NO ₃ ⁻ | SO ₄ ²⁻ | N | S |
| This study | China | Grid level | 2005-2020 | 3.4 | 5.3 | 1.7 | 10.3 | 4.2 | 15.5 | 1.2 | 3.3 | 4.6 | 6.4 | 32.9 | 23.1 |
| Nowlan et al. (2014) | China | Grid level | 2005-2007 | 0.2 | | | | | | | | | | | |
| Lye and Tian (2007) | China | Grid level | 2003 | 2.9 | | | | | | | 7.1 | 2.8 | | | 12.9 |
| Jia et al. (2014) | China | Grid level | 1980-2010 | | | | | | | | | | | | |
| Jia et al. (2016) | China | Grid level | 2005-2014 | 0.6 | 1.1 | 0.1 | 5.4 | 0.3 | | | | 13.9 | | | |
| Tan et al. (2022) | China | Grid level | 2010 | | | | | | | | 4.2 | 3.4 | | | |
| Itahashi et al. (2018) | China | Grid level | 2010 | | | | | | | | 3.5 | 2.5 | | | |
| Zhao et al. (2017) | China | Grid level | 2008-2012 | 0.3 | 1.7 | 1.0 | 0.5 | 2.6 | | | 6.6 | 3.4 | | | 18.1 |
| Xu et al. (2015) | China | 43 sites | 2010-2014 | 0.2-9.8 | 0.2-16.6 | 0.1-4.5 | 0.5-16.0 | 0.1-11.7 | | | 1.0-19.1 | 0.5-20.1 | | | 39.9 |
| Xu et al. (2019) | China | 32 sites | 2010-2015 | 3.1 | 5.2 | 1.4 | 9.6 | 3.7 | | | 11.4 | 10.3 | | | |
| Wen et al. (2020) | China | 66 sites | 2011-2018 | | | 22.5 | | | | | | 19.4 | | | |
| Pan et al. (2012) | North China | 10 sites | 2007-2010 | 0.8-4.5 | | 2.2-3.1 | 8.1-64.2 | 1.7-5.5 | | | 10.3-22.0 | 3.4-10.2 | | | 60.6 |
| Pan et al. (2013) | North China | 10 sites | 2007-2010 | | | | | | 32.4 | 12.8 | | | | 19.6 | 64.8 |
| Zhu et al. (2015) | China | Grid level | 2013 | | | | | | | | 7.3 | 5.9 | | | |
| Yu et al. (2016) | China | 43 sites | 2009-2014 | | | | | | | | 32.9 | 116.0 | | | |
| Yu et al. (2019) | China | Grid level | 1985-2015 | 0.8 | 2.0 | 2.7 | 0.5 | 4.3 | | | 5.9 | 4.2 | | | 20.4 |
| Li et al. (2019) | China | Grid level | 2010 | | | | | | | | | | | 71.5 | |
| Li et al. (2020) | China | Grid level | 2011-2016 | | | | | | | | 5.9 | 13.3 | 33.4 | | |
| Liu et al. (2016a) | Southwest China | 1 site | 2003-2013 | | | | | | | | 17.5 | 8.2 | 21.7 | | |
| Liu et al. (2016b) | China | 225 data records | 2003-2014 | | | | | | | | 6.8 | 5.4 | | | |
| Liu et al. (2016c) | China | 174 sites | 2000-2013 | | | | | | | | | | 23.0 | | |
| Liu et al. (2017a) | China | Grid level | 2010-2012 | | | | | | | | 5.8 | | | | |

Table S4 (continued)

| Reference | Study region | Research scale | Study period | Dry deposition | | | | | Wet/bulk deposition | | | Total deposition | | |
|---------------------------|-----------------|----------------|--------------|-----------------|------------------|------------------------------|-----------------|------------------------------|---------------------|-------------------------------|------------------------------|------------------------------|-------------------------------|---------|
| | | | | NO ₂ | HNO ₃ | NO ₃ ⁻ | NH ₃ | NH ₄ ⁺ | SO ₂ | SO ₄ ²⁻ | NH ₄ ⁺ | NO ₃ ⁻ | SO ₄ ²⁻ | |
| Liu et al. (2017b) | China | Grid level | 2012 | | | 1.5 | | | | | | | | |
| Liu et al. (2021) | China | Grid level | 2008-2016 | | | | | | | | 6.5 | | | |
| Luo et al. (2016) | China | 16 sites | 2010-2012 | | | | | | 2.3-26.5 | 0.5-3.4 | | | | |
| Ge et al. (2014) | China | Grid level | 2007 | | | | | | | | 9.1 | 9.1 | 48.8 | 35.0 |
| Kuribayashi et al. (2012) | China | 6 sites | 2001-2005 | | | | | | 23.5 | 3.8 | | | | 49.4 |
| Zhang et al. (2017) | China | Grid level | 2007-2014 | 0.005-8.54 | | | | | | | | | | |
| Zhou et al. (2021) | China | Grid level | 2013-2018 | 2.1-3.1 | | | | | 7.5-18.4 | | | | | |
| Qiao et al. (2015a) | Sichuan, China | 1 site | 2010-2011 | | | | | | | | 1.4 | 1.3 | 8.1 | |
| Qiao et al. (2015b) | Sichuan, China | Grid level | 2010-2011 | | | | | | | | 0.3 | 2.8 | | |
| Zhang et al. (2022) | Tibetan Plateau | 27 sites | | | | | | | | | | | | |
| Larssen et al. (2011) | South China | 4 sites | 2001-2004 | | | | | | | | 0.4-0.9 | 0.2-0.5 | 0.9-1.9 | 0.4-2.5 |
| Jiang et al. (2020) | Hunan, China | 5 sites | 2015-2016 | | | | | | 8.6 | | | | 18.2 | 26.8 |

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.

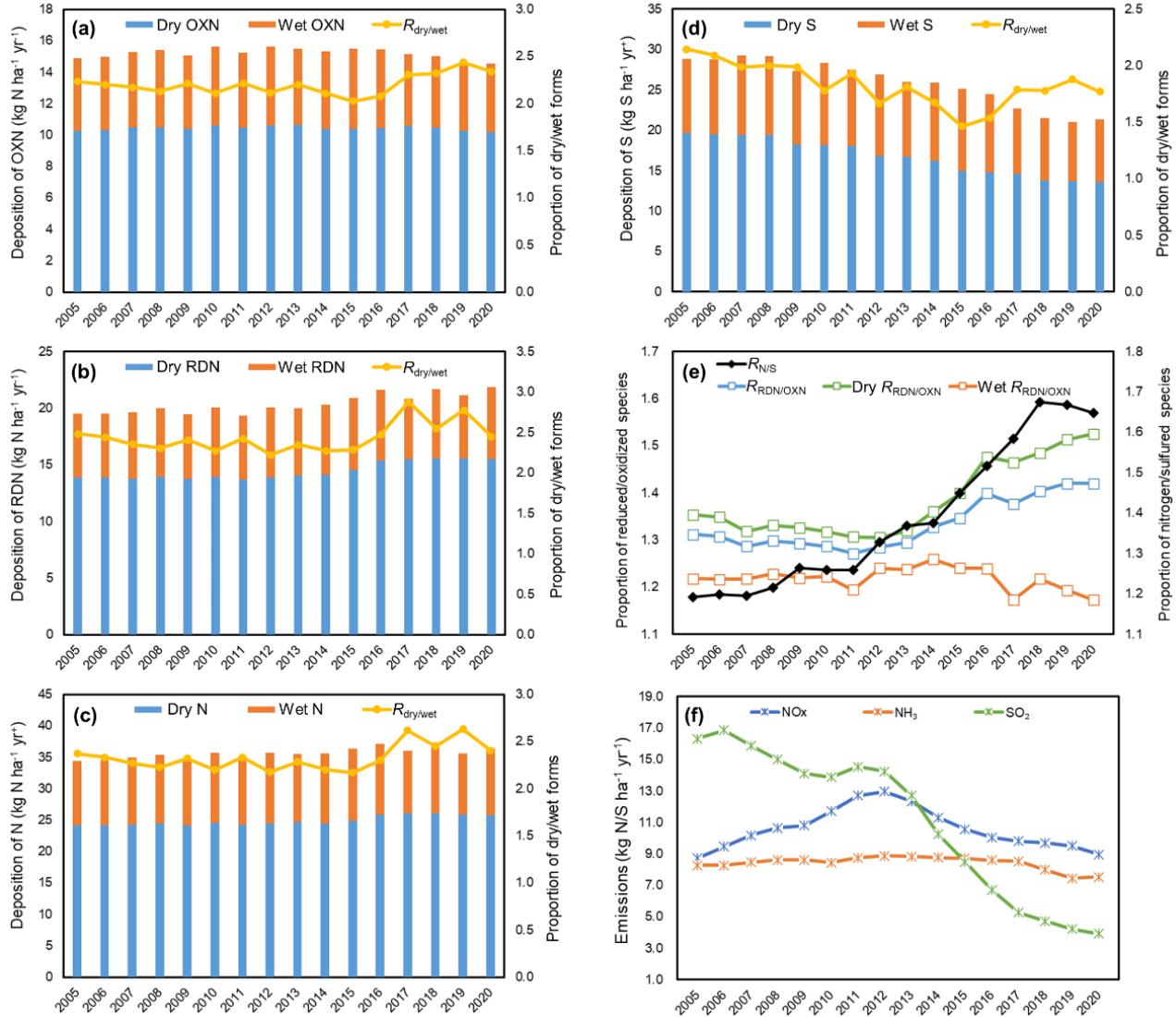


Figure 7 in the revised manuscript: The spatial distributions of N and S deposition flux in 2005-2020.

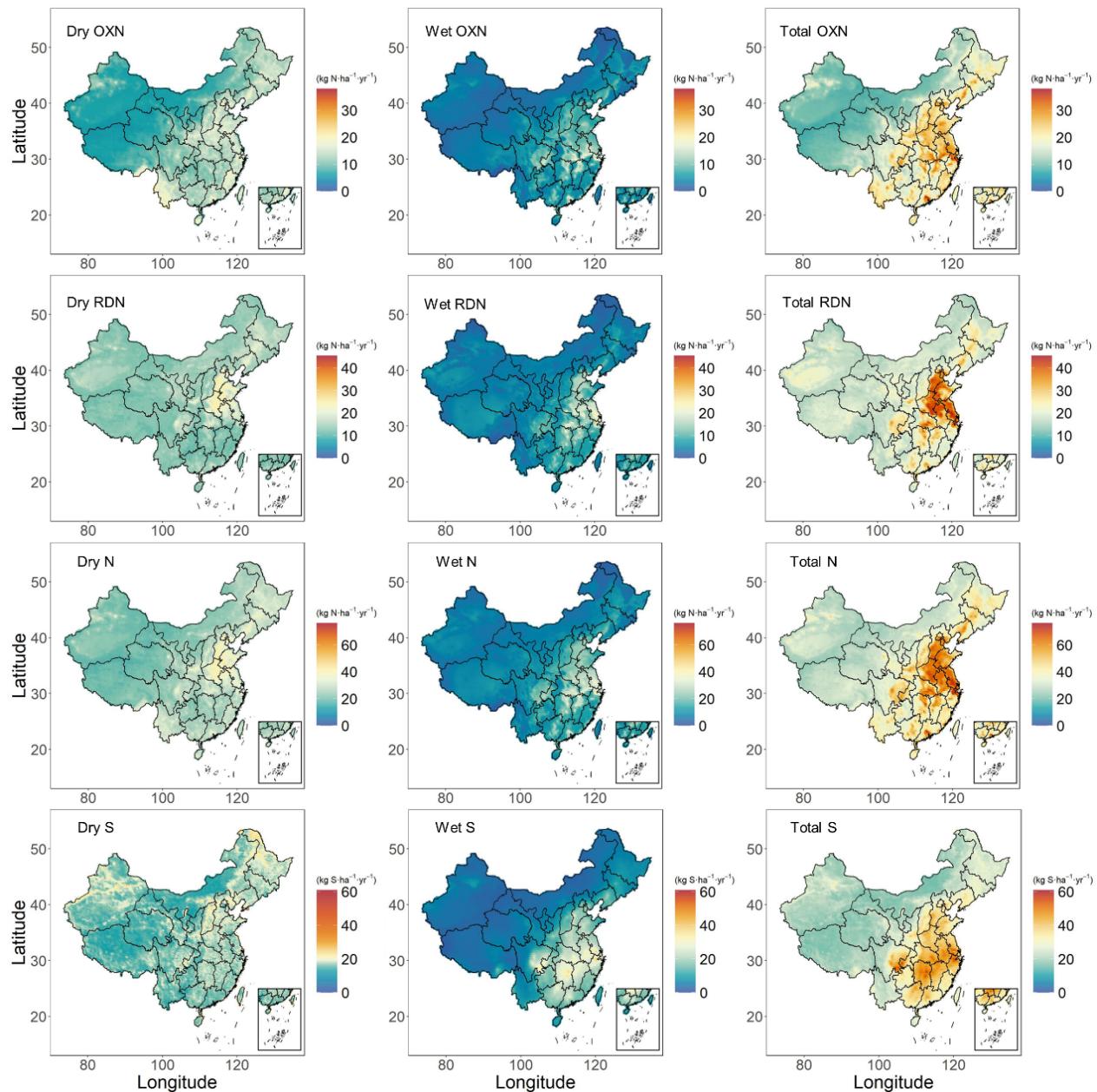
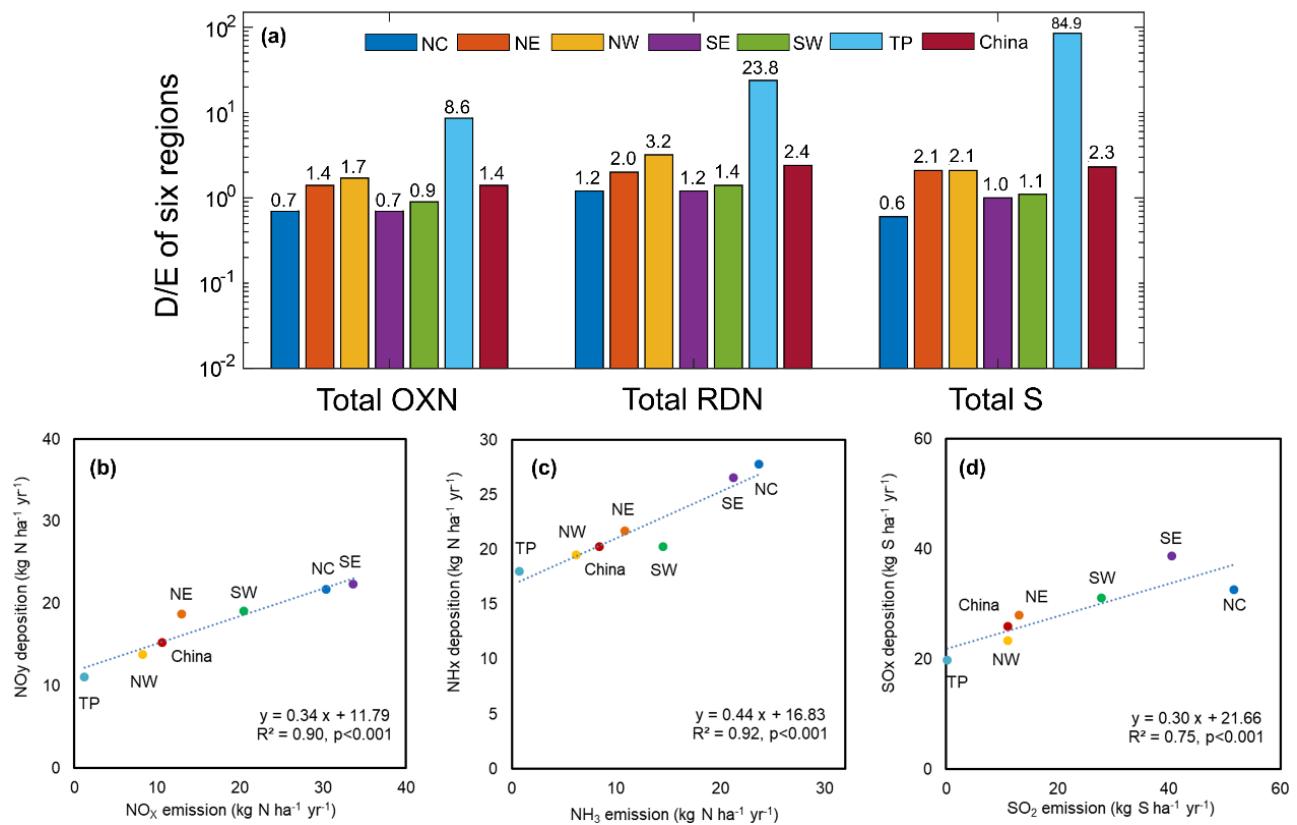


Figure 9 in the revised manuscript: Annual mean D/E ratio of OXN, RDN and sulfur from 2005 to 2020 in different regions (a) and linear relationship between regional deposition and emissions (b-d).



References

- Ge, B. Z., Wang, Z. F., Xu, X. B., Wu, J. B., Yu, X. L., and Li, J.: Wet deposition of acidifying substances in different regions of China and the rest of East Asia: modeling with updated NAQPMS, Environ. Pollut., 187, 10-21, <https://doi.org/10.1016/j.envpol.2013.12.014>, 2014.
- Itahashi, S., Yumimoto, K., Uno, I., Hayami, H., Fujita, S.-i., Pan, Y., and Wang, Y.: A 15-year record (2001–2015) of the ratio of nitrate to non-sea-salt sulfate in precipitation over East Asia, Atmos. Chem. Phys., 18, 2835-2852, <https://doi.org/10.5194/acp-18-2835-2018>, 2018.
- Jia, Y., Yu, G., Gao, Y., He, N., Wang, Q., Jiao, C., and Zuo, Y.: Global inorganic nitrogen dry deposition inferred from ground- and space-based measurements, Sci. Rep., 6, 19810, <https://doi.org/10.1038/srep19810>, 2016.
- Jia, Y., Yu, G., He, N., Zhan, X., Fang, H., Sheng, W., Zuo, Y., Zhang, D., and Wang, Q.: Spatial and decadal variations in inorganic nitrogen wet deposition in China induced by human activity, Sci. Rep., 4, 3763, <https://doi.org/10.1038/srep03763>, 2014.
- Jiang, W., Zhu, X., Shen, J., Huang, Z., Gong, D., Li, Y., and Wu, J.: Atmospheric sulfur deposition in a paddy rice region in the hilly region in central china, China Environ. Sci., 40, 4848-4856, <http://www.zghjkx.com.cn/EN/Y2020/V40/I11/4848>, 2020. (in Chinese)
- Kuribayashi, M., Ohara, T., Morino, Y., Uno, I., Kurokawa, J.-i., and Hara, H.: Long-term trends of sulfur deposition in East Asia during 1981–2005, Atmos. Environ., 59, 461-475, <https://doi.org/10.1016/j.atmosenv.2012.04.060>, 2012.
- Larssen, T., Duan, L., and Mulder, J.: Deposition and leaching of sulfur, nitrogen and calcium in four forested catchments in China: implications for acidification, Environ. Sci. Technol., 45, 1192-1198, <https://doi.org/10.1021/es103426p>, 2011.
- Li, R., Cui, L., Fu, H., Zhao, Y., Zhou, W., and Chen, J.: Satellite-based estimates of wet ammonium ($\text{NH}_4\text{-N}$) deposition fluxes across China during 2011-2016 using a space-time ensemble model, Environ. Sci. Technol., 54, 13419-13428, <https://doi.org/10.1021/acs.est.0c03547>, 2020.

Li, R., Cui, L., Zhao, Y., Meng, Y., Kong, W., and Fu, H.: Estimating monthly wet sulfur (S) deposition flux over China using an ensemble model of improved machine learning and geostatistical approach, *Atmos. Environ.*, 214, 116884, <https://doi.org/10.1016/j.atmosenv.2019.116884>, 2019.

Liaw, A. and Wiener, M.: Classification and regression by randomForest, 18-22,

Liu, L., Zhang, X., and Lu, X.: The composition, seasonal variation, and potential sources of the atmospheric wet sulfur (S) and nitrogen (N) deposition in the southwest of China, *Environ Sci. Pollut. Res. Int.*, 23, 6363-6375, <https://doi.org/10.1007/s11356-015-5844-1>, 2016a.

Liu, L., Zhang, X., Wang, S., Lu, X., and Ouyang, X.: A review of spatial variation of inorganic nitrogen (N) wet deposition in China, *PLoS One*, 11, e0146051, <https://doi.org/10.1371/journal.pone.0146051>, 2016b.

Liu, L., Zhang, X., Wang, S., Zhang, W., and Lu, X.: Bulk sulfur (S) deposition in China, *Atmos. Environ.*, 135, 41-49, <https://doi.org/10.1016/j.atmosenv.2016.04.003>, 2016c.

Liu, L., Yang, Y., Xi, R., Zhang, X., Xu, W., Liu, X., Li, Y., Liu, P., and Wang, Z.: Global wet-reduced nitrogen deposition derived from combining satellite measurements with output from a chemistry transport model, *J. Geophys. Res.*, 126, <https://doi.org/10.1029/2020jd033977>, 2021.

Liu, L., Zhang, X., Xu, W., Liu, X., Lu, X., Chen, D., Zhang, X., Wang, S., and Zhang, W.: Estimation of monthly bulk nitrate deposition in China based on satellite NO₂ measurement by the Ozone Monitoring Instrument, *Remote Sens. Environ.*, 199, 93-106, <https://doi.org/10.1016/j.rse.2017.07.005>, 2017a.

Liu, L., Zhang, X., Zhang, Y., Xu, W., Liu, X., Zhang, X., Feng, J., Chen, X., Zhang, Y., Lu, X., Wang, S., Zhang, W., and Zhao, L.: Dry particulate nitrate deposition in China, *Environ. Sci. Technol.*, 51, 5572-5581, <https://doi.org/10.1021/acs.est.7b00898>, 2017b.

Luo, X., Pan, Y., Goulding, K., Zhang, L., Liu, X., and Zhang, F.: Spatial and seasonal variations of atmospheric sulfur concentrations and dry deposition at 16 rural and suburban sites in China, *Atmos. Environ.*, 146, 79-89, <https://doi.org/10.1016/j.atmosenv.2016.07.038>, 2016.

Lye, C. and Tian, H.: Spatial and temporal patterns of nitrogen deposition in China: Synthesis of observational data, *J. Geophys. Res.*, 112, <https://doi.org/10.1029/2006jd007990>, 2007.

Nowlan, C. R., Martin, R. V., Philip, S., Lamsal, L. N., Krotkov, N. A., Marais, E. A., Wang, S., and Zhang, Q.: Global dry deposition of nitrogen dioxide and sulfur dioxide inferred from space-based measurements, *Global Biogeochem. Cycles*, 28, 1025-1043, <https://doi.org/10.1002/2014gb004805>, 2014.

Pan, Y. P., Wang, Y. S., Tang, G. Q., and Wu, D.: Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China, *Atmos. Chem. Phys.*, 12, 6515-6535, <https://doi.org/10.5194/acp-12-6515-2012>, 2012.

Pan, Y. P., Wang, Y. S., Tang, G. Q., and Wu, D.: Spatial distribution and temporal variations of atmospheric sulfur deposition in Northern China: insights into the potential acidification risks, *Atmos. Chem. Phys.*, 13, 1675-1688, <https://doi.org/10.5194/acp-13-1675-2013>, 2013.

Qiao, X., Xiao, W., Jaffe, D., Kota, S. H., Ying, Q., and Tang, Y.: Atmospheric wet deposition of sulfur and nitrogen in Jiuzhaigou National Nature Reserve, Sichuan Province, China, *Sci. Total Environ.*, 511, 28-36, <https://doi.org/10.1016/j.scitotenv.2014.12.028>, 2015a.

Qiao, X., Tang, Y., Hu, J., Zhang, S., Li, J., Kota, S. H., Wu, L., Gao, H., Zhang, H., and Ying, Q.: Modeling dry and wet deposition of sulfate, nitrate, and ammonium ions in Jiuzhaigou National Nature Reserve, China using a source-oriented CMAQ model: Part I. Base case model results, *Sci. Total Environ.*, 532, 831-839, <https://doi.org/10.1016/j.scitotenv.2015.05.108>, 2015b.

Tan, J., Su, H., Itahashi, S., Tao, W., Wang, S., Li, R., Fu, H., Huang, K., Fu, J. S., and Cheng, Y.: Quantifying the wet deposition of reactive nitrogen over China: Synthesis of observations and models, *Sci. Total Environ.*, 851, 158007, <https://doi.org/10.1016/j.scitotenv.2022.158007>, 2022.

Wen, Z., Xu, W., Li, Q., Han, M., Tang, A., Zhang, Y., Luo, X., Shen, J., Wang, W., Li, K., Pan, Y., Zhang, L., Li, W., Collett, J. L., Jr., Zhong, B., Wang, X., Goulding, K., Zhang, F., and Liu, X.: Changes of nitrogen deposition in China from 1980 to 2018, *Environ. Int.*, 144, 106022, <https://doi.org/10.1016/j.envint.2020.106022>, 2020.

Xu, W., Zhang, L., and Liu, X.: A database of atmospheric nitrogen concentration and deposition from the nationwide monitoring network in China, *Sci. Data*, 6, 51, <https://doi.org/10.1038/s41597-019-0061-2>, 2019.

Xu, W., Luo, X. S., Pan, Y. P., Zhang, L., Tang, A. H., Shen, J. L., Zhang, Y., Li, K. H., Wu, Q. H., Yang, D. W., Zhang, Y. Y., Xue, J., Li, W. Q., Li, Q. Q., Tang, L., Lu, S. H., Liang, T., Tong, Y. A., Liu, P., Zhang, Q., Xiong, Z. Q., Shi, X. J., Wu, L. H., Shi, W. Q., Tian, K., Zhong, X. H., Shi, K., Tang, Q. Y., Zhang, L. J., Huang, J. L., He, C. E., Kuang, F. H., Zhu, B., Liu, H., Jin, X., Xin, Y. J., Shi, X. K., Du, E. Z., Dore, A. J., Tang, S., Collett, J. L., Goulding, K., Sun, Y. X., Ren, J., Zhang, F. S., and Liu, X. J.: Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China, *Atmos. Chem. Phys.*, 15, 12345-12360, <https://doi.org/10.5194/acp-15-12345-2015>, 2015.

Yu, G., Jia, Y., He, N., Zhu, J., Chen, Z., Wang, Q., Piao, S., Liu, X., He, H., Guo, X., 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.

Yu, H., He, N., Wang, Q., Zhu, J., Xu, L., Zhu, Z., and Yu, G.: Wet acid deposition in Chinese natural and agricultural ecosystems: Evidence from national-scale monitoring, *J. Geophys. Res.: Atmospheres*, 121, 10,995-911,005, <https://doi.org/10.1002/2015jd024441>, 2016.

Zhang, B., Li, Z., Feng, Q., Gui, J., Zhao, Y., and Zhang, B.: Environmental significance of atmospheric nitrogen deposition in the transition zone between the Tibetan Plateau and arid region, *Chemosphere*, 307, 136096, <https://doi.org/10.1016/j.chemosphere.2022.136096>, 2022.

Zhang, X. Y., Lu, X. H., Liu, L., Chen, D. M., Zhang, X. M., Liu, X. J., and Zhang, Y.: Dry deposition of NO₂ over China inferred from OMI columnar NO₂ and atmospheric chemistry transport model, *Atmos. Environ.*, 169, 238-249, <https://doi.org/10.1016/j.atmosenv.2017.09.017>, 2017.

Zhao, Y., Zhang, L., Chen, Y., Liu, X., Xu, W., Pan, Y., and Duan, L.: Atmospheric nitrogen deposition to China: A model analysis on nitrogen budget and critical load exceedance, *Atmos. Environ.*, 153, 32-40, <https://doi.org/10.1016/j.atmosenv.2017.01.018>, 2017.

Zhou, K., Zhao, Y., Zhang, L., and Xi, M.: Declining dry deposition of NO₂ and SO₂ with diverse spatiotemporal patterns in China from 2013 to 2018, *Atmos. Environ.*, 262, 118655, <https://doi.org/10.1016/j.atmosenv.2021.118655>, 2021.

Zhu, J., He, N., Wang, Q., Yuan, G., Wen, D., Yu, G., and Jia, Y.: The composition, spatial patterns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems, *Sci. Total Environ.*, 511, 777-785, <https://doi.org/10.1016/j.scitotenv.2014.12.038>, 2015.