PREFACE

PREFACE TO THE SPECIAL ISSUE: REACTIVE NITROGEN IN THE AIR: EMISSIONS, PROCESS, DEPOSITION AND IMPACTS

Toward a better understanding of cascading consequences of atmospheric reactive nitrogen along its transport pathway

PAN Yuepeng

State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China

Over the past century, the creation of nitrogenous fertilizer has sustained food production and thereby the global population. Nevertheless, all the nitrogen used in food production is added to the environment, as is the nitrogen emitted to the atmosphere during fossil-fuel combustion (Gruber and Galloway 2008). Of particular concern is the consequence of large reactive nitrogen leaking from the 'nitrogen cascade', especially ammonia and nitrogen oxides, which can first cause air pollution and then, after it has been transported to remote areas and deposited on the ground, lead to soil acidification, water eutrophication and biodiversity loss (Galloway et al. 2003). This special issue presents a selection of 13 papers that advance our understanding of cascading consequences of reactive nitrogen species along their emission, transport, deposition, and the impacts in the atmosphere.

Emissions of gaseous nitrogen to the atmosphere

China is the top emitter of ammonia in the world—even larger than the sum of the EU and US. As one of the emission hotspots in East China, Jiangsu Province had a stable agricultural ammonia emissions level between 2000 and 2006. However, a clear increasing trend was found between 2007 and 2012, and after then a decreasing trend till 2017 (Huang et al. 2020). Of the total agricultural ammonia emissions in Jiangsu Province, 78.1% was from livestock and poultry farming, and the remaining 21.9% was from fertilizer application. This situation is true at the city scale, as presented in high-resolution (1 km × 1 km, monthly) ammonia emissions inventory of agricultural sources in Hefei City (Hou and Yu 2020). They identified two major contributors to livestock waste from broilers (34.5%) and laying hens (22.2%). Note that the average emissions density of the whole city was 2.4 t km⁻², and the higher values were mostly in areas with denser populations.

Aside from ammonia, other reactive nitrogen emissions from soils, e.g., nitrous acid and nitric oxide, is also a key process of the global nitrogen cycle and has significant implications for atmospheric chemistry. In the laboratory, Wu et al. (2020) found that freeze-dried soil had the closest emission features of nitrous acid and nitric oxide with those of fresh soil, while air-drying and oven-drying methods significantly increased gaseous fluxes. Therefore, global soil reactive nitrogen emissions could be overestimated if air- and oven-dried soils are used.

Atmospheric transport of unexpected emissions

Biomass burning also emits nitrogen oxides and ammonia, and this is of importance in agricultural regions. Wang et al. (2020) found that 91% of fire points in Northeast China from 2003 to 2017 were from croplands, and mainly occurred during March–April (37%) and October–November (46%). In the Pearl River Delta region, Guo et al. (2020) observed the linkage between severe haze episodes and biomass burning during October 2014. They also found a regional feature of haze pollution, with periodically elevated nitrate aerosols.

In summer 2015, an unexpected fire and explosion accident occurred in Tianjin Port. Of the aerosol constitutes, nitrate was most affected. Its concentrations peaked at 16.5 μg m⁻³ one week after the explosion, as observed at an island 300 km downwind of Tianjin (Zong et al. 2020). After the explosion, the stable nitrogen isotope of aerosol nitrate decreased significantly from +7.33‰ to −1.58‰. Through the inverse computation of dry deposited nitrate, the explosion-affected area should be less than 1.42 × 10⁴ km², or 20% of the total area of the Bohai Sea.

In addition to atmospheric transport, the coastal ocean also receives much riverine nitrogen and thus can become
eutrophic. The anthropogenic nitrogen in eutrophic ocean waters might be reversed to land, according to the precipitation observation at a receptor site in subtropical forest (Chang et al. 2020). They suggested that typhoons passing over eutrophic oceans can bring substantial organic nitrogen via storms and hence be an important source of nutrients to the forest.

Atmospheric deposition pattern and tracing techniques

Compared with site-based precipitation collection, moss nitrogen was suggested to be a simple technique in estimation of wet nitrogen deposition. A significant linear relationship between moss nitrogen content and wet deposition was found by Chen et al. (2020). They also determined the nitrogen isotopic composition of moss (Haplocladium microphyllum) tissues and found a more negative value at suburban and rural sites than that at urban sites, due to major contributions from agricultural and nonagricultural sources, respectively.

In addition, remote-sensing technology can provide additional information on nitrogen pollution at a regional scale. Combining the tropospheric columns from satellite measurements and the simulated profiles from an atmospheric chemistry transport model, Wang et al. (2020) estimated a total nitrogen deposition of $54.5 \pm 17.2$ kg N ha$^{-1}$ yr$^{-1}$ in North China, with high values in urban and farmland areas. In the region, atmospheric nitrogen was mainly deposited via gas (52%), followed by precipitation (36%) and particles (12%).

Impacts and mitigation of atmospheric reactive nitrogen

Due to strict control measures in China, sulfur dioxide emissions have largely been reduced in recent years. As a result, fine particle have become more sensitive to nitrogen dioxides than ammonia, indicating that nitrogen oxides emission control is more effective in reducing surface particulate concentrations in China (Xu et al. 2020). This is partially due to the abundant ammonia in the atmosphere, which needs to be substantially reduced.

Mitigation of atmospheric ammonia pollution can be achieved not only by decreasing emissions from sources, but also by increasing ammonia deposition around sources. Yi et al. (2020) found that 80% of the emitted ammonia was dry deposited within 100 m of the studied paddy fields (0.6 ha) in the subtropical hilly regions of China. Thus, not all volatilized ammonia can enter the atmosphere to form aerosols. This finding indicates that measures to increase the level of ammonia deposition around sources, such as by planting trees, are advocated to reduce the amount of ammonia pollution. In another study, Zhang et al. (2020) concluded that returning crop straw back into cropland via either incorporation or prescribed in-situ burning under favorable climate conditions could be a promising straw management practice for mitigating ammonia loss and increasing soil fertility in calcareous soils.

In summary, reactive nitrogen control in the atmosphere, especially ammonia, remains a great challenge due to its huge emissions from various sources, as well as unclear atmospheric processes and sinks. There is also increasing concern regarding its impacts on ecosystems. Note that previous nitrogen deposition impacts were mostly derived from field experiments by spraying nitrogen solution onto soils. However, this conventional technique largely ignores the dry deposition of ammonia (as well as other gaseous reactive nitrogen species). Due to the increasing trend of atmospheric ammonia concentrations worldwide and its dominant role in nitrogen deposition and haze pollution, Pan et al. (2020) suggest that the next generation of experiments should mimic nitrogen deposition on natural ecosystems by further considering the dry deposition of ammonia.

Acknowledgments

I am deeply grateful to the Editor-in-Chief of AOSL, Prof. Huijun Wang, who supported the publication of this special issue. Special thanks go to the authors, editors and reviewers. Without their invaluable help this special issue could not have been realized.

ORCID

Yuepeng PAN http://orcid.org/0000-0002-5547-0849

References

Chang, M., W. Chen, S. Deng, X. Wang, and S. Zhou. 2020. “Are Typhoon and Marine Eutrophication the Possible Missing Sources of High Dissolved Organic Nitrogen in Wet Deposition?” Atmospheric and Oceanic Science Letters 13 (3). doi:10.1080/16742834.2019.1679016.

Chen, Z., T. Huang, R. Fan, H. Yang, Y. Yang, and C. Huang. 2020. “Atmospheric Nitrogen Deposition in Yangtze River Delta: Insights Gained from the Nitrogen Content and Isotopic Composition of the Moss Haplocladium Microphyllum.” Atmospheric and Oceanic Science Letters 13 (3). doi:10.1080/16742834.2019.1688629.

Galloway, J. N., J. D. Aber, J. W. Erisman, S. P. Seitzinger, R. W. Howarth, E. B. Cowling, and B. J. Cosby. 2003. “The Nitrogen Cascade.” BioScience 53 (4): 341–356. doi:10.1641/0006-3568(2003)053[0341:TNC]2.0.CO;2.
Gruber, N., and J. N. Galloway. 2008. “An Earth-system Perspective of the Global Nitrogen Cycle.” *Nature* 451 (7176): 293–296. doi:10.1038/nature06592.

Guo, J., S. Zhou, X. Sun, M. Huang, H. Dong, M. Chang, Q. Fan, S. Fan, and X. Wang. 2020. “The Regional Nature of Nitrate-dominant Haze Pollution during Autumn over the Pearl River Delta Area.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1740055.

Hou, X., and X. Yu. 2020. “An Ammonia Emissions Inventory For Agricultural Sources In Hefei, China.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1747355.

Huang, J., R. Xiong, L. Fang, T. Li, and W. Shen. 2020. “Estimation of Interannual Trends of Ammonia Emissions from Agriculture in Jiangsu Province from 2000 to 2017.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1736499.

Pan, Y., S. Tian, D. Wu, W. Xu, X. Zhu, C. Liu, D. Li, et al. 2020. “Ammonia Should Be Considered in Field Experiments Mimicking Nitrogen Deposition.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1733919.

Wang, L., X. Jin, Q. Wang, H. Mao, Q. Liu, G. Weng, and Y. Wang. 2020. “Spatial and Temporal Variability of Open Biomass Burning in Northeast China from 2003 to 2017.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1742574.

Wang, Z., X. Zhang, L. Liu, M. Cheng, and J. Xu. 2020. “Spatial and Seasonal Patterns of Atmospheric Nitrogen Deposition in North China.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2019.1701385.

Wu, D., L. Deng, Y. Liu, D. Xi, H. Zou, R. Wang, Z. Sha, Y. Pan, L. Hou, and M. Liu. 2020. “Comparisons of the Effects of Different Drying Methods on Soil Nitrogen Fractions: Insights into Emissions of Reactive Nitrogen Gases (HONO and NO).” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1733388.

Xu, G., Q. Zang, Y. Yao, and X. Zhang. 2020. “Changes in PM$_{2.5}$ Sensitivity to NO$_x$ and NH$_3$ Emissions Due to a Large Decrease in SO$_2$ Emissions from 2013 to 2018.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1738009.

Yi, Y., J. Shen, C. Yang, J. Wang, Y. Li, and J. Wu. 2020. “Dry Deposition of Ammonia around Paddy Fields in the Subtropical Hilly Area in Southern China.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1738208.

Zhang, B., M. Zhou, H. Lin, N. Tite, Y. Wang, and B. Zhu. 2020. “Effects of Different Long-term Crop Straw Management Practices on Ammonia Volatilization from Subtropical Calcareous Agricultural Soil.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2020.1736498.

Zong, Z., Z. Sun, Y. Tan, C. Tian, L. Qu, and L. Ji. 2020. “Impact of an Accidental Explosion in Tianjin Port on Enhanced Atmospheric Nitrogen Deposition over the Bohai Sea Inferred from Aerosol Nitrate Dual Isotopes.” *Atmospheric and Oceanic Science Letters* 13 (3). doi:10.1080/16742834.2019.1682926.