

# Reduced nitrogen dominated nitrogen deposition in the United States, but its contribution to nitrogen deposition in China decreased

Xuejun Liu<sup>a,1</sup>, Wen Xu<sup>a</sup>, Enzai Du<sup>b</sup>, Yuepeng Pan<sup>c</sup>, and Keith Goulding<sup>d</sup>

Recently, Li et al. (1) report that reduced nitrogen (N) dominated both wet and dry deposition of inorganic N, following long-term N oxides (NO<sub>x</sub>) emission controls introduced in 1990. They systematically compare wet deposition of inorganic N, measured by the US National Atmospheric Deposition Program (NADP) between 1990–1992 and 2010–2012, and calculate N dry deposition from measurements of concentrations made by Clean Air Status and Trends Network, NADP Ammonia Monitoring Network, and Interagency Monitoring of Protected Visual Environments NH<sub>x</sub> in 2011–2013. Their results show the transition from oxidized N-dominated wet deposition in the early 1990s to reduced N-dominated wet deposition in the early 2010s. According to their analysis (1), this transition reflects increases in agricultural ammonia (NH<sub>3</sub>) emissions and the success of regulatory policies in decreasing NO<sub>x</sub> emissions, which was consistent with an earlier report on wet deposition (2). If correct, this is a significant change that needs to be noted by the science community and policy makers working to reduce atmospheric pollution in the United States (3).

In rapidly developing countries like China, however, with the simultaneous intensive growth in industry and agriculture, another N deposition pattern was observed. Liu et al. (4) found that wet (i.e., bulk) N deposition increased significantly (with average increase of 0.41 kg N·ha<sup>-1</sup>·y<sup>-1</sup>) while the ratios of reduced N to oxidized N in precipitation decreased significantly between the 1980s (~5.0) and 2000s (~2.0). Their N deposition data were consistent with China's national NH<sub>3</sub> and NO<sub>x</sub> emission trends and similar decreasing ratios of NH<sub>3</sub>–N:NO<sub>x</sub>–N from 1980 to 2010, suggesting the increased importance of oxidized N emissions and deposition in China. More recently, Xu et al. (5) reported that the reduced N

content of wet deposition was on average 55% (34–71%) in the early 2010s (2010–2014) (5), compared with 62% between the mid-1990s and 2000s (6) and 80% in the 1980s (4). Although reduced N still dominates N deposition in China especially in northern China (7), oxidized N has been increasing in importance and will soon dominate, unless strict emission control measures are adopted (8).

One other important point to note is that Li et al. (1) do not include dry NO<sub>2</sub>, dry particulate organic N, and wet organic N deposition in total N deposition estimate. Benedict et al. (9) showed the deposition of these N species can be up to 0.7 kg N·ha<sup>-1</sup>·y<sup>-1</sup> in the Rocky Mountain area. Moreover, Li et al.'s important finding fails to consider particulate ammonium and nitrate dry deposition from coarse particles. This omission will especially lead to an underestimation of dry nitrate deposition because the majority of the dry-deposited nitrate results from coarse particulate matter rather than fine particles (e.g., PM<sub>2.5</sub>) (7). Taken together, Li et al. (1) may have therefore calculated a higher contribution of reduced N in total N deposition budget. If wet and dry deposition of the non-included N species (i.e., NO<sub>2</sub>, coarse nitrate, and organic N) averaged 1 kg N·ha<sup>-1</sup>·y<sup>-1</sup>, the reduced N contribution to total N deposition in the United States would be no more than 50%. Assuming that reduced N, essentially NH<sub>3</sub>, dominates, emissions and deposition could result in inappropriate air pollution policies.

## Acknowledgments

This paper was supported by Chinese State Basic Research Program Grant 2014CB954200, National Natural Science Foundation of China Grants 40425007 and 41405144, and the Sino–UK Virtual Centre for Improving Nitrogen in Agronomy.

<sup>a</sup>Center for Resources, Environment and Food Security, Key Laboratory of Plant–Soil Interactions of Ministry of Education, College of Resources and Environmental Sciences, China Agricultural University, Beijing 100193, China; <sup>b</sup>State Key Laboratory of Earth Surface Processes and Resource Ecology, and College of Resources Science and Technology, Beijing Normal University, Beijing 100875, China; <sup>c</sup>State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China; and <sup>d</sup>The Sustainable Soil and Grassland Systems Department, Rothamsted Research, West Common, Harpenden, Hertfordshire AL5 2JQ, United Kingdom

Author contributions: X.L. designed research; X.L. and W.X. performed research; X.L. analyzed data; and X.L., E.D., Y.P., and K.G. wrote the paper. The authors declare no conflict of interest.

<sup>1</sup>To whom correspondence should be addressed. Email: liu310@cau.edu.cn.

- 
- 1 Li Y, et al. (2016) Increasing importance of deposition of reduced nitrogen in the United States. *Proc Natl Acad Sci USA* 113(21):5874–5879.
  - 2 Du EZ, de Vries W, Galloway JN, Hu X, Fang J (2014) Changes in wet nitrogen deposition in the United States between 1985 and 2012. *Environ Res Lett* 9(9): 095004.
  - 3 Stokstad E (2014) Air pollution. Ammonia pollution from farming may exact hefty health costs. *Science* 343(6168):238.
  - 4 Liu X, et al. (2013) Enhanced nitrogen deposition over China. *Nature* 494(7438):459–462.
  - 5 Xu W, et al. (2015) Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. *Atmos Phys Chem* 15(21):12345–12360.
  - 6 Du EZ, Liu XJ (2014) High rates of wet nitrogen deposition in China: A synthesis. *Nitrogen Deposition, Critical Loads and Biodiversity*, eds Sutton MA, et al. (Springer Science and Business Media, Dordrecht, The Netherlands), Chap 6, pp 49–56.
  - 7 Pan YP, et al. (2013) Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China. *Atmos Chem Phys* 12(14):6515–6535.
  - 8 Liu X, et al. (2016) Evidence for a historic change occurring in China. *Environ Sci Technol* 50(2):505–506.
  - 9 Benedict KB, et al. (2013) A seasonal nitrogen deposition budget for Rocky Mountain National Park. *Ecol Appl* 23(5):1156–1169.