Ammonia should be considered in field experiments mimicking nitrogen deposition

PAN Yuepeng, TIAN Shili, WU Dianming, XU Wen, ZHU Xiaying, LIU Chunyan, LI Dejun, FANG Yunting, DUAN Lei, LIU Xuejun, and WANG Yuesi

State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China; Key Laboratory of Geographic Information Sciences, Ministry of Education, School of Geographic Sciences, East China Normal University, Shanghai, China; College of Resources and Environmental Sciences, China Agricultural University, Beijing, China; National Climate Center, China Meteorological Administration, Beijing, China; Key Laboratory of Agro-ecological Processes in Subtropical Region, Institute of Subtropical Agriculture, Chinese Academy of Sciences, Changsha, Hunan, China; CAS Key Laboratory of Forest Ecology and Management, Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang, Liaoning, China; State Key Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, China

ABSTRACT
Excess nitrogen deposition has significant impacts on water eutrophication, soil acidification, elevated greenhouse gas emissions, and biodiversity loss. These impacts mostly derive from conventional manipulative experiments in the field by adding nitrogen solution directly onto grassland or forest floors. For forest ecosystems, previous field experiments have usually ignored the nitrogen cycles in the canopy, which are important in response to airborne nitrogen input. Although whole-forest canopy nitrogen fertilization has recently been conducted to promote our understanding of nitrogen deposition processes, spraying nitrogen solution onto plants still largely ignores the dry deposition of ammonia (as well as other gaseous reactive nitrogen species). To date, there have been a limited number of field studies that have investigated the bi-directional exchange of ammonia between the atmosphere and plants, not to mention the impacts of ammonia on natural ecosystems. Due to the increasing trend of atmospheric ammonia concentrations worldwide and its dominant role in nitrogen deposition and haze pollution, the next generation of experiments should mimic nitrogen deposition on natural ecosystems by further considering the dry deposition of ammonia.

Since the early 20th century, the global nitrogen (N) cycle has been significantly altered by increasing fertilizer production and fossil fuel combustion (Pan et al. 2016; Vitousek 1997). Globally, reactive N production increased from 15 Tg N yr⁻¹ in 1860 to 187 Tg N yr⁻¹ in 2005 (Gruber and Galloway 2008). More than half of this human-produced N was ultimately returned to the ground via wet and dry deposition. Following Europe (Dise and Wright 1995) and North America (Fenn et al. 1998), East Asia has become a new hotspot of atmospheric N deposition over recent decades (Pan et al. 2012; Xu et al. 2015). When the additional N deposition exceeds the N demand of natural ecosystems, cascading environmental problems may occur, including water eutrophication, soil acidification, elevated greenhouse gas emissions, and biodiversity loss (Fowler et al. 2013; Galloway et al. 2003; Huang et al. 2015).

To quantify the impacts of atmospheric N deposition on natural ecosystems, manipulative field experiments mimicking N deposition have been extensively conducted, e.g., by adding N solution directly onto grassland or forest floors (Fang et al. 2009; Mo et al. 2007; Xia, Niu, and Wan 2009 (Figure 1a)). However, these conventional experiments tend to focus on underground
processes and ignore the impacts of N deposition on the canopy, which plays an important role in the N cycle of forest ecosystems (Zhang et al. 2015).

To address these canopy-associated concerns, whole-forest canopy N fertilization experiments have recently been designed (Figure 1(b)). For example, aircraft fertilization was performed above the canopy in a mature spruce–hemlock forest in the United States (Gaige et al. 2007). Also, pump lifting systems and canopy spraying experiments were conducted in a subtropical temperate deciduous forest in China (Zhang et al. 2015). These canopy-based N addition experiments have revealed many important N cycling processes that are different from those revealed by experiments targeting forest floors. Thus, the incomplete knowledge gained from the conventional approach focusing on forest floors cannot be used to extrapolate the impacts of N deposition on forest ecosystems (Zhang et al. 2015).

Although this new canopy-based N fertilization technology has improved our knowledge of atmospheric N deposition processes in forest ecosystems, attempts thus far are still insufficient to simulate atmospheric N deposition that occurs naturally. Spraying N solutions onto plants assumes that atmospheric N species are only deposited as rainfall (wet deposition). Such a traditional technique can be a good proxy for atmospheric wet N deposition owing to its episodic manner in nature. However, dry deposition of gaseous and particulate reactive N is a dependable process involved in the continuous input of airborne N into ecosystems (Wesely 1989). To the best of our knowledge, few field experiments (if any) have evaluated the impacts of dry N deposition, not to mention comparisons of dry versus wet N deposition effects (Sheppard et al. 2011). Also, the roles of different N species, e.g., oxidized versus reduced forms, have not been fully considered. In particular, gaseous ammonia (NH₃) exchange between the atmosphere and leaves has not been well resolved in the field.

In fact, dry deposition contributes half of the total deposited N worldwide, with NH₃ being the dominant species (Li et al. 2016; Pan et al. 2012; Xu et al. 2015). Compared with the decreasing NOₓ concentrations, atmospheric NH₃ concentrations have substantially increased over the past decade (Warner et al. 2017), and thus the resulting depositions of reduced N have grown over China and the United States (Liu et al. 2016; Pan et al. 2018). In addition, a unique long-term field manipulation found that dry deposition of NH₃ gas, rather than wet deposition of ammonium ions, played a vital role in shifting peatland plant species composition (Sheppard et al. 2011). This finding highlighted the potential for NH₃ to destroy natural ecosystems. Clearly, future field and modeling experiments regarding N deposition effects on ecosystems will be incomplete if they do not take dry deposition into account.

To date, there has only been one experiment that manipulated the NH₃ dry deposition into an ombrotrophic bog ecosystem (Sheppard et al. 2011), but the scenario remains largely unclear in other natural ecosystems (Adriaenssens et al. 2012; Cape et al. 2009; Pitcairn et al. 1998). Therefore, aside from wet deposition, we suggest there is an urgent need for further consideration of the effects of dry deposition (of different N forms) in future manipulative field experiments (Figure 1(c)). We propose that the first step should be to conduct a new generation of experiments that mimics N deposition; specifically, Free-Air NH₃ Enrichment (FANE) experiments should be conducted, which will allow the investigation of the response of native ecosystems to rising NH₃ concentrations in the atmosphere.

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ORCID

Yuepeng PAN http://orcid.org/0000-0002-5547-0849
Dianming WU http://orcid.org/0000-0002-0414-9430
Yunting FANG http://orcid.org/0000-0001-7531-546X

References

Adriaenssens, S., J. Staelens, K. Wuyts, S. Van Wittenbergh, T. Wuytack, K. Verheyen, P. Boeckx et al. 2012. “Canopy Uptake of 15NH3 by Four Temperate Tree Species and the Interaction with Leaf Properties.” Water, Air, & Soil Pollution 223 (9): 5643–5657. doi:10.1007/s11270-012-1304-4.

Cape, J. N., L. J. van der Eerden, L. J. Sheppard, I. D. Leith, and M. A. Sutton. 2009. “Evidence for Changing the Critical Level for Ammonia.” Environmental Pollution 157 (3): 1033–1037. doi:10.1016/j.envpol.2008.09.049.

Dise, N. B., and R. F. Wright. 1995. “Nitrogen Leaching from European Forests in Relation to Nitrogen Deposition.” Forest Ecology and Management 71 (1): 153–161. doi:10.1016/0378-1127(94)06092-W.

Fang, Y., P. Gundersen, J. Mo, and W. Zhu. 2009. “Nitrogen Leaching in Response to Increased Nitrogen Inputs in Subtropical Monsoon Forests in Southern China.” Forest Ecology and Management 257 (1): 332–342. doi:10.1016/j.foreco.2008.09.004.

Fenn, M. E., M. A. Poth, J. D. Aber, J. S. Baron, B. T. Bormann, D. W. Johnson, A. D. Lemly et al. 1998. “Nitrogen Excess in North American Ecosystems: Predisposing Factors, Ecosystem Responses, and Management Strategies.” Ecological Applications 8 (3): 706–733. doi:10.1890/1051-0761(1998)008[0706:NEINAE]2.0.CO;2.

Fowler, D., M. Coyle, U. Skiba, M. A. Sutton, J. N. Cape, S. Reis, L. J. Sheppard et al. 2013. “The Global Nitrogen Cycle in the Twenty-first Century.” Philosophical Transactions of the Royal Society B: Biological Sciences 368 (1621): 20130164. doi:10.1098/rstb.2013.0164.

Gaige, E., D. B. Dail, D. Y. Hollinger, A. A. Davidson, I. J. Fernandez, H. Sievering, A. White et al. 2007. “Changes in Canopy Processes following Whole-Forest Canopy Nitrogen Fertilization of a Mature Spruce-Hemlock Forest.” Ecosystems 10 (7): 1133–1147. doi:10.1002/102001-007-9081-4.

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.” Biogeochemistry 53 (4): 341–356. doi:10.1021/0006-3568(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: 293. doi:10.1038/nature06592.

Huang, Y., R. Kang, J. Mulder, T. Zhang, and L. Duan. 2015. “Nitrogen Saturation, Soil Acidification, and Ecological Effects in a Subtropical Pine Forest on Acid Soil in Southwest China.” Journal of Geophysical Research: Biogeosciences 120 (11): 2457–2472.

Li, Y., B. A. Schichtel, J. T. Walker, D. B. Schwede, X. Chen, C. M. B. Lehmann, M. A. Puchalski et al. 2016. “Increasing Importance of Deposition of Reduced Nitrogen in the United States.” Proceedings of the National Academy of Sciences of the United States of America 113 (21): 5874–5879. doi:10.1073/pnas.1525736113.

Liu, X., W. Xu, E. Du, Y. Pan, and K. Goulding. 2016. “Reduced Nitrogen Dominated Nitrogen Deposition in the United States, but Its Contribution to Nitrogen Deposition in China Decreased.” Proceedings of the National Academy of Sciences of the United States of America 113 (26): E3590–3591. doi:10.1073/pnas.1607507113.

Mo, J., W. Zhang, W. Zhu, Y. Fang, D. Li, and P. Zhao. 2007. “Response of Soil Respiration to Simulated N Deposition in a Disturbed and a Rehabilitated Tropical Forest in Southern China.” Plant and Soil 296 (1): 125–135. doi:10.1007/s11104-007-9303-8.

Pan, Y., S. Tian, D. Liu, Y. Fang, X. Zhu, Q. Zhang, B. Zheng et al. 2016. “Fossil Fuel Combustion-related Emissions Dominate Atmospheric Ammonia Sources during Severe Haze Episodes: Evidence from 15N-stable Isotope in Size-resolved Aerosol Ammonium.” Environmental Science & Technology 50 (15): 8049–8056. doi:10.1021/acs.est.6b00634.

Pan, Y., S. Tian, Y. Zhao, L. Zhang, X. Zhu, J. Gao, W. Huang et al. 2018. “Identifying Ammonia Hotspots in China Using a National Observation Network.” Environmental Science & Technology 52 (7): 3926–3934. doi:10.1021/acs.est.7b05235.

Pan, Y. P., Y. S. Wang, G. Q. Tang, and D. Wu. 2012. “Wet and Dry Deposition of Atmospheric Nitrogen at TenSites in Northern China.” Atmospheric Chemistry and Physics 12 (14): 6515–6535. doi:10.5194/acp-12-6515-2012.

Pitcairn, C. E. R., I. D. Leith, L. J. Sheppard, M. A. Sutton, D. Fowler, R. C. Munro, S. Tang et al. 1998. “The Relationship between Nitrogen Deposition, Species Composition and Foliar Nitrogen Concentrations in Woodland Flora in the Vicinity of Livestock Farms.” Environmental Pollution 102 (1): 41–48. doi:10.1016/S0269-7491(98)00013-4.

Sheppard, L. J., I. D. Leith, T. Mizunuma, J. Neil Cape, A. Crossley, S. Leeson, M. A. Sutton et al. 2011. “Dry Deposition of Ammonia Gas Drives Species Change Faster than Wet Deposition of Ammonium Ions: Evidence from a Long-term Field Manipulation.” Global Change Biology 17 (12): 3589–3607. doi:10.1111/j.1365-2486.2011.02478.x.

Vitousek, P. 1997. “Human Domination of Earth’s Ecosystems.” Science 277 (5325): 494–499. doi:10.1126/science.277.5325.494.

Warner, J. X., R. R. Dickerson, Z. Wei, L. L. Strow, Y. Wang, and Q. Liang. 2017. “Increased Atmospheric Ammonia over the World’s Major Agricultural Areas Detected from Space.” Geophysical Research Letters 44 (6): 2875–2884. doi:10.1002/2016GL072305.

Wesely, M. L. 1989. “Parameterization of Surface Resistances to Gaseous Dry Deposition in Regional-scale Numerical Models.” Atmospheric Environment 23 (6): 1292–1304. doi:10.1007/0006-9818(89)090153-4.
Xia, J., S. Niu, and S. Wan. 2009. “Response of Ecosystem Carbon Exchange to Warming and Nitrogen Addition during Two Hydrologically Contrasting Growing Seasons in a Temperate Steppe.” Global Change Biology 15 (6): 1544–1556. doi:10.1111/gcb.2009.15.issue-6.

Xu, W., X. S. Luo, Y. P. Pan, L. Zhang, A. H. Tang, J. L. Shen, Y. Zhang et al. 2015. “Quantifying Atmospheric Nitrogen Deposition through a Nationwide Monitoring Network across China.” Atmospheric Chemistry and Physics 15 (21): 12345–12360. doi:10.5194/acp-15-12345-2015.

Zhang, W., W. Shen, S. Zhu, S. Wan, Y. Luo, J. Yan, K. Wang, et al. 2015. “CAN Canopy Addition of Nitrogen Better Illustrate the Effect of Atmospheric Nitrogen Deposition on Forest Ecosystem?” Scientific Reports 5: 11245. doi:10.1038/srep11245.