Clean Air Status and Trends Network (CASTNET, SPD111), National Atmospheric Deposition Program/National Trends Network (NTN, TN04), National Atmospheric Deposition Program/Ammonia Monitoring Network (AMoN, TN04) site in Claiborne County, TN.
Photo courtesy of U.S. Environmental Protection Agency, CASTNET.

Evolution of Monitoring and Modeling

of Reactive Nitrogen Deposition in the United States

by John T. Walker and Greg M. Beachley

The authors consider three overarching examples where continued evolution of monitoring and modeling are needed to improve nitrogen deposition budgets.

Deposition of reactive nitrogen (Nr)—that is, all forms of nitrogen that are biologically, photochemically, or radiatively active—can contribute to eutrophication and acidification, changes in biodiversity, reduced resilience to climate variability, and other impacts in terrestrial and aquatic ecosystems.[1] Accurate and complete atmospheric deposition budgets of nutrients and acidity are fundamental to critical load frameworks, which are used by the U.S. Environmental Protection Agency (EPA) to quantitatively link deposition to negative effects on soils, water, vegetation, visibility, and other aspects of public welfare.

The critical load describes the amount of atmospheric deposition to an ecosystem below which harmful effects do not occur and has become an important component of the review of the secondary U.S. National Ambient Air Quality Standards (NAAQS).[1-4] Critical loads are also used by land management agencies to guide air pollution management for national parks, forests, and wilderness areas.[5,6] An estimate of total Nr deposition is needed to determine if an ecosystem is receiving more or less Nr than the critical load (i.e., the exceedance). Nr deposition budgets used for critical loads assessments are developed from measurements, models, and combinations of the two.[7]

Members of the National Atmospheric Deposition Program Total Deposition Science Committee (NADP/TDep), along with collaborators from federal agencies and academia, recently completed a detailed report on the state of the science of Nr deposition budgets in the United States.[8] The report highlights that while much progress has been made in improving deposition budgets over the past decade, further improvements remain limited by important data and knowledge gaps. Policy-relevant research needs identified in the report address monitoring, process-level measurements, modeling, and source apportionment. In this article, we summarize three overarching examples where continued evolution of monitoring and modeling are needed to inform changing trends in the atmospheric composition of Nr, better understand spatial patterns of deposition in urban environments, and improve the accuracy of modeled deposition estimates to account for specific land-types.

Sources, Patterns, and Processes of NHx Deposition

Due to the decline in emissions of oxides of nitrogen (NOx) under the U.S. Clean Air Act, reduced forms of atmospheric Nr (NHx = ammonia (NH_3) + ammonium (NH_4^+)) are becoming an increasingly important component of Nr deposition[9] across the United States. However, development of a more complete understanding of the magnitude and spatial patterns of NHx deposition is limited by the completeness and accuracy of NH_3 emission inventories, as well as monitoring and modeling of NHx deposition. This is particularly true of agricultural sources and regions, as confined animal feeding operations (CAFOs, 55%) and fertilized soils (25%) account for approximately 80% of NH_3 emissions in the United States.[10] While increasing trends in NH_3 concentrations have been documented,[11,12] linking these trends to changes in emissions is difficult due to limitations in inventories.[13,14] Additionally, quantifying the dry component of NHx deposition remains difficult due to uncertainties in modeling the bidirectional air–surface exchange of NH_3.[15] While models are improving, there remains a paucity of flux measurement datasets by which to evaluate and improve air-surface exchange models for North American ecosystems.[16]

As further described in an accompanying article elsewhere in this issue, better characterization of the spatial variability of atmospheric NH_3 concentrations in agricultural areas is needed for evaluation and improvement of emission inventories, improvement of chemical transport models (CTMs) to more accurately simulate particulate matter formation and deposition, and treatment of NH_3 dry deposition in measurement-model fusion approaches.[7] The NADP Ammonia Monitoring Network (AMoN), which began in 2007, is currently the only national monitoring effort for NH_3 in the United States (currently ~100 sites). However, most of the sites are located in the eastern United States, many in counties with relatively low NH_3 emissions. Broad geographical gaps in monitoring exist over areas of the West and Midwest, where agricultural NH_3 emissions are large, and many smaller hot-spot areas are also missed.

As shown in Figure 1, landscapes often contain a patchwork of agricultural sources and natural land use, creating high spatial and temporal variability in NH_3 concentrations and deposition that can be challenging for both monitoring and modeling. Expansion of AMoN monitoring in agricultural areas, informed by satellite measurements and emission inventories to identify the most valuable and representative new monitoring locations, would help to better characterize spatial and temporal patterns in agricultural regions at relatively low cost.

Urban Deposition Issues

The primary monitoring networks that support deposition assessments in the United States—NADP and Clean Air Status and Trends Network (CASTNET)—were originally designed in the 1970s and 1980s to track changes in acidic deposition resulting from NOx and sulfur oxides (SOx) emission reductions from electricity generating units. Monitoring sites were therefore intentionally located in primarily rural locations to be regionally representative. For this reason, urban areas are not well characterized by these networks and deposition in urban environments is instead extrapolated from measurements in non-urban locations (e.g., NADP wet deposition mapping protocol) or modeled (e.g., TDep dry deposition[7]).

However, both measurements and models show urban areas (Figure 1, lower right) as hot-spots for deposition of oxidized and reduced forms of Nr, owing to a high density of mobile source emissions. Numerous studies have documented gradients in Nr deposition from urban to rural areas in the United States (e.g., Los Angeles,[17-19] New York,[20] Boston,[21,22] Phoenix,[23] and Salt Lake City[24]).

Expanded routine monitoring is needed to better understand the role of atmospheric Nr deposition in urban water quality and to better inform management of total maximum daily loads (TMDL) and other water quality issues downstream.[25] This would involve the expansion of networks such as NADP National Trends Network (NTN) in urban areas, which would benefit from coordination of deposition and water quality monitoring.[26] Air concentrations of oxidized Nr (NOx, NOy, NO_2) are already monitored in many urban areas (e.g., EPA Air Quality System <www.epa.gov/aqs>). Utilization of these datasets in measurement-model fusion techniques such as NADP TDep[7] is needed. Existing monitoring could be complemented with urban sampling of NH_3 using low-cost passive sampling such as employed by AMoN[27] to better understand patterns of NHx deposition.[19] Improvements to deposition algorithms in CTMs will also be needed to more accurately represent Nr dry deposition in urban environments.

Land-Use-Specific Deposition Estimates

CTMs and measurement-model fusion techniques[7] used for North American deposition assessments provide estimates of deposition as averages over model grid cells. Grid cells may be on the order of 10 km × 10 km or larger in size and often contain multiple types of land use and land cover, each of which has different physical, biological, and biogeochemical characteristics that affect air–surface exchange. Through their influence on air–surface exchange, these characteristics can result in large differences in deposition among the ecosystems present in the cell. To estimate fluxes to the grid cell, models average sub-grid variability in land surface characteristics or the deposition velocities derived from the model, leading to often large differences between grid-average and ecosystem-specific fluxes. Calculation of a critical load exceedance for a specific ecosystem using a grid-average deposition estimate may therefore contain large uncertainty.

Differences may be particularly large for species that deposit rapidly, such as nitric acid (HNO_3), or that depend on biogeochemical characteristics of the vegetation and soil such as NH_3. Deposition of HNO_3 is limited primarily by turbulent transfer from the atmosphere to the receptor surface, the resistance to deposition being a strong function of the roughness and surface area of the vegetation. HNO_3 will therefore deposit more rapidly to a forest (Figure 1, bottom left) than a grass field. Recent studies show that deposition of HNO_3 to forests can differ substantially from the corresponding grid average value,[28,29] highlighting the impact of sub-grid heterogeneity and model grid size on deposition estimates from CTMs.

Figure 1. Examples of landscapes where improvements in monitoring and modeling of Nr deposition are needed: (1) mixed agricultural (light colored fields and CAFOs) and natural land use types (top center); (2) rural forest (bottom left); and (3) highly urbanized (bottom right).
Source: Images of agricultural and urban landscapes attributed to Google DigitalGlobe and TerraMetrics Map Data.

NH_3 is exchanged bidirectionally with the surface depending on the difference between the atmospheric concentration and the surface compensation point, which is a function of the nitrogen status and acidity of the exchange surface.[15] While forests may be net sinks for atmospheric NH_3, crops are expected to be net sources due to higher nitrogen content of the soil and vegetation resulting from fertilization.[30] In model grid cells containing a mix of crops and natural land use types (Figure 1, top center), the grid-average flux may therefore be much different than the actual flux to the natural ecosystems. These studies highlight the need for CTMs and measurement-model fusion techniques to output land-use-specific fluxes for critical load applications, an option that is becoming available in newer versions of CTMs.[28,29,31]

These brief examples highlight aspects of monitoring and modeling that must continue to progress to reduce uncertainties in Nr deposition budgets used for policy assessments in the United States. Additional detail on these and other topics can be found in the recent EPA report on the subject[8] available at the National Atmospheric Deposition Program website at http://nadp.slh.wisc.edu/committees/tdep/reports/nrDepWhitePaper.aspx.**em**

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