Urban wet deposition monitoring station, Denver, CO.
Photo courtesy of Gregory Wetherbee, U.S. Geological Survey.

Long-Term Trends
in Reactive Nitrogen Deposition
in the United States

by Greg M. Beachley, Chris M. Rogers, Tom F. Lavery, John T. Walker, and Melissa A. Puchalski

The authors identify several examples where improvements in monitoring, modeling, and emissions inventories are needed to better characterize the linkages between trends in emissions and changes in the atmospheric composition of reactive nitrogen.

Long-term monitoring of ambient air quality and deposition is necessary to characterize trends in human and ecosystem exposure and to gauge the effectiveness of air pollution control programs. Such datasets are rare because of the difficulty and capital required to consistently and accurately collect and analyze samples over time from a spatially adequate number of regionally representative sites. Most of the national air pollutant monitoring networks producing these datasets were established in the 1970s and 1980s and focused on the human health-based U.S. National Ambient Air Quality Standards (NAAQS) criteria pollutants (e.g., sulfur dioxide [SO_2], nitrogen dioxide [NO_2], ozone [O_3], and particulate matter < 2.5 µm [$PM_{2.5}$]) or reporting acid rain trends and visibility impairment.

Under Title IV of the U.S. Clean Air Act Amendments (CAAA), electric generating units (EGU) were required to make significant reductions in emissions of SO_2 and oxides of nitrogen (NOx; i.e., nitric oxide [NO] and nitrogen dioxide [NO_2]). While NOx has continued to be regulated under stationary and mobile emissions programs (e.g., NOx Budget Trading Program), reduced nitrogen (NHx; i.e., particulate ammonium [pNH_4] plus gaseous ammonia [NH_3]) remains unregulated despite its contributions to $PM_{2.5}$ formation and total reactive nitrogen (Nr) deposition. Several long-term monitoring networks have measured components of Nr species for several decades (see Table 1).

While monitoring data are used to assess regional long-term trends in air concentrations[1] and wet deposition of some Nr species,[2] the NADP Total Deposition (TDep) measurement-model fusion method[3] is widely used for assessing trends in total (wet + dry) Nr deposition in the United States. Briefly, the TDep method combines measured concentrations and wet deposition with modeled values where measurements are lacking (spatial gaps or unmeasured species).

In this article, we use measurements, TDep products, and emission inventories to discuss current trends in atmospheric concentrations and deposition of Nr and their relationship to trends in emissions. This analysis identifies several examples where improvements in monitoring, modeling, and emissions inventories are needed to better characterize the linkages between trends in emissions and changes in the atmospheric composition of Nr.

Current Monitoring Trends in Emissions, Ambient Concentrations, and Deposition

Annual emissions of SO_2 and NOx have decreased substantially (by 83% and 57%, respectively) from the period from 1990–1992 to 2014–2016 (see Figure 1). This is attributable to EGU controls (i.e., EGU emission reductions of 85% for SO_2 and 77% for NOx),[4] market-driven changes to cleaner fuels, and mobile source controls (i.e., mobile source reductions of 89% for SO_2 and 46% in NOx).[4]

This decline is reflected in the long-term monitoring of ambient concentrations over the same period. The decreasing SO_2 concentration trend measured at eastern CASTNET sites (86%; see Figure 2 and summarized in Table 2) shows a linear relationship between EGU emissions and ambient concentrations (R^2= 99%).[5] Data that support linkages between emissions and environmental results provide accountability for regulators and the regulated community.

Deriving this type of relationship between emissions and concentrations is more convoluted for Nr species. Atmospheric processing converts the NOx emitted by sources (reported by emissions monitors) to a diverse number of oxidized N compounds (NOy), which monitoring networks either measure as total NOy (by chemical conversion of all NOy compounds to NO prior to detection) or as a fraction of NOy (e.g., filter-based methods report out on total nitrate [the sum of nitric acid (HNO_3) and particulate nitrate (pNO_3]). Also, NOx emissions are more distributed across source types (e.g., 26% EGUs, 52% transportation, 22% other in 1990)[4] (Figure 1). Large decreases in NOx emissions during the period from 1990–1992 to 2014–2016 are reflected in a

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marked decrease in ambient concentrations of total nitrate (48%) at CASTNET eastern reference sites (Figure 2), and in NO_2 satellite observations.[6]

For NHx species, emission sources emit NH_3, which in the atmosphere can readily convert to pNH_4 or remain as NH_3 depending on meteorological conditions and availability of acidic pollutants as precursors to pNH_4. Monitoring networks need to measure both forms to accurately represent NHx. CASTNET ambient pNH_4 concentrations show a similar

Table 1. Existing U.S. monitoring networks that measure components of reactive nitrogen (Nr) in the atmosphere or precipitation (wet deposition). Data from these networks are used by state, local, and federal agencies; researchers; and industries to assess trends in atmospheric pollution and deposition.

Network	Nr Measurements	Measurement Interval	Website
Clean Air Status and Trends Network (CASTNET)	Ambient concentrations of pNH_4^+, particulate nitrate (pNO_3^-), nitric acid (HNO_3)	Weekly	https://epa.gov/castnet
National Atmospheric Deposition Program (NADP) National Trends Network (NTN)	Concentrations of NO_3- and NH_4^+ in precipitation; precipitation amounts	Weekly	http://nadp.slh.wisc.edu/NTN/
NADP's Atmospheric Integrated Research Monitoring Network (AIRMoN)	Concentrations of NO_3^- and NH_4^+ in precipitation; precipitation amounts	Daily (event-based)	http://nadp.slh.wisc.edu/AIRMoN/
NADP's Ammonia Monitoring Network (AMoN)	Ambient concentrations of NH_3	Bi-weekly	http://nadp.slh.wisc.edu/AMoN/
Chemical Speciation Network (CSN)	Ambient concentrations of pNO_3^-, pNH_4^+	Daily (1:3 or 1:6 day)	https://www3.epa.gov/ttn/amtic/speciepg.html
Interagency Monitoring of Protected Visual Environments (IMPROVE)	Ambient concentrations of pNO_3^-, particulate nitrite (pNO_2^-)	Daily (1:3 day)	http://vista.cira.coloradostate.edu/improve
National Core (NCore) Multipollutant Network; State and Local Air Monitoring Stations (SLAMS); National Air Monitoring Stations (NAMS)	Concentrations of NO, total oxidized nitrogen (NOy); PM speciation (CSN or IMPROVE)	Hourly	https://www3.epa.gov/ttn/amtic/ncore; https://www3.epa.gov/airquality/montring.html
Photochemical Assessment Monitoring Stations (PAMS)	Concentrations of NO, NOy, NOx	Hourly	https://www3.epa.gov/ttn/amtic/pamsmain.html
Near-road NO_2 Monitoring	Concentrations of NO_2	Hourly	https://www3.epa.gov/ttnamti1/nearroad.html
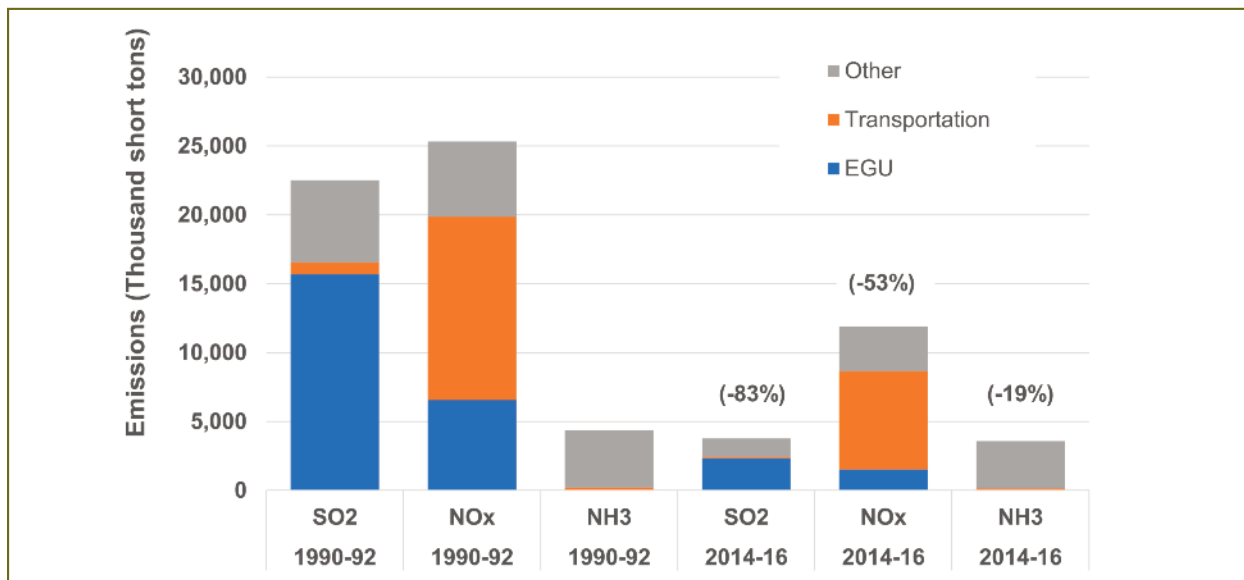
Figure 1. Three-year averaged annual emissions trends for SO_2, NOx, and NH_3 by source category.[4]
Note: Source categories are grouped as regulated EGUs, transportation, and other, which includes everything from fuel combustion from non-EGU sources, industrial processes, and agricultural sources to events such as wildfires and prescribed burns. The percent decrease in overall emissions is noted above the 2014-16 bars.

decreasing trend (63%) as those reported for SO_2 and total nitrate (Figure 2), yet NH_2 emissions have decreased at a much slower rate (-19%) since 1990–1992.[4] Measured annual ambient NH_3 concentrations at 21 NADP/AMoN sites with long-term sampling records increased 24% from 2008–2010 to 2014–2016 (see Figure 3). An increasing NH_2 trend (7 ± 2%) was also identified in a study on similar sites that accounted for variability in seasonality and regional location.[8]

Trends in total (wet + dry) deposition in the United States were derived from TDep results and should be reflective of those for emissions and ambient concentrations. Total Sulfur

(S) deposition decreased by 58% from 2000–2002 to 2014–2016 and total NOy deposition decreased by 35% over the same period, showing significant but less dramatic trends than measured concentrations (see Figure 4; summarized in Table 2). However, total NHx, deposition increased by 30% over the same time period, and comprised a decrease in dry pNH_4 deposition (-17%) and increases in wet NH_4^+ deposition (+24%) and dry NH_3 deposition (+54%), which contribute 2%, 30%, and 18% to the total Nr budget, respectively (Figure 4).

Ambient NH_3 can be entrained in precipitation, thus higher

Table 2. Summary of percent differences for oxidized sulfur, oxidized nitrogen, and reduced nitrogen in emissions, concentrations, and total deposition over different time periods of comparison. All percent differences are obtained from three-year averages at the beginning and end of the time period as indicated.

Species	Time Period	Emissions	Concentrations	Total Deposition
SO_2	1990–92 to 2014–16	-83	-86	-
	2000–02 to 2014–16	-76	-80	-58◇
NOy	1990–92 to 2014–16	-53ζ	-48¥	-
	2000–02 to 2014–16	-48ζ	-48¥	-35
NHx	1990–92 to 2014–16	-19†	-63‡	-
	2000–02 to 2014–16	-15†	-58‡	30
	2008–10 to 2014–16	-17†	-39‡; 24†; 7 ± 2†δ	19

Notes:
◇- reported as total S; ζ - reported as NOx; ¥ - reported as total nitrate;
† - reported as NH_3; ‡ - reported as pNH_4^+; δ – Ref.: Butler et al., 2016
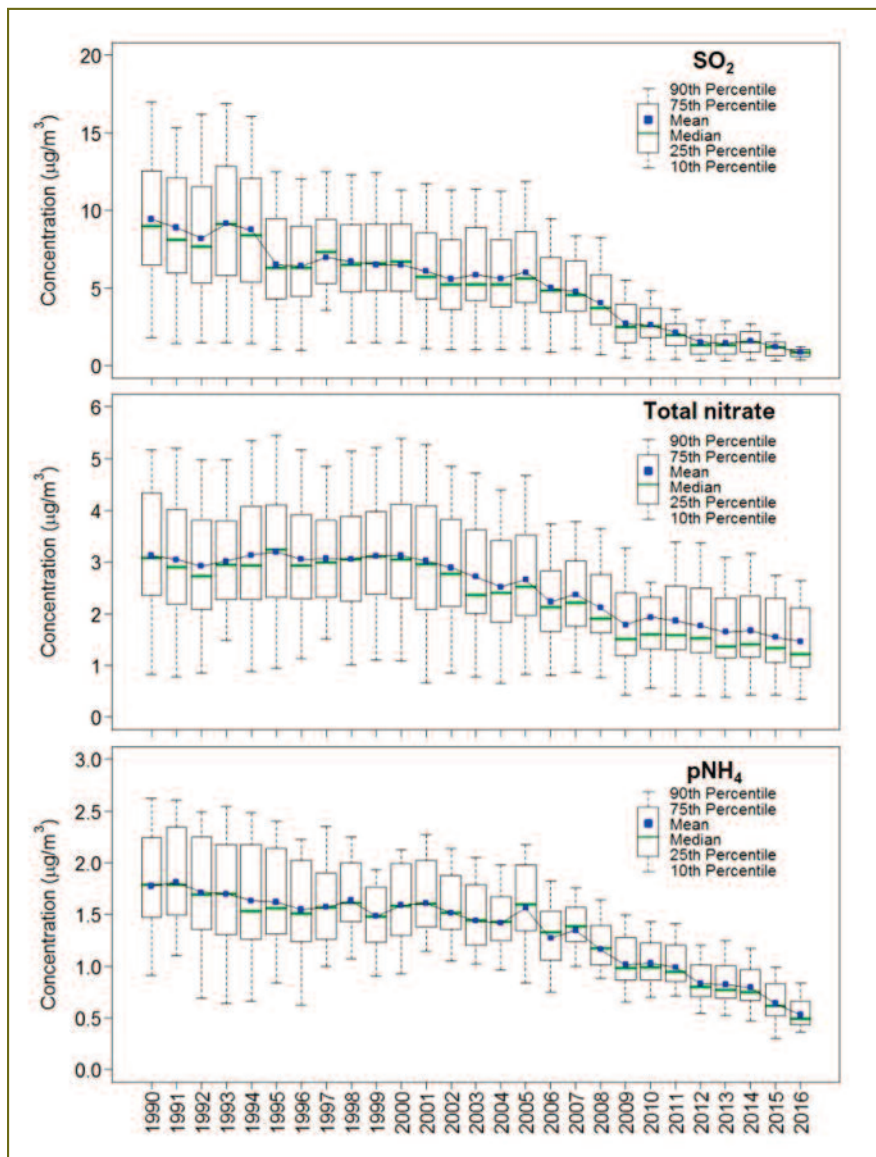
Figure 2. Trends in annual aggregate mean SO_2 (top), total nitrate (middle), and pNH_4 (bottom) concentrations from CASTNET eastern reference sites.[7]
Note: The CASTNET reference sites are split into eastern and western regions due to the spatial density of the measurement sites, concentration differences, a difference in filterpack collection flow rate, and different start dates of operation. Only eastern sites are discussed as they are in closer proximity to EGUs and more reflective of the trends.

NHx concentration trend (-4%), which is more proximate to the National Emissions Inventory (NEI) NH_3 emissions trend of -17% over this period, though still a substantial difference.

The TDep NOy total deposition maps from 2000–2002 to 2014–2016 show that the reductions in NOy deposition have been significant downwind of large EGU sources in the Eastern United States (see Figure 5). Urban areas are now easily identified as the major NOy hotspots. The total NHx deposition map shows increases in agricultural source regions (e.g., Midwest United States, eastern NC, southeastern PA) (Figure 5). The total Nr deposition predicted by the TDep method is now approximately half NOy and half NHx (Figure 4). This trend has also been observed in other studies.[2,10]

Trends Analysis Limitations

The trends analyses in Nr emissions, concentrations, and deposition described in the previous section are not without limitations. Linking trends in emissions and atmospheric concentrations for Nr species is not as straightforward as for S, as there are more Nr species, more reactivity, and more sources to convolute these linkages. Emissions inventories for non-EGU sources are not robust and improvements are needed

NH_3 concentrations likely explain the observed increases in wet NH_4^+ deposition. The decreasing pNH_4 concentrations and the increasing NH_3 concentrations suggest that less of the NH_3 emissions are partitioning to the particle phase. This is supported by the concurrent decline in SO_2 and NOy emissions and concentrations, which reduces the potential for acidic pollutants to react with gaseous NH_3 and convert to PM.[8] Summing the concentration averages for pNH_4 and NH_3 over the period from 2008–2010 to 2014–2016 and calculating the difference provides a rough estimate of the

for all Nr species. These sources have greater uncertainties, as they are more variable with time (e.g., agricultural and biogenic sources), are episodic (e.g., wildfires), and are typically calculated via mass-balance techniques.[11] Studies suggest current inventories for mobile emissions are overestimated for NOx[12-14] and underestimated for NH_3.[15,16] Agricultural sources (e.g., livestock production, emissions from fertilized soils) account for 80% of U.S. NH_3 emissions[4,17,18] and are poorly characterized by agricultural practice and activity data in emissions inventory development.[19]
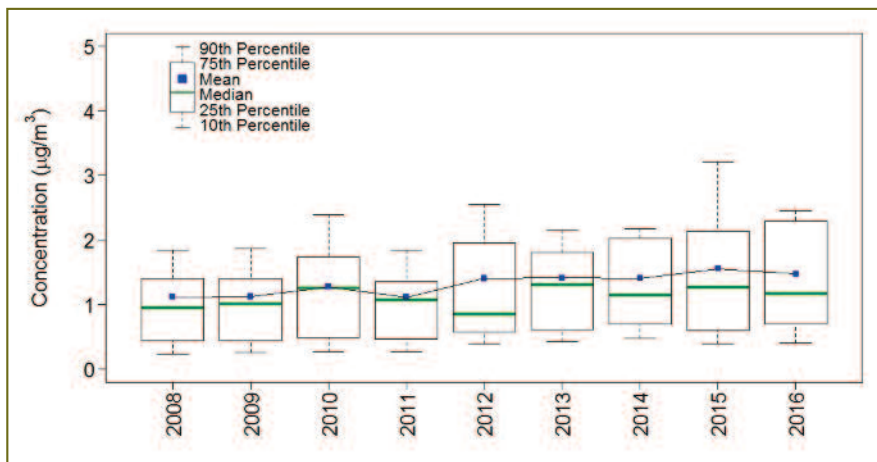
Figure 3. Trend in annual aggregate mean NH$_3$ concentrations from 21 NADP/AMoN sites.
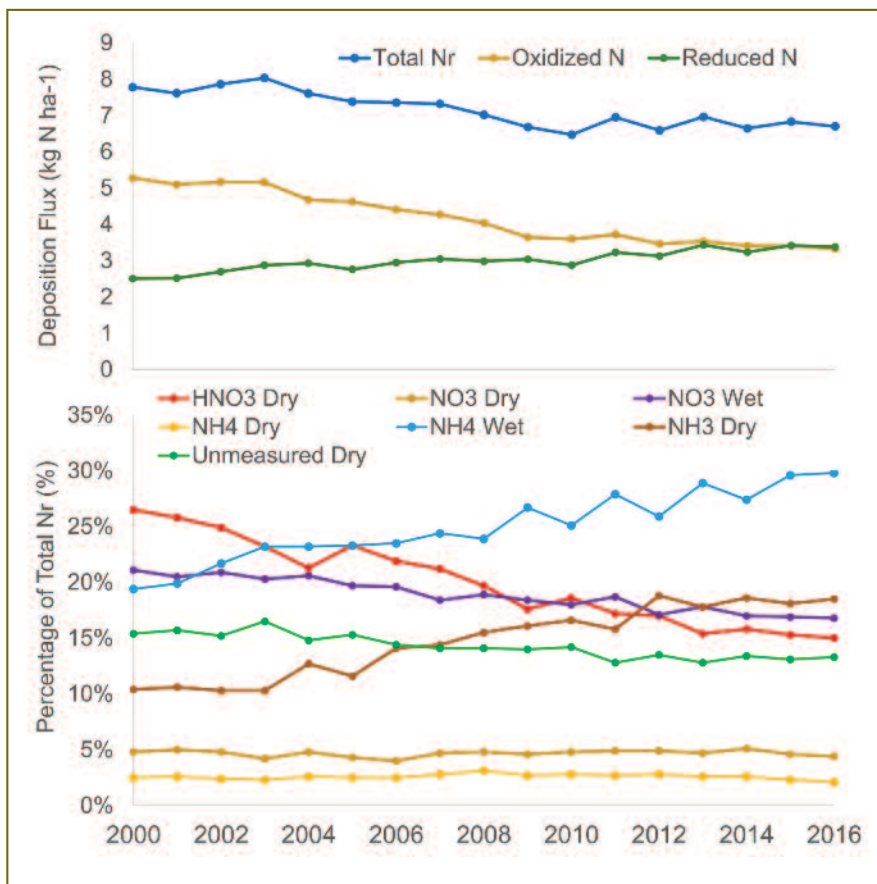
Figure 4. Trends in Nr deposition output by the TDep measurement-model fusion method.
Note: Top plot is the deposition flux of total Nr and its oxidized and reduced components (kg-N ha^{-1}). The lower plot is the percentage of total Nr deposition for each modeled species and its deposition pathway.

relationship with the modeled bi-directional deposition velocities. Also, existing network measurements for NO$_2$ (e.g., U.S. AQS), are also not currently utilized. Planned newer versions of the TDep method will address these limitations in the near future. Approximately 13% of the total Nr deposition budget is either not measured or not utilized by the TDep method (Figure 4). A fraction of this is organic nitrogen (ON), which is uncharacterized.

More Research Needed

Routine Nr monitoring could be expanded to include bulk sampling of ON in precipitation and PM to develop more complete Nr budgets. Additionally, low-cost passive samplers for NH$_3$ and NO$_2$ could be added to existing networks to help characterize gradients from urban and agricultural areas to rural, non-source impacted areas. This could be conducted in tandem with satellite assessments to identify new monitoring locations and to better understand measurement spatial representativeness. Further development of low-cost methods for directly measuring dry deposition, suitable for routine network operation, is also a high priority. Finally, there are constant improvements in the accuracy of chemical transport models (CTMs) used to develop long-term time series of concentrations and deposition. These new estimates need to be reconciled with older estimates, especially for trends assessment where consistency is essential.

Satellite measurements of tropospheric NO$_2$ and NH$_3$ concentrations can augment current monitoring and modeling strategies for Nr and address some of these limitations. Satellite data products have been used to quantify regional and point-source scale emissions,[20,21] including episodic emissions (e.g., wildfires)[22,23] to improve emissions inventories.[24,25] Also,

There are substantial limitations to the available Nr concentration measurements and how those measurements are used to assess total deposition. The TDep methodology does not utilize measured NH$_3$ concentrations because of a non-linear
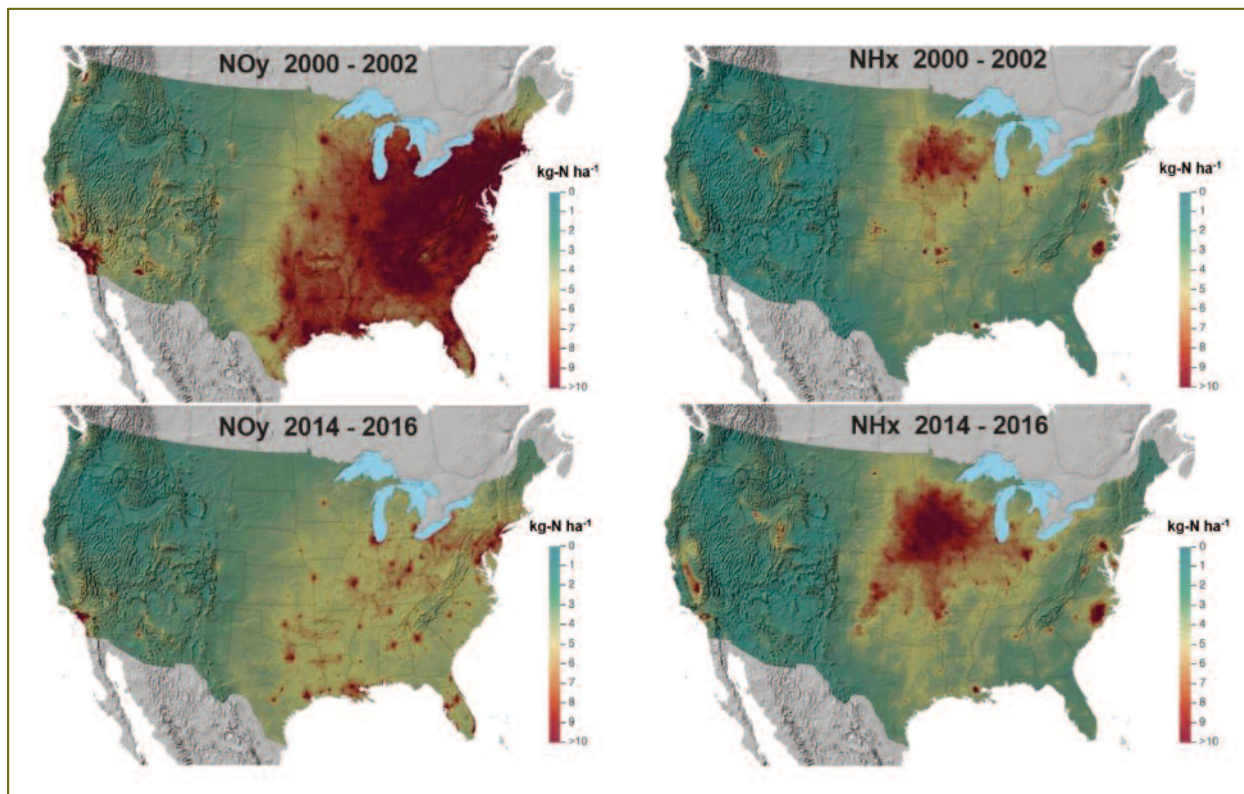
Figure 5. TDep method deposition maps of NOy and NHx from 2000–2002 to 2014–2016.[9]

satellite-derived long-term trends for concentrations of NO_2[26,27] and NH_3[28-30] support surface monitoring trend data and provide information on spatial variability[31] not achievable with surface networks. Satellite data products have been used in conjunction with measurements, CTMs, and deposition models to estimate trends in Nr deposition[32,33] or to evaluate and improve the CTMs[28,34-36] and thus improving modeled deposition estimates providing more accurate trends. **em**

Greg M. Beachley and **Melissa A. Puchalski** are with the U.S. Environmental Protection Agency's (EPA) Office of Air and Radiation in Washington, DC; **Chris M. Rogers** is with Wood Environment & Infrastructure Solutions Inc., Jacksonville, FL; **Tom F. Lavery** is an environmental consultant based in Cranston, RI; and **John T. Walker** is with EPA's Office of Research and Development in Durham, NC. E-mail: beachley.gregory@epa.gov.

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