



Clean Air Status and Trends Network (CASTNET) site in Beaufort, NC.  
Photo courtesy of the U.S. Environmental Protection Agency (EPA).

# Need for **Improved Monitoring** of Spatial and Temporal Trends of Reduced Nitrogen

by **Melissa A. Puchalski, John T. Walker, Gregory M. Beachley, Mark A. Zondlo, Katherine B. Benedict, Richard H. Grant, Bret A. Schichtel, Christopher M. Rogers, April B. Leytem, Joann Rice, Kristi Morris, James J. Schauer, and Rui Wang**

The authors discuss needed improvements in monitoring and characterization of reduced inorganic nitrogen.

**U.S. regulations have been effective at reducing emissions** of oxidized nitrogen (NO<sub>x</sub>) and associated secondary pollutants (e.g., ozone, nitrate).<sup>1,2</sup> NO<sub>x</sub> (nitrogen oxide [NO] + nitrogen dioxide [NO<sub>2</sub>]) reductions have resulted in decreasing trends and shifting spatial patterns in wet and dry nitrate deposition as measured by urban and rural monitoring networks.<sup>3,4</sup> As NO<sub>x</sub> emissions have decreased, the atmospheric reactive nitrogen (Nr) budget is shifting toward a greater contribution from reduced inorganic nitrogen forms (NH<sub>x</sub> = ammonia gas [NH<sub>3</sub>] + ammonium aerosol [NH<sub>4</sub><sup>+</sup>]).<sup>5</sup> Unlike NO<sub>2</sub>, NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> are not regulated under the U.S. Clean Air Act; however, as a component of Nr deposition and contributor to particulate matter < 2.5 μm (PM<sub>2.5</sub>), NH<sub>x</sub> is considered in the current review of the NO<sub>x</sub>/SO<sub>x</sub>/PM secondary National Ambient Air Quality Standards (NAAQS).<sup>6</sup> While the primary NAAQS protect human health, the secondary NAAQS protect public welfare, which includes visibility and ecosystem health.

Livestock waste and fertilized soils account for approximately 80% of NH<sub>3</sub> emissions in the United States.<sup>7</sup> Automobiles (urban) and wildfires are also important sources of NH<sub>3</sub> and are likely underestimated in current inventories.<sup>4,8-10</sup> In the atmosphere, NH<sub>3</sub> reacts with acidic pollutants to form particulates (e.g., ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>], ammonium bisulfate [(NH<sub>4</sub>)HSO<sub>4</sub>], ammonium nitrate [NH<sub>4</sub>NO<sub>3</sub>]), which contribute to PM<sub>2.5</sub>, climate forcing, and poor visibility.<sup>11</sup> NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> are also components of Nr deposition, which can cause soil acidification,<sup>12</sup> NO and nitrous oxide (N<sub>2</sub>O) emissions from microbial activity in soils,<sup>13,14</sup> reductions in vegetation species diversity,<sup>15</sup> and eutrophication of aquatic ecosystems.<sup>16</sup> Measuring and modeling NH<sub>3</sub> dry deposition is challenging due to the presence of a surface compensation point and subsequent bidirectional exchange with the atmosphere.<sup>17,18</sup>

Ambient NH<sub>4</sub><sup>+</sup> has been monitored for more than 30 years by existing networks, but less information exists for gaseous NH<sub>3</sub>. Over the last decade, ground-based monitoring and remote sensing have shown NH<sub>3</sub> to be increasing in some regions of the United States,<sup>19,20</sup> yet uncertainties in emission inventories and gaps in monitoring make linking long-term trends in NH<sub>3</sub> emissions with ambient NH<sub>x</sub> difficult.<sup>21</sup> Expanded monitoring of NH<sub>x</sub> is needed to better characterize these linkages and provide data to evaluate and improve chemical transport models (CTMs) to more accurately predict PM formation, visibility impacts, and ecosystem exposure. Better understanding of spatial variability of NH<sub>3</sub> concentrations and bidirectional exchange is also needed to improve the representation of NH<sub>3</sub> in measurement-model fusion techniques for total deposition.<sup>22</sup>

### Current Monitoring of NH<sub>x</sub>

Particulate NH<sub>4</sub><sup>+</sup> has been measured by atmospheric

monitoring networks in the United States for more than 30 years. None of the existing networks measure total NH<sub>x</sub>, but data can be combined to develop estimates of reduced atmospheric nitrogen. The key U.S. networks that measure components of NH<sub>x</sub> include:

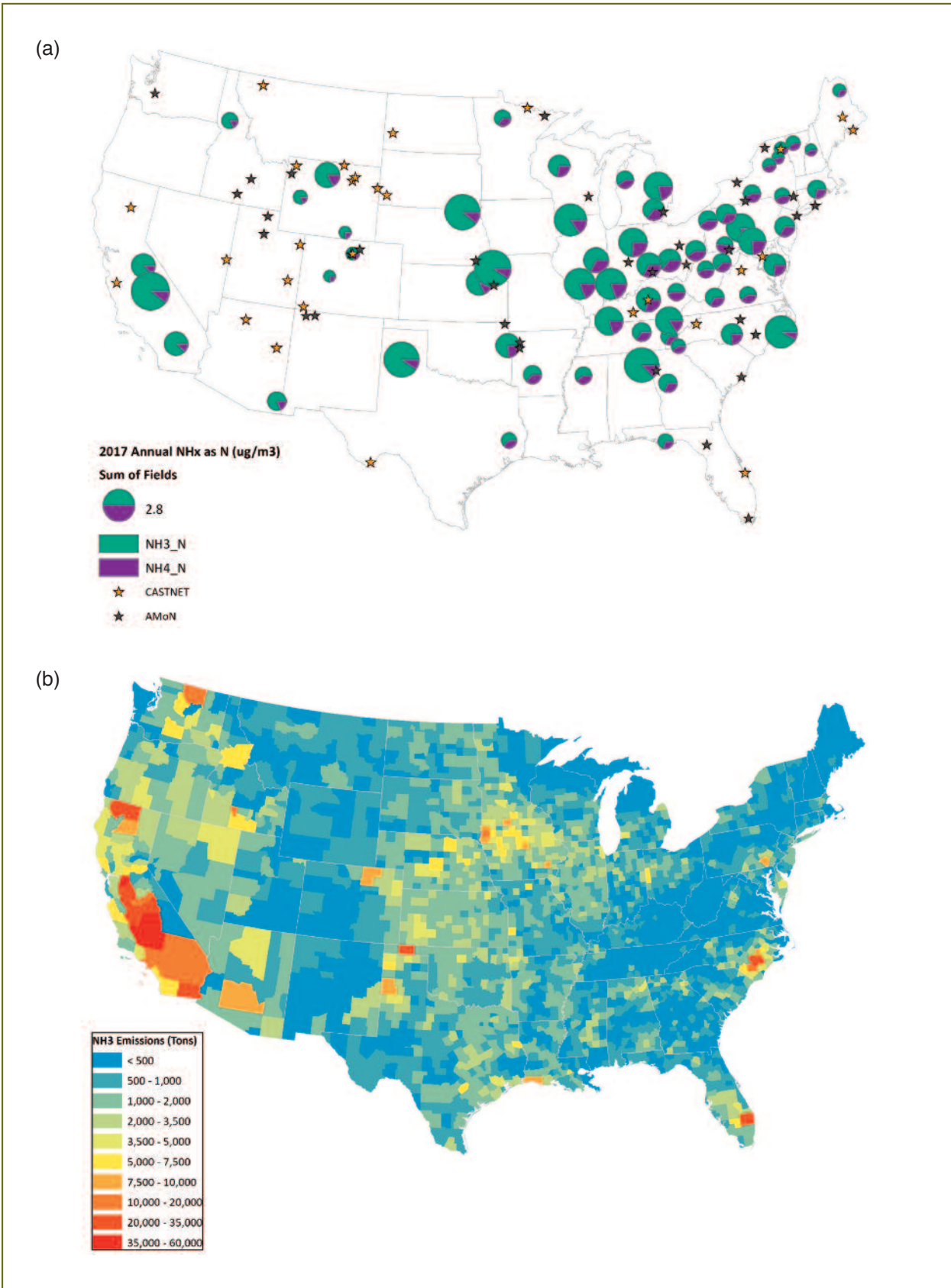
- The **Clean Air Status and Trends Network (CASTNET)** measures weekly concentrations of particulate NH<sub>4</sub><sup>+</sup>, as well as nitric acid (HNO<sub>3</sub>), sulfur dioxide (SO<sub>2</sub>), nitrate (NO<sub>3</sub><sup>-</sup>), and sulfate (SO<sub>4</sub><sup>2-</sup>), at 92 sites. CASTNET data have provided estimates of dry deposition used to assess spatial and long-term trends since 1990.
- **National Atmospheric Deposition Program's (NADP) Ammonia Monitoring Network (AMoN)** uses passive samplers to measure two-week integrated NH<sub>3</sub> concentrations. Established in 2007, the network includes 105 sites.
- The **Chemical Speciation Network (CSN) and Interagency Monitoring of Protected Visual Environments (IMPROVE)** networks collect speciated 24-hour PM<sub>2.5</sub> samples on a one-in-three-day schedule. CSN samples are analyzed for NH<sub>4</sub><sup>+</sup>.<sup>23</sup> In IMPROVE, NH<sub>4</sub><sup>+</sup> is derived from assumed compositions of the measured SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>.<sup>24</sup>

In the case of both CASTNET and CSN, NH<sub>4</sub><sup>+</sup> concentrations are biased low due to losses of NH<sub>4</sub><sup>+</sup> from dissociation of particulate nitrate from the Teflon and nylon filters, respectively.<sup>25,26</sup> While the NH<sub>4</sub><sup>+</sup> concentrations are comparable, IMPROVE and CSN use a PM<sub>2.5</sub> cutpoint and CASTNET uses an open face filter pack.<sup>27</sup>

The spatial distribution of ambient measurements of NH<sub>x</sub>, calculated as the sum of NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> at co-located AMoN/CASTNET sites, is presented in Figure 1a, showing a predominance of NH<sub>3</sub> at all locations. The majority of co-located AMoN/CASTNET sites are located in the eastern United States, many in counties with relatively low NH<sub>3</sub> emissions. Some hotspots are captured (i.e., eastern NC); however, large areas of agricultural NH<sub>3</sub> emissions across the Midwest and West are not well resolved by the existing networks, which can be seen by the discrepancy between Figure 1a and 1b. Outside of agricultural regions, monitoring is lacking in the Northwest, Gulf Coast region, and Florida.

### Expansion of Routine Monitoring

Establishing additional co-located time-integrated NH<sub>4</sub><sup>+</sup> and NH<sub>3</sub> sites would be a cost-effective way to better resolve broad spatial and temporal patterns of NH<sub>x</sub>. Satellite measurements and NH<sub>3</sub> inventories are useful for identifying high-value locations for establishing new sites where monitoring is currently lacking. Figure 2 shows AMoN sites layered with the 2008–2016 average satellite NH<sub>3</sub> measurements,



**Figure 1.** (a) Annual NHx measurements across the continental United States from CASTNET and AMoN in 2017; (b) map of 2014 NEI NH<sub>3</sub> emissions for all sectors.<sup>7</sup>

*Note:* (a) Pie charts represent total NHx as the sum of particulate NH<sub>4</sub><sup>+</sup> (measured by CASTNET) and NH<sub>3</sub> (measured by AMoN). The size of the circle is representative of the total NHx concentration. Site locations that are not co-located are represented by a star.



highlighting monitoring gaps in high emission regions and difficulties in selecting representative sites. For example, the highest AMoN  $\text{NH}_3$  concentration (2016–2018 average  $16.8 \mu\text{g}/\text{m}^3$ ) is measured at UT01 (Cache Valley) located adjacent to a feedlot. This site is representative of the Cache Valley, but not representative of the northern UT region. The highest ground-level concentrations may not be well represented in the satellite observations due to a lack of vertical mixing, which illustrates the synergies between satellite and ground-based measurements.

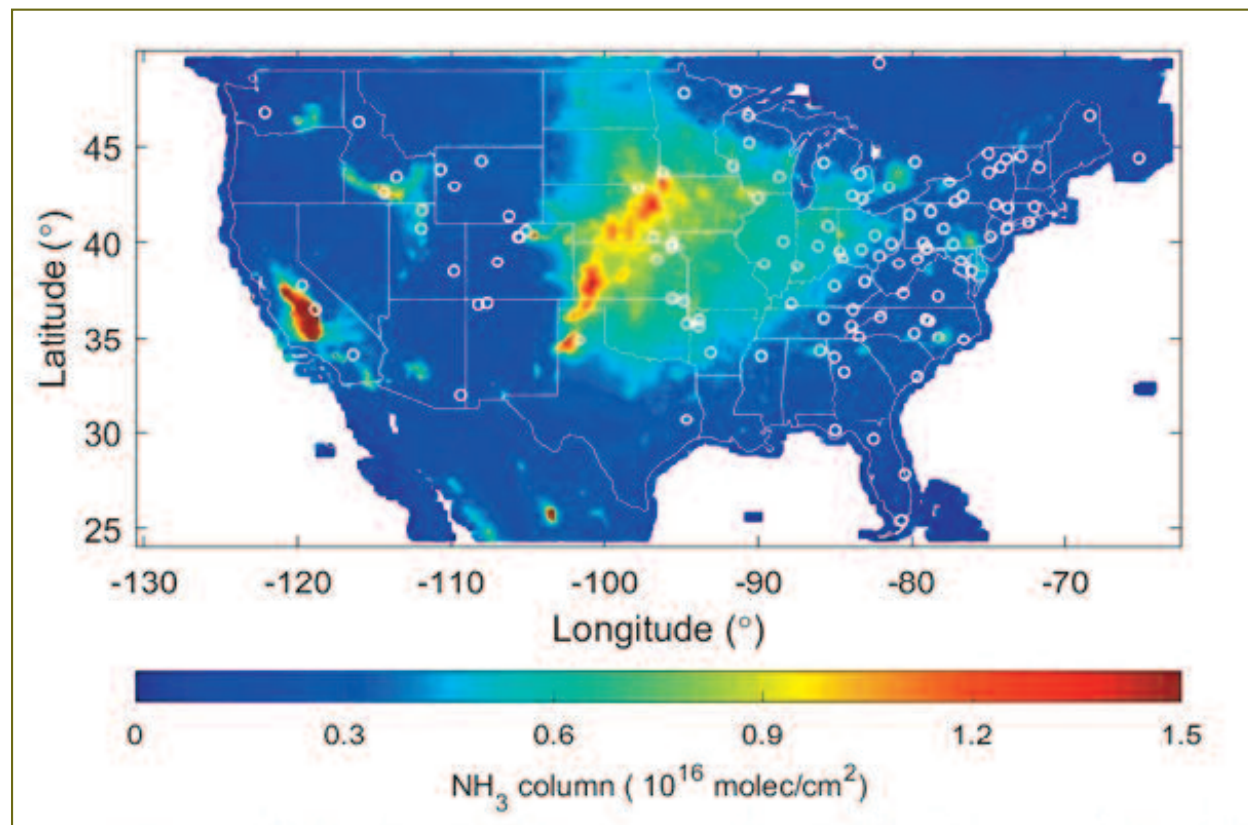
Characterization of patterns across smaller source regions and land-use types is also needed. As an example, a regional passive  $\text{NH}_3$  monitoring network has been conducted during the warmest months since 2010 in Colorado.<sup>31,32</sup> This network provides data on  $\text{NH}_3$  gradients across an agricultural–rural–suburban–urban region with concentrations ranging from less than  $5 \mu\text{g m}^{-3}$  in rural–suburban locations to  $42 \mu\text{g m}^{-3}$  near large feedlots,<sup>31</sup> highlighting some of the gaps in existing national networks where the spatial gradients are not well represented.

CSN and IMPROVE offer additional opportunity for expanded rural and urban monitoring. Capturing the daily integrated  $\text{NH}_x$  concentrations with speciated  $\text{PM}_{2.5}$

measurements would be beneficial for model evaluation and source apportionment. Acid-impregnated filters deployed in a pilot IMPROVE study in the West and Midwest showed good recovery of  $\text{NH}_x$ .<sup>33</sup> However, recent testing of acid-impregnated filters in CSN and IMPROVE deployed in the Southeast exhibited a negative bias in CSN as compared to the reference method; therefore, additional testing in humid environments is needed. If biases in the technique can be adequately understood, deployment of the acid-impregnated filters at existing sites could provide additional integrated  $\text{NH}_x$  concentrations in urban and rural environments at more than 300 sites for a relatively low cost.

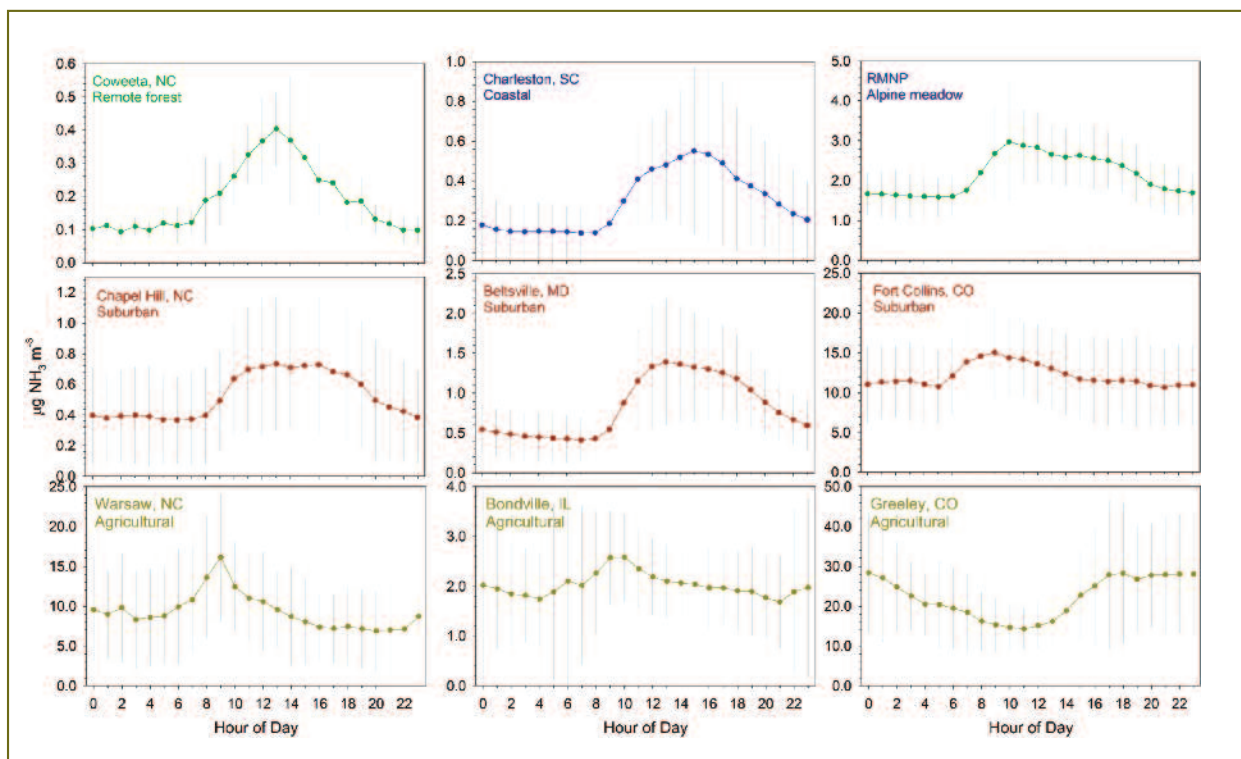
### High Time Resolution Measurements

Modeling the diurnal  $\text{NH}_3$  concentration profile in CTMs is important to properly simulate PM formation and atmospheric deposition processes, but is difficult due to uncertainties in emission inventories, boundary layer dynamics, local dispersion, and bi-directional fluxes.<sup>17,34,35</sup> Figure 3 summarizes diurnal  $\text{NH}_3$  concentration profiles in several locations, highlighting the need for high-time resolution measurements in different environments. Profiles in agricultural areas (e.g., crops and concentrated animal feeding operations regions) can differ substantially from profiles measured in forested, coastal, and suburban locations. Patterns similar to the examples in Figure



**Figure 2.** Average (2008–2016) satellite  $\text{NH}_3$  measurements from the Infrared Atmospheric Sounding Instruments (IASI v2.2R) using an oversampling algorithm for high-spatial resolution.<sup>28–30</sup>

Note: AMoN sites are noted by open circles.



**Figure 3.** Diurnal concentration profiles of  $\text{NH}_3$  at suburban, forested, coastal, and agricultural sites during summer months.

*Note:* Concentrations were measured using high temperature conversion/chemiluminescence (Warsaw, NC),<sup>18</sup> the Monitor for Aerosols and Gases (MARGA, Coweeta, NC; Chapel Hill, NC; Charleston, SC; Bondville, IL; Beltsville, MD),<sup>39,40</sup> Picarro ( $\text{NH}_3$ ) analyzer (Rocky Mountain National Park [RMNP]); or a Particle Measuring System (Air Sentry II, 129 Fort Collins, CO; Greeley, CO).<sup>41</sup> Data represent mean hourly concentration  $\pm$  1 standard deviation.

3 have been observed at other locations in the United States.<sup>36-38</sup>

The diurnal concentration patterns in Figure 3 may not correlate directly to  $\text{NH}_3$  emission rates, but rather reflect the net result of emissions, deposition, and atmospheric processes. Time-resolved monitoring of air concentrations is not a substitute for direct emission measurements, which are also needed to improve current inventories.

Methods for continuous  $\text{NH}_3$  measurements include cavity ring-down, ion mobility, and quantum cascade laser spectroscopy, high temperature conversion with nitric oxide chemiluminescence, online wet denuder/conductivity techniques, chemical ionization mass spectrometry, and others.<sup>42</sup> As an option for total  $\text{NH}_x$ , CASTNET designed an enhanced chemiluminescence  $\text{NO}/\text{NO}_y/\text{total N}_r$  analyzer (nitrotrain). Hourly concentrations of  $\text{NH}_x$  are reported by subtracting  $\text{NO}_y$  from total  $\text{N}_r$ . The nitrotrain is currently deployed at Duke Forest, NC, in a routine-network mode. Deployment in other networks (e.g., NCore Network) could utilize existing infrastructure to measure hourly  $\text{NH}_x$  concentrations at 80+ urban and rural monitoring sites. Adsorption onto inlet and tubing surfaces, as well as particle filtration,<sup>42</sup> continue to present

significant challenges for deployment of continuous  $\text{NH}_3$  methods in a routine monitoring framework.

## Conclusions

Improved characterization of  $\text{NH}_x$  will support the development of effective environmental regulations, guide the development of best management practices for emissions, and improve CTMs. A combination of time-integrated sampling and high-time resolution measurements are needed to better characterize spatial gradients, long-term trends and atmospheric processes. Ideally, any expansion of NADP's AMoN would be accompanied by  $\text{NH}_4^+$  measurements, trace acidic pollutants, and meteorology to characterize gas-particle interactions and improve models used to assess  $\text{PM}_{2.5}$  reduction strategies. Selecting new site locations should be informed by remote sensing and emission inventory data. Expanding  $\text{NH}_x$  monitoring presents an opportunity for greater engagement of agricultural, ecological, and atmospheric communities in the assessment of  $\text{NH}_x$  and for cooperation between federal, state, or local agencies, universities, tribes, or other private organizations.

Improvements in emission inventories are also urgently needed, particularly for agriculture. In addition to the emission

factors and emission models themselves, improvements in the activity data underlying the inventory are needed. For example, information on number of animals, facility characteristics and manure management, locations of emission sources, and timing and amount of fertilizer application are often lacking or inaccurate. A targeted approach to low-cost, time-integrated

NH<sub>x</sub> measurements and high resolution, multi-pollutant intensive field studies is critical for the refinement of NH<sub>3</sub> emission inventories, improved model performance and their application to developing meaningful air quality management strategies. **em**

**Melissa A. Puchalski** and **Greg M. Beachley** are with the U.S. Environmental Protection Agency's (EPA) Office of Air and Radiation in Washington, DC; **John T. Walker** is with EPA's Office of Research and Development in Durham, NC; **Mark A. Zondlo** and **Rui Wang** are with the Department of Civil and Environmental Engineering at Princeton University, Princeton, NJ; **Katherine B. Benedict** is with the Department of Atmospheric Science at Colorado State University, Fort Collins, CO; **Richard H. Grant** is with the Department of Agronomy at Purdue University, West Lafayette, IN; **Bret A. Schichtel** is with the U.S. National Park Service, Cooperative Institute for Research in the Atmosphere at Colorado State University, Fort Collins, CO; **Christopher M. Rogers** is with Wood Environment & Infrastructure Solutions Inc. in Newberry, FL; **April B. Leytem** is with the U.S. Department of Agriculture, Agricultural Research Service in Kimberly, ID; **Joann Rice** is with EPA's Office of Air Quality Planning and Standards in Durham, NC; **Kristi Morris** is with the U.S. National Park Service, Air Resources Division in Lakewood, CO; and **James J. Schauer** is with the Wisconsin State Laboratory of Hygiene at the University of Wisconsin-Madison, Madison, WI. E-mail: puchalski.melissa@epa.gov.

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