The Pressing Need for Reactive Nitrogen Measurements for Space

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Reactive nitrogen (Nr) has significant impacts on ecosystems, air quality, and climate; it is thus a pollutant of critical concern for several reasons. Deposition of Nr in the U.S. has been found to have causal impacts on acidification and N enrichment, leading to eutrophication, alterations of primary productivity, algal blooms, and loss of biodiversity. However, existing national ambient air quality standards have been shown to inadequately protect ecosystem welfare against these threats, even in regions where Nr levels are declining owing to reductions in NO_x emissions from transportation and power generation. Simultaneously, the sensitivity of Nr deposition to ammonia (NH_3) emissions is increasing, as are NH_3 emissions themselves. Meanwhile, efforts to regulate Nr deposition have been stymied by uncertainties in Nr sources and in the relationship between emissions and deposition as described by air quality models, largely owing to lack of measurement constraints on key model species and processes. In addition to the impacts of Nr deposition on ecosystems, Nr species influence air quality and climate through their impacts on aerosols and direct and indirect impacts on greenhouse gases such as N_2O and O_3. Nr mitigation approaches must be balanced with the benefits of anthropogenic enhancement of nitrogen fixation, which is essential for crop production and food security to support the world's current and future population. Here we make the case that a geostationary satellite mission for North America over the next decade is critical for addressing ecosystems, food security, air quality, and climate management needs for reactive nitrogen.

1.What are the key challenges or questions for Earth System Science across the spectrum of basic research, applied research, applications, and/or operations in the coming decade?

A burgeoning challenge for air quality control is figuring out how to mitigate Nr deposition. Many national parks and federally protected areas in the U.S. are currently subject to hazardous levels of Nr deposition. When deposited in excess (i.e., above critical loads), Nr can lead to soil acidification, eutrophication of surface water, and nutrient imbalances in sensitive ecosystems. Despite likely continued decreases in NO_x emissions from transportation and power plants in the U.S. in coming decades, critical load exceedances are estimated to persist in 2050 in most national parks [Ellis et al., 2013]. Exceedances may even increase in locations already exceeding critical loads such as Rocky Mountain National Park [Paulot et al., 2013]. Meanwhile, the roles of emissions of NO_x, SO_x and NH_3 in governing Nr deposition are undergoing a transitional phase, with reductions to the former two species being potentially outweighed by increases in the latter. Disentangling the relative importance of each is critical for developing effective strategies for mitigating Nr deposition. Simultaneously, application of NH_3 for agricultural practices, which is essential yet currently unregulated, can be guided by regional voluntary programs for best management practices as well as enhancements to alert systems

which are currently based only on meteorological forecasts (http://www.rmwarningsystem.com, https://www.colorado.gov/pacific/sites/default/files/AP_PO_Appendix-V-Options-to-Reduce-Ammonia-Emissions.pdf).

Reactive nitrogen species also impact air quality. NO_x and NH_3 directly contribute to fine particulate matter concentrations (i.e., $PM_{2.5}$) through formation of ammonium sulfate and ammonium nitrate. These species constitute approximately half of the $PM_{2.5}$ concentrations in the U.S., which are responsible for more than 100,000 premature deaths per year [Fann et al., 2012]; NO_x also contributes to health concerns via O_3. The external air pollution damages of emissions of NO_x and NH_3 owing to premature deaths are estimated to be tens to hundreds of thousands of dollars per year per kg of N emitted, depending upon location and season [Fann et al., 2009; Dedoussi and Barrett, 2014; Paulot and Jacob, 2014; Buonocore et al., 2014].

Climate forcing of greenhouse gases and aerosols are also impacted by Nr. The direct preindustrial to present radiative forcing of ammonium nitrate aerosol has been estimated to be -0.10 ± 0.04 W m^{-2} [Myhre et al., 2013] to -0.19 ± 0.18 W m^{-2} [Shindell et al., 2013]. The presence of NH_3 can impact the thermodynamic state of secondary inorganic aerosols, which can modulate aerosol direct radiative forcing [Martin et al., 2004; Wang et al., 2008] as well as indirect forcing through clouds [e.g., Abbatt et al., 2006; Sorooshian et al., 2008]. NH_3 has also been shown to promote the formation of absorbing organic aerosol compounds that exert a positive radiative forcing [e.g., Bones et al., 2010; Updyke et al., 2012]. Additionally, N_2O is a potent greenhouse gas, with a 100-year global warming potential nearly 300 times stronger than CO_2 [Myhre et al., 2013], while NO_x emissions contribute to the formation of O_3, which is itself a greenhouse gas and criteria pollutant. Lastly, deposition of Nr may alter the carbon cycle by lowering biogenic CO_2 emissions [Pinder et al., 2012, 2013].

Despite this vast array of important issues associated with Nr, our understanding of the sources and distributions of Nr is severely limited. National emissions inventories of NOx may have uncertainties of 100% or more [Travis et al., 2015]. Inventories of NH_3 are even less certain, with constraints from bottom-up versus top-down approaches differing by factors of 2-5 in specific seasons and locations [Zhu et al., 2013; Paulot et al., 2014; Nowak et al., 2012]. Annual N_2O budgets are better constrained globally, given its well-mixed distribution, but can contain regional uncertainties up to 25% owing to seasonal variability and stratosphere-troposphere exchange [e.g., Wells et al., 2015]. Constraining sources and distributions of Nr is thus an outstanding challenge.

2. Why are these challenge/questions timely to address now especially with respect to readiness?

To understand the burgeoning nature of Nr as a pollutant, it is worth considering the status of current U.S. legislation related to Nr. Primary NAAQS for NO_x and $PM_{2.5}$ as criteria pollutants are already in place to mitigate ambient air pollution. The EPA has considered formation of air quality standards to address the effects of reactive nitrogen deposition, including aquatic and terrestrial acidification and nutrient enrichment. These new standards would be in the form of secondary standards on SO_x and NO_x emissions, and air quality models would be used to relate these to Nr deposition. As part of the EPA process of enforcing the Clean Air Act, this topic is subject to periodic review, resulting in an Integrated Science Assessment in 2008 [EPA, 2008]; a

Clean Air Scientific Advisory Committee (CASAC), an independent review board that provides advice to the EPA, also reviewed the relevant science. While causal relationships were found, both the EPA and CASAC determined that the information present at the time was insufficient to warrant formation of a new secondary standard owing to uncertainties in how Nr would be monitored and the reliability of air quality models. The observational needs have only become exacerbated by the increased complexity of the Nr sources included in such models, from the bidirectional exchange of NH_3 to online coupling of Nr soil pools with the atmosphere [e.g., Hudman et al., 2012; Bash et al., 2013; Zhu et al., 2015b]. The time is thus ripe to bring new measurements sources to bear on this problem.

3. Why are space-based observations fundamental to addressing these challenges/questions?

There is a large data gap in measuring Nr that underscores the tremendous potential for refining our understanding of Nr distributions using satellite observations. Current and planned in situ and space-born measurements are inadequate to holistically evaluate Nr as a pollutant, given their short mission lifetime, limited spatial and temporal coverage, and the lack of co-measurements of Nr species (NH_3, NO_2, and N_2O). In situ observations are currently insufficient, as highlighted in the EPA Integrated Science Assessment and CASAC reviews for development of secondary standards to address deleterious levels of Nr deposition. The EPA has routinely monitored NO_x concentrations in more than 100 locations around the country. These measurements, along with trends in remote sensing data, have helped establish the benefits of regulations that lead to cuts in NO_x emissions, with broad decreases to NO_2 evident in much of the country. However, many of the routine in situ measurements have contamination issues, and are often a complex mixture – NO_y – of various forms of nitrogen oxides. Widespread monitoring of NH_3 did not begin until 2007, with the introduction of the Ammonia Monitoring Network [AMoN, Puchalski et al., 2011], which now includes more than 50 sites. Both networks however lack the spatial and temporal coverage to describe variability in NO_x and NH_3, whose relatively short atmospheric lifetimes (~hours) leads to highly heterogeneous distributions that are difficult to completely characterize throughout the country.

In the 2018-2019 timeframe, the TEMPO instrument [Hilsenrath and Chance, 2013] will begin providing the first geostationary measurements of NO_2 over North America. Not only will TEMPO provide data at much higher spatial resolution than current satellites (2×5 km^2), the data will be available every hour, rather than once per day. However, the lifetime of this instrument is nominally only two years, which is too short to support long-term air quality management needs, and the instrument will not be able to detect NH_3 or N_2O.

The potential value of space-born measurements of NH_3 has been demonstrated in several recent studies using current low-earth-orbit platforms, yet the current instruments have several critical shortcomings in their ability to reduce our uncertainty in the sources and fates of NH_3, which would be addressed by geostationary observations [Zhu et al., 2015a]. The first remote sensing observations of lower tropospheric NH_3 were from the TES instrument [Beer et al., 2008]. This product was subsequently used to constrain NH_3 emission in the U.S. in Zhu et al. [2013], although success was limited owing to sparseness of quality TES data (a few hundred measurements per month over North America). The IASI instrument is also capable of retrieving NH_3 [Clarisse et al., 2009]. With its scanning capabilities, IASI provides much greater spatial coverage than TES; however, the spectral resolution of IASI is much lower, and given similar

instrument noise characteristics, its detection limits are higher than TES. More recently, NH_3 retrievals have been generated from the CrIS instrument [Shephard and Cady-Pereira, 2015], a scanning instrument with four times lower noise than TES or IASI, launched aboard the Suomi National Polar-orbiting Partnership in 2011. Still, these observations are limited in terms of the temporal sampling necessary to evaluate the diurnal variability of Nr sources.

Despite these advances in surface monitoring and remote sensing of Nr, the scientific needs continue to outpace the set of available measurements, which are insufficient to constrain key uncertainties in current AQ models such as the diurnal variability of Nr emissions, their coupling through soil processes and atmospheric interactions, and their response to changing levels of associated compounds such as SO_x [Zhu et al., 2015b; Bash et al., 2015]. As our air quality models of these of processes evolve, and as new air quality policies are enacted that rely on such models, there is a growing need to observe Nr at hourly and kilometer scales throughout North America that can only be addressed by new geostationary remote sensing measurements.

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