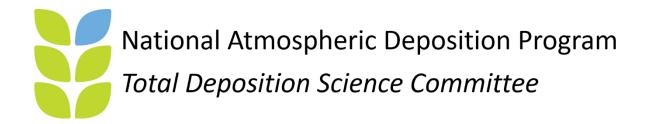
### 2019 NADP Total Deposition Science Committee Agricultural Workshop:

"Connecting stakeholder and science perspectives to better understand the linkages between agriculture and reactive nitrogen deposition"





### Contents

Workshop organizing committee	1
Background	5
Summary of key points	ô
Session 1: Modeling and source apportionment of reactive nitrogen deposition	Э
Daven Henze (UC Boulder): Source attribution of reactive nitrogen deposition using models and measurements	Э
Jesse Bash (EPA ORD): Updates and sensitivities to modeling NH <sub>3</sub> flux in the Community Multiscale Air Quality (CMAQ) Model version 5.3	
Panel Discussion: Daven Henze (UC Boulder), Jesse Bash (EPA), Viney Aneja (NCSU), Amanda Cole (ECCC), Mike Barna (NPS)	5
Session 2: Emissions of reactive nitrogen	2
Peter Adams (Carnegie Mellon University): Process-based ammonia emissions inventories from livestock – status and needs	2
Kang Sun (University at Buffalo - SUNY): Observational data-driven constraints on the emissions and lifetimes of reactive nitrogen	3
Panel Discussion: Peter Adams (CMU), Kang Sun (UaB), April Leytem (USDA ARS), Marc Houyoux (EPA OAQPS), Ian Rumsey (EPA ORD)44	
Session 3: Spatial and temporal patterns of reactive nitrogen deposition4	3
Jeff Collett (Colorado State University): Optimizing a ground-based reactive nitrogen monitoring strategy with respect to needs and resolution in a limited resource world44	3
Mark Shephard (Environment and Climate Change Canada): Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America	4
Panel Discussion: Jeff Collett (CSU), Mark Shephard (ECCC), Mark Zondlo (Princeton University), Melissa Puchalski (EPA OAP), Randy Martin (USU)60	C
Session 4: Federal stakeholders	ŝ
Peter Vadas (USDA ARS): Ammonia research in USDA-ARS: Objectives, accomplishments, and future directions	5
Jim Cheatham (NPS): Partnership to reduce nitrogen deposition impacts in Rocky Mountain National Park	3
Gail Tonnesen (EPA Region 8): Need for improved ammonia emissions and ambient monitoring data for modeling PM <sub>2.5</sub> and regional haze82	2
Daniel Cornelius (Intertribal Agricultural Council, University of Wisconsin-Madison): Native American agriculture and natural resources: Impacts relating to atmospheric deposition	ô

Panel Discussion: Peter Vadas (USDA ARS), Jim Cheatham (NPS), Gail Tonnesen (EPA Region 8), Dan Cornelius (Intertribal Agricultural Council/University of Wisconsin-Madison)	90
Session 5: Commodity groups and state agencies	94
Bill Hammerich (Colorado Livestock Association): Commodity group perspective - Colorado Livestock Association	
Alan Blaylock (Nutrien): Fertilizer industry use of atmospheric deposition data	<del>)</del> 7
Greg Zwicke (USDA NRCS): USDA NRCS and reactive nitrogen	)4
Panel Discussion: Bill Hammerich (Colorado Livestock Association), Alan Blaylock (Nutrien), Greg Zwicke (USDA NRCS), Lisa Devore (CDPHE)10	)7
Wrap-up and next steps11	1

### Workshop organizing committee

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### Background

In 2019, members of the NADP Total Deposition Science Committee and collaborators completed a whitepaper outlining science needs for continued development of reactive nitrogen (Nr) deposition budgets in the U.S.

(http://nadp.slh.wisc.edu/committees/tdep/reports/nrDepWhitePaper.aspx). Improving current understanding of the role of agriculture in Nr deposition was identified as an overarching theme where important data and knowledge gaps persist. Advancements needed to address these gaps span a number of research areas including:

- characterization of spatial and temporal patterns of reduced N [ammonia gas (NH<sub>3</sub>) + ammonium aerosol (NH<sub>4</sub><sup>+</sup>)] deposition
- better understanding of Nr emissions from agricultural and non- agricultural sources
- improvement of atmospheric deposition models
- more accurate source apportionment of Nr deposition

A 1-day workshop "Connecting Stakeholder and Science Perspectives to Better Understand the Linkages Between Agriculture and Reactive Nitrogen Deposition" was convened to discuss recent advances and future directions in these topic areas and to better understand the interests and needs of stakeholders that can contribute to, and benefit from, better understanding the role of agriculture in Nr deposition. The workshop was held on November 4<sup>th</sup>, 2019 in Boulder, CO in conjunction with the Fall NADP Science Symposium and was attended by approximately 100 participants from academia, state and federal government, tribal nations, and private industry. The workshop consisted of invited presentations and panels, beginning with a science-focused morning session organized around the research areas mentioned above. This was followed by a more stakeholder-oriented afternoon session, also comprising invited speakers and panelists including participants from federal and state agencies, tribal communities, and commodity and agricultural organizations. Here we summarize the presentations and panel discussions, drawing on slides as well as audio of the presentations and panels. Following the format of the TDep white paper, presentations are organized into three parts: Introduction, State of the Science, and Future Directions. Panel discussions are organized by the main points and themes of the discussions.

### Summary of key points

Below is a summary of the key points raised in each session.

Session 1: Modeling and source apportionment of reactive nitrogen deposition

- There are several techniques available to apportion sources of Nr deposition depending on the specific question being addressed and available data.
- Modeling of land-use specific deposition is a step forward for ecosystem- and locationspecific deposition assessments.
- Key model uncertainties include parameterizations for NH<sub>3</sub> air-surface exchange processes and emissions inventories, particularly in capturing episodic emission pulses.
- More comprehensive (e.g. gas + particles, physical parameters that drive dry deposition) and higher time resolution (e.g., NH<sub>3</sub>) measurements are needed for model evaluation.
- Additional measurements of ecosystem Nr pools (e.g., soil and vegetation chemistry) are needed to improve parameterizations of NH<sub>3</sub> compensation points.
- Research is needed to make models more dynamic using techniques such as machinelearning, data assimilation, or sub-grid modeling.
- More model intercomparisons would be helpful in establishing comparability of models used for deposition assessments, evaluating model uncertainties, and prioritizing new measurements.
- More direct communication and collaboration between field scientists and modelers should be encouraged to ensure that model parameters accurately reflect measurements and real-world practices.

### Session 2: Emissions of reactive nitrogen

- Process-based emission models allow us to capture important short-term variability in emissions and to incorporate management strategies into models.
- Satellite measurements of NH<sub>3</sub> and NO<sub>2</sub> can play an important role in the identification and spatial allocation of emissions.
- More ground-based measurements of emissions and supporting process level information is needed but there is a sensitivity of producers to conducting those measurements in some places.
- Improving activity and management data and making those data more easily accessible are key needs for improving emission inventories for both CAFOs and crops.
- The scientific community will need to work with local commodity groups and producers to collect this information.

### Session 3: Spatial and temporal patterns of reactive nitrogen deposition

- Monitoring networks for Nr generally have good spatial coverage.
- A robust routine measurement of NH<sub>4</sub><sup>+</sup> is needed.

- Higher time resolution measurements of NH<sub>3</sub> are needed for model evaluation and modeling bidirectional flux.
- In the future, hybrid networks of fewer but more comprehensive base monitoring sites combined with supersites at select locations may be a cost-effective approach to address diverse needs for monitoring and process-level research.
- Satellites can complement ground-based monitoring to track trends in atmospheric composition over source regions.
- Satellites can help optimize site selection for ground-based monitoring and processoriented studies.
- Methods for assimilating satellite data into measurement-model fusion techniques are needed.

### Session 4: Federal stakeholders

- With respect to Nr deposition and better understanding the role of agriculture, the needs of federal stakeholders are diverse. Taking advantage of opportunities (e.g. workshops, conferences, stakeholder listening sessions) to communicate and identify areas of potential collaboration are therefore important.
- Evaluation of BMPs for NH<sub>3</sub> emissions (e.g., identifying most feasible options, quantifying emission reductions, assessing downwind impacts to deposition) represents a key opportunity for cross-agency collaboration and closer engagement with agricultural producers.
- The need for improved representation of agricultural practices in soil N cycling and emission models represents an opportunity for partnership with USDA ARS.
- There are wide-ranging opportunities (e.g. Intertribal Agriculture Council (IAC) conferences, outreach to tribal resource management groups, tribal colleges and universities) to partner with tribal nations on monitoring efforts and research activities related to Nr deposition.

### Session 5: Commodity groups and state agencies

- Solving these large-scale issues will be most successful if built on collaborative efforts with stakeholders, agencies, and non-governmental organizations. Farmers are appreciating a collaborative effort more than ever, rather than "top-down" regulatory approaches.
- Building partnerships with industry and commodity groups will take time but is doable if built on trust, transparency and communication.
- There are a number of lessons learned (e.g., early engagement with agricultural stakeholders, inclusion of stakeholders in planning and scientific discussions, involvement of stakeholders in development of objectives and solutions) from the RMNP Air Quality Initiative on building stakeholder relationships that can translate to future efforts in other parts of the U.S.

• Building communication with local agricultural commodity groups and extension services that engage with farmers directly will be key to addressing important data and knowledge gaps related to Nr emissions.

Products for the workshop will include the summary developed here, which will be posted to the NADP TDep website, and an Agricultural Stakeholder Engagement Plan to be developed by the TDep Stakeholder Workgroup. The stakeholder engagement plan will follow up on the opportunities identified during the workshop. Another product under consideration is a communication piece geared toward the agricultural community, framed by the needs of stakeholders and supported by the science discussed during the workshop and outlined in the TDep white paper on reactive N.

## Session 1: Modeling and source apportionment of reactive nitrogen deposition

Daven Henze (UC Boulder): Source attribution of reactive nitrogen deposition using models and measurements

### Introduction

In order to reduce human impacts of reactive nitrogen (Nr) deposition to sensitive ecosystems, we must understand the effect that regional and local anthropogenic and natural Nr sources have on specific ecosystems. Source apportionment or source attribution modeling is a key method to understand these effects. Daven Henze (CU Boulder) gave a keynote presentation to provide an overview of the current state of the science of source apportionment modeling, and to identify challenges that are present in the field and some potential research opportunities that may help to address those challenges.

Source attribution modeling is the main technique to quantify the contributions of individual sources of Nr to Nr deposition to a particular ecosystem. There are a range of modeling and measurement techniques; some highlighted applications were conducted in the US, China and Europe using approaches including chemical indicators, isotopic measurements, and constraints from remote sensing observations. Two major categories of air quality model sensitivity analyses are typically used to estimate source contributions: source (tagging, perturbation) and receptor (adjoint, trajectory) oriented modeling strategies. These two strategies can be used in tandem.

### State of the Science

Many challenges exist in the field of source attribution (Figure 1), the most significant were identified as the lack of adequate measurements for modeling constraints and evaluation. This includes observations of Nr deposition, including process-level and those made at a network-scale which often lack adequate spatial and temporal resolution. Modeling estimates are often uncertain as they heavily depend on both local and regional meteorological data to predict transport and transformation processes. Also, emissions inventories are a significant limitation (e.g. natural emissions vs those from industrial, transportation and agricultural activities) as they are uncertain in terms of magnitudes, distributions, and trends.

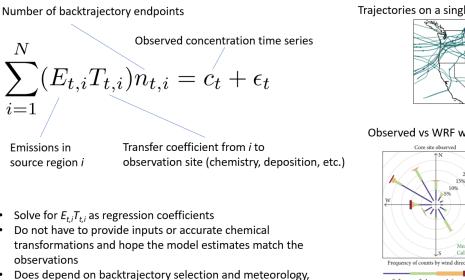
#### Challenges and opportunities for source attribution of Nr deposition TES: July 4 – 19, 2005 Lack of direct observational constraints on all components of Nr deposition Model estimates depend upon - Meteorology at fine scales and across great distances - Precipitation - Uncertain emission inputs - Source attribution methods - May require arbitrarily defined source regions - May be highly subject to uncertainties in model estimates CrIS: June 13, 2012 - Innovative methods to address each of these issues - Expanding observational datasets: - situ monitoring of NH<sub>3</sub> (AMoN in US and China) - Recent and upcoming remote sensing observations NO<sub>2</sub>: TROPOMI, TEMPO (2022) - NH<sub>3</sub>: CrIS, AIRS, IASI 239 241 Longitude Many regions undergoing transition of Nr sources

## Figure 1. Outline of challenges and opportunities for source attribution of Nr deposition with example maps of remote sensing observations of NH<sub>3</sub> concentrations derived from CrIS and TES.

Progress has been made to address some of these limitations and many research opportunities remain (Figure 1). This includes the development and future application of innovative source apportionment methods to address limitations and improvements in the observational datasets (from satellites and new monitoring, e.g. AMoN NH<sub>3</sub>).

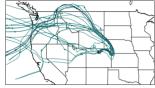
Henze directed the audience to Elliot et al. (2019) to highlight isotopic tracer methods that can be used in source attribution methods. There has been much research to develop the source isotopic signatures that can distinguish different source types, but there remains overlap in these source signatures that will limit the effectiveness of the method. More research is being conducted to refine these source signatures, including studies to understand effects of atmospheric fractionation and accumulation in bio-monitors that can be used as a surrogate for deposition.

### Trajectory mass-balance (e.g., Gebhart et al., 2011; 2014)

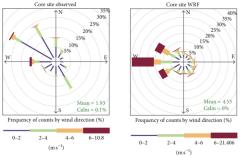


which are explored through ensembles

Trajectories on a single day (Gebhart et al., 2011)



Observed vs WRF wind rose (Gebhart et al., 2014)



### Figure 2. Basic format of trajectory mass-balance equation and wind trajectories and wind roses from Gebhart et al., 2011 and 2014.

The work of Gebhart et al., 2011 and 2014 was presented to illustrate the concept of a trajectory mass-balance model (Figure 2). These studies illustrate a receptor-based technique that uses wind trajectories and/or wind direction frequency to limit solutions to a basic mass balance equation where source strength weighted by transformation factor and trajectory factor are equal to concentrations at a receptor site. Major uncertainties with this method are from the trajectories and defining the source regions.

Sensitivity of all model concentrations to one model source

### Forward Model (source-oriented) Adjoint Model (receptor-oriented)

Sensitivity of model concentration in specific location to many model sources



cost scales with # of receptors

### Figure 3. Generic key differences between instrumented modeling capabilities with Eulerian Chemical Transport Models (CTMs): receptor-oriented (adjoint) vs. source-oriented (forward model)

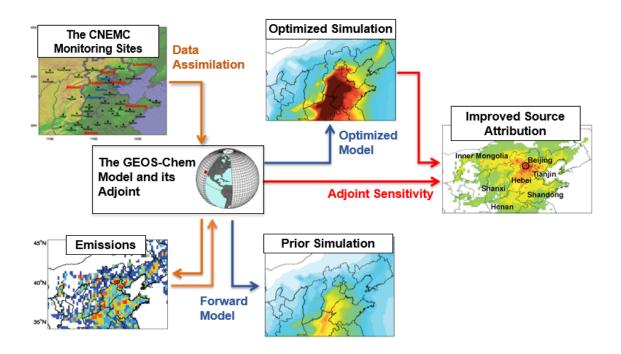
Generically, source apportionment models are typically the receptor-oriented model or the source-oriented model (Figure 3). The receptor-oriented model is demonstrated by Gebhart's work, and the adjoint model framework is defined as beginning with a concentration at a receptor  $(t_n)$  and running backward to a source contribution or possible point of origin,  $t_0$ . Adjoint models are used to propagate infinitesimal differences in deposition at a receptor site back to where the model would apportion a small change in deposition to contributions from all the sources considered at to.

Conversely, source-oriented (i.e., forward) apportionment approaches that follow source contributions (usually by tracer species or perturbation) at  $t_0$  and allow the model to propagate that perturbation forward. Forward source-propagation techniques are evaluated by comparing model results at the receptor site to receptor observations. Example studies shown were Zhang et al., 2018 and Thompson et al., 2015.

Which modeling strategy you select would depend on the study conditions and available data. An adjoint or receptor-oriented approach would be best suited for a study with a small number of receptor sites and a larger number of sources. A source-oriented approach would be preferable in a study with a small number of sources.

Hybrid techniques (e.g. Malm et al., 2016) combine the receptor-based methods and the source-methods. Source contribution estimates are used to derive regional source correlation patterns (derived from the singular value decomposition) and these patterns are used in the basic receptor model mass balance equation against observed time-series measurements. Simplistically, the source correlation patterns limit the number of possible solutions based on information from the source model.

The take-home message is that these differing techniques can often be combined in ways to optimize data inputs and improve the subsequent modeling steps (and outputs). For instance, adjoint models fit into the general framework of data assimilation of inverse modeling (i.e. the use of observed or modeled data as an endpoint to optimize model inputs).

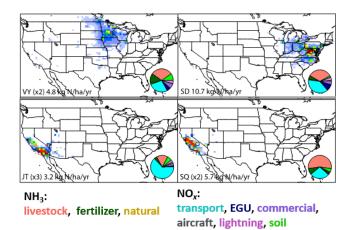


## Figure 4. Zhang et al., 2016 study is shown as a flow diagram of how different modeling techniques (data assimilation in orange, forward modeling in blue, and source attribution modeling in red) were coupled together to improve source attribution.

The Zhang et al., 2016 study is presented as an example that ties different modeling strategies together and incorporates data assimilation (Figure 4). In this study, speciated PM observations (CNEMC) were used in an inverse model to provide modeled 'optimized' emissions inputs (orange arrows from data assimilation to GEOS-Chem and from GEOS-Chem to Emissions). These "optimized" emissions inputs were fed back into the GEOS-Chem model (orange arrow from Emissions to GEOS-Chem) and rerun to get the "Optimized Simulation" ("optimized model" blue arrow). The change in the "Optimized Simulation" can be seen when compared with the "Prior Simulation". The adjoint sensitivity analysis is then run (red arrows) to get the final "Improved Source Attribution".

Up to this point, the model examples are used for emissions and concentrations. Paulot et al., 2013 applied these techniques to Nr deposition on a global scale. This was the first study to apply adjoint modeling techniques to calculate the source attribution of deposited species in a

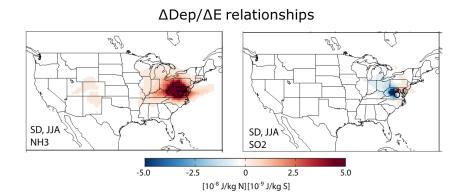
CTM. The paper identified the broad spatial scales of the emission footprints (often several 100's of km) upwind of several biodiversity hotspots around the word, and it also examined chemical couplings in secondary inorganic aerosol formation, transport, and deposition that can govern then magnitude and even the sign of the sensitivity of Nr deposition to emissions of  $NH_3$ ,  $SO_2$  and  $NO_x$ .



- Consider 8 Federal Class I areas in the US
- GEOS-Chem adjoint, 0.5° x 0.667°, 2010, stemming from NASA AQAST Tiger Team (D. Jacob, A. Russell, J. Milford, B. Schichtel, J. Vimont, R, Scheffe, J. Kelly, L. Pardo)
- Investigate contributions and exceedance sensitivities

### Figure 5. Lee et al., 2016 study describing the use of a GEOS-Chem adjoint model to investigate Nr source footprints (i.e. sensitivities) or Nr exceeding Critical Loads in Federal Class I areas (Voyagers, Shenandoah, Joshua Tree, and Sequoia). The pie charts inset into each map represent the source contributions of Nr considered (either NH<sub>3</sub> or NO<sub>x</sub> from different source types).

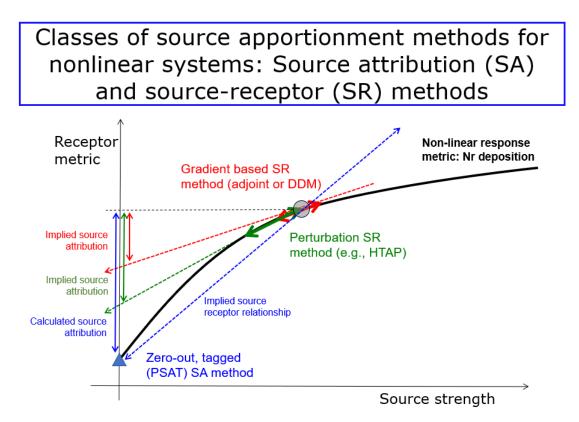
Focusing on a U.S. study in more detail, Lee et al., 2016 used a GEOS-Chem adjoint model to apportion Nr that deposited to 8 Federal Class I areas (Figure 5). The pie charts inlaid at the bottom right of each map reflect the speciation (i.e. NH<sub>3</sub> or NO<sub>x</sub>) and contributions from different source categories of Nr observed at the site. The maps themselves show the cumulative integration of the source sensitivity of deposition observed at the receptor location (i.e. the "source footprint"). The color scheme represents the quantity of the integrated source sensitivity in terms of percentage of the total. Red is a 90% accumulation, meaning that ~90% of the Nr that is deposited at the receptor location originates from this area. Blue represents a much smaller portion of the overall Nr observed at the receptor and source footprints at this level of accumulation are further away (100s of km) from the park. This technique helps to illustrate the regional Nr transport vs. the impacts of local Nr sources. There is a lot of local scale meteorology at smaller time-scales that drive the variability of deposition and broad, longer scale background features that drive deposition from sources that are further away.



 $SO_2$  forms  $(NH_4)_2SO_4$  aerosol in the summer that would have transported further away than  $NH_3$ 

Figure 6. Sensitivity study from Lee et al., 2016 of changes in Nr deposition in response to modeled input emission changes (i.e. perturbation) of NH<sub>3</sub> and SO<sub>2</sub> ( $\Delta Dep/\Delta E$ ) in Shenandoah NP in summer.

The study then looks at the sensitivity of the Nr deposition at the receptor site to changes in an emission source ( $\Delta Dep/\Delta E$ , Figure 6). The example used was Shenandoah National Park where an increase in NH<sub>3</sub> emissions would cause an increase in Nr deposition in the park, but an increase in SO<sub>2</sub> emissions would cause a net decrease in park Nr deposition. The reason for this is that the model has SO<sub>2</sub> reacting with NH<sub>3</sub> to form (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The NH<sub>3</sub> will tend to deposit locally (i.e. to the park), but the aerosol (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> will be transported through the park, thus decreasing local deposition. Atmospheric transformation effects such as this should be considered when trying to implement policies for deposition reduction.



## Figure 7. Conceptual schematic to illustrate the difference between classes of source apportionment methods on a current atmospheric state (black circle) and a non-linear response metric of Nr deposition.

A challenge with source attribution of Nr deposition is the nonlinear relationship between the magnitude of Nr emissions in one location and the magnitude of Nr deposition in another. This type of relationship is depicted in Figure 7, where the non-linear "response metric" in this case would be Nr deposition (represented by the solid black line in Figure 7). A generalized walk-through of the different source apportionment methods was presented as a visualization of two classes of methods: those based on a local linear gradient calculation, often referred to as a "source receptor" (SR) relationship and those that perform a complete source attribution (SA) calculation.

The SR class includes specific methods such as adjoint or the Decoupled Direct Method (DDM) exemplified in red. These methods calculate the change in the response metric (Nr deposition) due to a very small (infinitesimally small, mathematically) perturbation in emissions (i.e. source strength) which is represented as the solid red bidirectional arrow. These methods precisely quantify the linear local gradient of the source receptor relationship, which can be extrapolated for larger changes in emissions (i.e. dotted red line), including to the point where the source strength is zeroed out (i.e. a 100% emissions perturbation, shown in Figure 7 as the intersection of the dotted red line with the y-axis). The change in "height" of the new extrapolated intersection point from the original "height" of the current atmospheric state (dotted black line

in Figure 7) is referred to as the "implied source attribution" (vertical red arrow in Figure 7) and represents an approximation of the Nr deposition attributed to the source that was zeroed out. The "implied source attribution" in the case shown by Figure 7 is estimated with the assumption that the response metric is linear, which is inherently assumed by SR methods. Because the Nr response in this example is a nonlinear relationship, the "implied source contribution" is significantly underestimated and is an example of a limitation of this SR method using the source receptor relationship to estimate the source attribution.

Another SR class source apportionment method is the perturbation-based approach used in the HTAP project (exemplified in Figure 7 in green). This method is similar to the adjoint or the Decoupled Direct Method (DDM) method illustrated prior but is more of an approximation of the local linear gradient of the source receptor relationship, as it is based on a -20% perturbation to the emissions.

In contrast, the source attribution class, SA, takes the opposite approach and directly calculates the source attribution of the response metric. In general, SA methods include tagging schemes such as PSAT, or the zero-out approach. The zero-out approach removes the source strength (also a 100% emissions perturbation) and calculates the source attribution of Nr deposition at the receptor (represented by the solid blue arrow in Figure 7). Thus, the relationship of the response metric at source strength of 0 (blue triangle) and the current atmospheric state (black circle) is estimated as a linear "implied source receptor relationship" (dotted blue line in Figure 7). This method accurately quantifies the source attribution, but incorrectly estimates the source-receptor relationship (i.e. response metric) as linear and would overestimate the extent to which a small change in emissions might change the response metric.

Overall, these two classes of approaches (SR vs SA) have different strengths and limitations and thus are suited to address different policy questions. The figure also illustrates how these different classes of source apportionment methods can be complementarily used to more accurately characterize a response metric of interest.

### **Future Directions**

Innovative source apportionment methods are combining qualitative and quantitative constraints from both measurements and models. This was demonstrated in the highlighted studies and the different techniques. These different strategies will provide different solutions and source attribution, but if we understand the nuances and the assumptions of the methods, we can strategically use them in tandem to provide more accurate results.

There are many opportunities for coupling these source apportionment strategies with available datasets. The availability of constantly improving (in terms of accuracy and spatial resolution) satellite datasets are a good complement to these methods.

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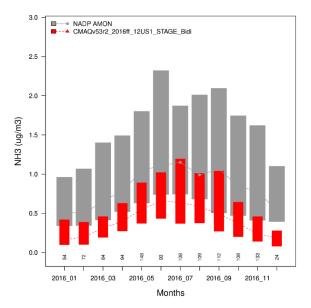
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### Introduction

The Community Multiscale Air Quality (CMAQ) model is used for numerous applications for atmospheric NH<sub>3</sub> (e.g. total deposition maps, atmospheric loading of N to the Chesapeake Bay, NH<sub>3</sub> emissions estimates for fertilizer). There has been an underprediction of modeled NH<sub>3</sub> concentrations as compared to observed NH<sub>3</sub> concentrations using the CMAQ model and other chemical transport models (CTM). This bias is a long-term artifact and is hypothesized to be due to limitations of the model representation of the bidirectional NH<sub>3</sub> deposition and also the underestimation of emissions in some areas.

Much recent progress has been made to "close the gap" with modeling the NH<sub>3</sub> emission and deposition processes by coupling agricultural models with CMAQ, introducing bidirectional exchange models. These include the new Surface Tiled Aerosol and Gaseous Exchange (STAGE) deposition scheme and Environmental Policy Integrated Climate (EPIC) model that are used in the latest version of CMAQ (v 5.3). Despite these improvements, the modeled NH<sub>3</sub> concentrations are still underpredicted (Figure 1).



### Figure 1. Comparison of monthly-binned NADP/AMoN samples vs. monthly-binned CMAQ/STAGE-modeled NH<sub>3</sub> at monitor sites for 2016.

This presentation discusses the new modeling configurations of the STAGE and EPIC modules and the sensitivities of the initial field and continental-scale simulation results with respect to model parameters and uncertainties of those parameters in agricultural and natural systems. Experiments discussed here explore the question of what is driving the bias between observed and modeled NH<sub>3</sub> concentrations from a bottom-up perspective.

### State of the Science

STAGE is a bidirectional dry deposition model that is an optional sub-module in CMAQ v5.3. This deposition model framework (manuscript in preparation) is an option alongside the M3DRY deposition model framework (Pleim et al., 2019). STAGE has several features that differ from M3DRY, primarily that fluxes are "tiled" for sub-grid land-use elements (e.g. deciduous, evergreen, water) and area-weighted up to the grid cell. This allows for the output of fluxes by specific land-use type, which is important for dry-deposition as key parameters that govern the deposition velocity are different for deciduous forests, evergreen forests, cropland, water, etc.

The resistance model itself is based on Kirchhoff's current law (as most resistance models are) and more specifically, the resistance parameterization framework (Figure 2) from Nemitz et al., 2001 and the Center of Ecology and Hydrology (CEH).

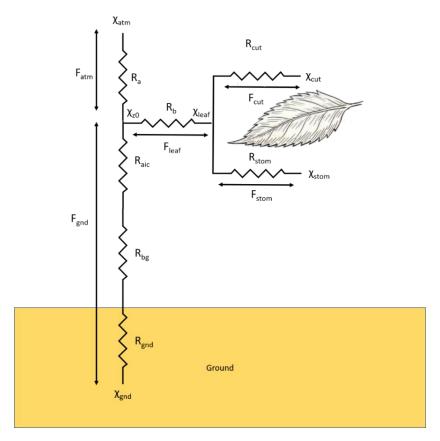


Figure 2. Framework of the STAGE NH<sub>3</sub> resistance model based on Kirchhoff's current law and adapted from Nemitz et al., 2001.

This framework incorporates the concentration gradients of species to the leaf and to the soil. The setting of these concentration gradients at different points in the model governs the bidirectional flux (whether you get evasion or deposition) at those points. STAGE is designed so that all species (e.g. Hg, NH<sub>3</sub>, VOCs, organic acids) can be evaluated for bidirectional flux via the compensation point,  $\chi$  (shown in Figure 2). The M3DRY framework has explicit equations for bidirectional flux only for the major species observed to have bidirectional fluxes (NH<sub>3</sub> and Hg). For the case of NH<sub>3</sub>, the compensation point is driven by the emission potential,  $\Gamma$  (ratio of NH<sub>4</sub><sup>+</sup> to H<sup>+</sup> concentrations) at the compensation point locations along the model at the leaf surface, the stomata, and the soil. If you specify a surface having a concentration > 0, then you get evasion, if the concentration is  $\leq 0$ , then you would get deposition (as in a unidirectional standard deposition model). Thus, setting up STAGE for bidirectional deposition is dynamic and easy.

Data that is input into STAGE includes the initial soil NH<sub>4</sub><sup>+</sup> fields. For natural soils, these are specified based on a limited set of soil chemistry measurements and extrapolated to different soil and vegetation types. More observational data is needed to improve these model inputs. For agricultural soils, the EPIC model provides the inputs on soil fertilization and mineralization, including soil NH<sub>4</sub><sup>+</sup> and pH.

EPIC is a USDA (United States Department of Agriculture) model initially developed to assess the effect of soil erosion on crop productivity and predict the effects of management decisions on agricultural and environmental considerations such as movements (of soil, water, nutrient, and pesticides), soil loss, water quality, and crop yields for areas with homogenous soils and management. As applied to CMAQ, EPIC is used to simulate biogeochemical processes relevant to N air-surface exchange in fertilized crops.

### EPIC emissions adjustments

Plant-demand nutrient needs drive the rate of fertilizer applications. The original EPIC 2016 simulation predicted that fertilizer application is 10.4 MT of N for the continental U.S. (CONUS), while the USDA Economic Research Service reported (from state data) 2014 fertilizer use as 12.1 MT of N (Figure 3). This represents a 20% under-estimation. In order to evaluate anomalies like this, post-processing tools were developed to adjust the EPIC fertilizer application output on a crop-by-crop; state-by-state to best reflect the observed data. This allows for the most realistic inputs to assess performance). Re-running the EPIC model with the 2014 data and extrapolating forward gives a much more reasonable 12.6 MT N (+4%).

	Published Values	EPIC	2016 STAGE post Processed EPIC	2016 STAGE Updated Г
EPIC Fertilizer Application	12.1 (MT N) <sup>1</sup>	10.4 (MT N)	12.6 (MT N)	12.6 (MT N)
EPIC Mineralization	-	23.0 (MT N)	7.4 (MT N)	7.4 (MT N)
Modeled Emissions	0.9-1.7 (MT N) <sup>2</sup>	1.4 (MT N)	0.9 (MT N)	1.6 (MT N)
Mean Annual Emissions Factor	7.1%-13.8% <sup>2</sup>	13.5%	6.9%	12.4%

1. 2014 USDA Economic Research Service (<u>https://www.ers.usda.gov/data-products/fertilizer-use-and-price.aspx</u>) 2. Using USDA ERS data and <u>Kilmont</u> & Brink 2004 emission factors

# Figure 3. Table (adapted from different slides of presentation) of emission rates and emission factors from literature (USDA ERS, Klimont and Brink, 2004) and model estimates from this study. Model runs include the EPIC model and the CMAQ + STAGE model estimates with the post-processed EPIC outputs both with and without the adjusted Γ values.

The original EPIC output  $NH_4^+$  from mineralization was more than a factor of 2 higher (23.0 MT N) than the output for the fertilizer application (initially 10.4 MT N) which is unrealistic. The algorithm was changed to the amount of organic N being applied plus 3% of the N uptake from the plants as "N uptake residue". This change had a significant effect and decreased the output  $NH_4^+$  from mineralization to a more reasonable 7.4 MT of N which was also in better agreement with estimates using the soil nitrogen mass balance method (Constantin et al., 2011).

The adjustments to the EPIC emissions mentioned above did not have a large impact on the  $NH_3$  net concentration bias. The increase in fertilizer application rate increased modeled  $NH_3$  concentrations by 5% (thus decreasing the bias to the observed  $NH_3$  concentrations), but the change in  $NH_4^+$  from mineralization decreased the modeled  $NH_3$  concentrations by -6% (increasing the bias). The increased  $NH_3$  concentrations from the fertilizer application rate occurred mostly during high  $NH_3$  periods and the decrease in  $NH_3$  concentrations from the mineralization manifested during periods of no fertilizer application and low  $NH_3$  concentrations.

Furthermore, there are known underestimates of N emissions, particularly in NH<sub>3</sub> from combustion sources (both industry and mobile; e.g. catalytic converters) and from the use of urea or NH<sub>3</sub> to create particles to suppress NO<sub>x</sub> or SO<sub>x</sub> emissions. Better representing these sources will likely reduce the modeled bias but they are estimated to be much smaller than animal husbandry and fertilizer NH<sub>3</sub> emissions which account for an estimated 81% of the NH<sub>3</sub> emissions (U.S. EPA, 2020). Thus, all of these results and considerations point towards the conclusion that merely improving N emission rate estimates is unlikely to resolve the modeled NH<sub>3</sub> concentration bias.

### Insights from measurements

All chemical transport models use estimates or parameter input into the algorithms used to represent bidirectional flux processes. Whenever possible, these estimates and inputs are based directly on field measurements and observational data or extrapolations of that data into different types of vegetation or meteorological conditions. The lack of measurements and field observations to confirm the accuracy of these parameters and extrapolations is the major knowledge gap in the field.

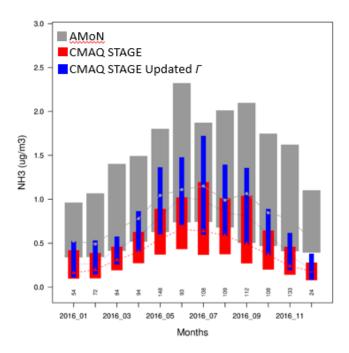
Examples of soil and vegetation  $\Gamma$  values that are input into CMAQ v5.3 are listed with measured values for grasslands, forest, and croplands in Figure 4. As part of an ongoing project to characterize biogeochemistry at AMoN sites for NH<sub>3</sub> flux modeling, EPA and Wood, Inc are developing expanded measurement databases of soil and vegetation emission potentials ( $\Gamma$ ) across different land cover types.

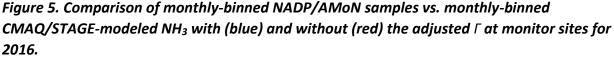
- Recent measurements from AMON site survey of the NH<sub>3</sub> emissions potential from vegetation and the soil surfaces
  - Emission potential ( $\Gamma$ ) ratio of molar NH<sub>4</sub> to H<sup>+</sup>
  - · Variable across land cover types
  - · Soil fluxes in natural systems appears to be driven by leaf litter

	Soil F	Vegetation <i>Г</i>
CMAQ v5.3	20	246-402
Grasslands Observations	1,000	1,000-2,000
Forest Observations	200 (Litter); 20 (Soil)	15-250
Croplands	100's to 1,000,000's	Variable with soil NH <sub>4</sub>

## Figure 4. Slide "Insights from Measurements" describing and listing emission potential ( $\Gamma$ ) calculated from field measurements for different land cover type as compared to the inputs for the CMAQ v5.3 model.

The calculated  $\Gamma$  using observed measurements proved quite different than the inputs to CMAQ for STAGE. They are highly variable across land cover types and across seasons. The CMAQ/STAGE model was rerun using approximations of the lower bound of these calculated  $\Gamma$  values (Figure 5) and the result was an improved model bias (Normalized mean bias decreased from -49% to -32%) and reduced error (RMSE decreased from 1.23 to 1.11 µg m<sup>-3</sup>). The correlation also slightly improved (Pearson's r increased from 0.66 to 0.68).





The adjustments to the  $\Gamma$  also did not have a marked effect on the agricultural emissions, which remained fairly stable, but increased emissions from semi-natural vegetation in most areas. For deposition, the level of the threshold of deposition was increased with the higher compensation points (with higher  $\Gamma$  and NH<sub>4</sub><sup>+</sup>), which increased the NH<sub>3</sub> concentrations. The resultant higher NH<sub>3</sub> concentrations also caused the deposition to increase once concentrations exceeded the elevated compensation points and the net effect produced deposition estimates that were very similar to the runs prior to the  $\Gamma$  adjustments. Thus, the most significant change was increasing the NH<sub>3</sub> concentrations, which helps to adjust the bias between modeled and observed NH<sub>3</sub> concentrations and the modeled emissions and deposition remained stable. This also has the implication that the models are underestimating the lifetime of NH<sub>3</sub>.

The updated NH<sub>3</sub> emission factors made in this study (Figure 3), resulted in the CMAQ/STAGE emissions to increase from 0.9 to 1.6 MT N and the Mean Annual Emissions Factor increases from 6.9 to 12.4%. Despite the nearly two-fold increase, this is still within range of published values as the STAGE run using the post-processed EPIC and original input  $\Gamma$  was near or just below the published range. The overall change appears to be significant, but the emissions represent a small change over a large area, e.g. this change primarily impacts non-agricultural land (73% of the land-use in the conterminous U.S. domain).

### **Future Directions**

It is important to note that this study represents an exploratory test run, and that these values will change when the CMAQ model is properly updated based on the observed data rather than

the quick adjustments tested here. However, these results allow us to conclude that, regarding  $NH_3$  emissions from fertilizer application, modeling the evasion of  $NH_3$  is dependent on the parameterization of  $NH_4^+$  in soil water solution.

Outputs from the EPIC model should be adjusted to better represent real-world management practices and the consideration of the nitrogen budget (e.g. the STAGE model mass balance checks at the soil layer level) is critical and should be included in top-down emissions assessments.

The adjustments to the soil and vegetation  $\Gamma$  values appear to significantly reduce the bias between modeled and observed NH<sub>3</sub> concentrations, without pushing the emissions and deposition estimates beyond reasonable values based on literature results from other studies. More work is needed in collecting field measurements and refining these input parameters particularly for seasonal dynamics in the soil and vegetation  $\Gamma$ .

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### Panel Discussion: Daven Henze (UC Boulder), Jesse Bash (EPA), Viney Aneja (NCSU), Amanda Cole (ECCC), Mike Barna (NPS)

The summary of the panel discussion is grouped into the following categories: audience questions for the presenters, panel discussion led both by moderator-posed questions and audience questions. Relevant questions and responses are grouped and summarized for clarity.

### **Follow-up Questions for Presenters**

### Question on sensitivity of modeled NH<sub>3</sub> deposition to pH.

Yanxu Zhang (Nanjing University) asked Daven Henze (University of Colorado Boulder) if he had looked into the sensitivity of the models to any impact of pH on NH<sub>3</sub> deposition. Daven responded that while he hadn't specifically looked at that effect on deposition, he has considered the role of heterogenous reactions (not necessarily a function of pH) in generating haze in extreme winter events, which would ultimately impact deposition (via phase partitioning). This is mostly done by reconciling differences between modeled and observed concentrations of sulfate and nitrate. Daven found that most sensitivity is limited to wintertime high humidity colder temperature episodes, and an overall impact to annual values may not be significant.

### Questions on some of the parameterizations and outputs from the STAGE model for NH3

A main conclusion of Jesse Bash's (EPA ORD) presentation highlighted the importance of the  $\Gamma$  ratio (and thus the NH<sub>4</sub><sup>+</sup> and H<sup>+</sup> concentrations) for modeling NH<sub>3</sub> emission from soils and vegetation and Charlie Driscoll (Syracuse University) asked for more insight as to how the H<sup>+</sup> concentration is specified in STAGE. For agricultural systems, the H<sup>+</sup> concentration is simulated by EPIC and soils are constrained within a specified pH range. In natural soil systems, the measured  $\Gamma$  values published in the literature are used. These values are extrapolated across different vegetation types and for different meteorological conditions and the number of measurements are limited. More measurements in different conditions are needed.

Viney Aneja (North Carolina State University) asked Jesse to elaborate on how the NH<sub>3</sub> emissions are treated in the model with respect to NH<sub>3</sub> concentration levels. His example was that at low NH<sub>3</sub> levels in the soil, fertilizer application will enrich N in soil and that will drive NH<sub>3</sub> deposition/evasion. At some point, a threshold level of N in soil will be reached, and NH<sub>3</sub> deposition/evasion will be governed more by physiochemical variables (e.g. temperature and pressure, relative humidity, soil moisture, or soil pH). Jesse responded that the model uses a Langmuir absorption curve to model the amount of NH<sub>4</sub><sup>+</sup> available in the soil pore solution but the model also takes into account the roles of pH, soil moisture, and soil temperature. This Langmuir algorithm is not completely correct, but useful in its representation of conditions and seems to make sense mechanistically.

Bret Schichtel (NPS) asked Jesse if the modeling results have been evaluated on a shorter time resolution to provide information on processes such as bidirectional flux and diurnal cycling. Jesse responded that the smaller time-resolution and field-scale estimates are not evaluated in the large Community Multiscale Air Quality (CMAQ) model, but instead in a box model. Some of the diurnal variability is captured in the box model, but it is a work in progress. He cited a current evaluation of observations from the Lillington study (Walker et al., 2013) and noted that key process such as the NH<sub>3</sub> in dew or wetted surfaces are not yet included in the model but will be added. The current box model is able to simulate some of the dynamics of the NH<sub>3</sub> diurnal profile in the nighttime and the early morning but can't reproduce the timing of the early morning dew peak.

Daven Henze remarked that the improvements made by the STAGE model are large steps forward in the field of modeling NH<sub>3</sub> concentrations, particularly in the bidirectional flux and the hourly distribution of emissions which have greatly improved the representation of the diurnal Nr concentrations.

### **Panel Discussion**

### Most important sources of uncertainty were related to modeled Nr deposition

The moderator opened the discussion by asking the panel what they felt were the most important sources of uncertainty in modeled Nr deposition.

Viney Aneja proposed that research should be conducted to see if a deposition model could be constructed in a framework different than the Ohm's resistance model framework (Wesely and Hicks, 2000). That a different method of modeling may help us to understand the processes of air surface exchange in new ways. Mike Barna (NPS) commented that he felt the framework of the resistance model is not the problem, but that the current resistance model framework is still poorly constrained. He felt that the two largest uncertainties in modeling the Nr deposition are the uncertainty in the Nr emission inventory and in modeling the dry deposition velocity. There are many research advancements to help with these uncertainties, the largest being the remote sensing datasets that would be particularly helpful in reducing uncertainties with the Nr emission inventory. He felt that addressing the gaps in the Nr emissions inventory will be easier than those for dry deposition.

In terms of the dry deposition velocity, perhaps a possible strategy could be to focus on simpler measurement techniques with the example being the use of monthly integrated flux measurements rather than measuring dry deposition at the hourly timescale with online techniques. A measurement strategy such as this could reduce cost and potentially increase the number of measurements.

Amanda Cole (Environment Climate Change Canada; ECCC) commented that many parameters affect deposition and it is therefore important to have very comprehensive measurements including meteorology and biogeochemistry at one location so that all processes (e.g. gas-

particle phase partitioning, aerosol scavenging) can be thoroughly assessed for developing the modeling algorithms. From the audience, Gail Tonneson (EPA Region 8) agreed with this, stating that the air quality model NH<sub>3</sub> estimates often have biases which are very difficult to identify. The biases could possibly come from different process including uncertainties in the emissions, gas-aerosol partitioning, vertical mixing, and the diurnal cycles of NH<sub>3</sub>, particularly uncertainties with the early morning dew evaporation peak often observed.

Jesse Bash agreed with Gail and Amanda's assessment of the utility of comprehensive measurements and added that co-located gas and aerosol phase data would be helpful in understanding if the models were low on NH<sub>3</sub> gas or perhaps low on NHx as a whole. He also added that observations on the pH of the dew and vegetation would be helpful to parameterize models to better understand the drivers of the morning NH<sub>3</sub> concentration peak thought to be associated with emission of NH<sub>3</sub> in evaporating dew.

### Accuracy of Nr estimates from biogeochemical models

Limei Ran (USDA NRCS) commented on the development of EPIC and its implementation in the CMAQ bidirectional model. Limei indicated the approach is based on N demand by crop which dynamically links the biogeochemical parameters to the atmosphere, and because of these dynamic interactions caution is needed in changing lone inputs.

The moderator commented on the increasing importance of the accuracy of Nr estimates from biogeochemical models and asked Limei what some of the biggest limitations and uncertainties were. Limei identified two main areas, improved crop management information and better fertilizer parameterization (e.g., type, amount, timing, method of application). Both will become more important as EPIC is applied for more local scale assessments. Peter Vadas (USDA ARS) added to this by acknowledging that a big limitation to EPIC and models like it (his vantage is from a phosphorus perspective, but that model shares many processes with NH<sub>3</sub>) was in its ability to properly model surface applications of fertilizer and particularly in terms of its relationship to meteorology. A potential way to deal with this is to communicate the modeling algorithms and how they are parameterized to the field experiment designers.

Jesse Bash agreed that models are likely missing a lot of emissions from manure-management processes, especially in application to the fields. Mike Barna concurred that these processes are unknown and likely represent a huge "pulse event" of NH<sub>3</sub> emissions that are certainly not captured in emissions inventories. A focus is needed to sample these pulse emission events but cannot be done with samplers with 2-week exposure periods (e.g., AMoN). There is the question of whether we are getting the annual bulk emissions correct, but also how would you distribute those emissions temporally?

B.H. Baek (University of North Carolina) commented from the audience that one way to incorporate episodic emissions could be deriving an estimate in a reverse approach, where if NH<sub>3</sub> emissions could be incorporated based on activity values, and if we can incorporate activity

into NEI, then that information could be used to calculate an estimate. This is not an approach that is normally done, but something that could be looked into given the need.

### Use of isotopic measurements in source apportionment

The moderator asked Amanda Cole to elaborate on research needed to advance the field of isotopic measurements for source apportionment applications. Amanda reiterated that much of the discussion in the TDep white paper chapter and associated paper (Elliott et al., 2019) dealt with development of techniques to quantify very small amounts of analyte, which is a significant hurdle. Also, experimental studies are needed to develop modeling algorithms to accurately characterize reaction rates and atmospheric processing (both chemical or physical) that contribute to isotopic fractionation that occurs during transport from emission to deposition. Amanda noted that she had seen some isotopic reaction rates included in a version of CMAQ. Lastly, Amanda noted that isotopic ratios from more diffuse sources need to be characterized. All of this is difficult and challenging but can be done. A first step could be to consolidate the existing data into an accessible database.

Peter Adams pointed out the large ranges in isotopic signatures and overlaps from different source categories, particularly for NH<sub>3</sub>, which could make the utilization of these signatures difficult in source attribution methods. In addition, in the case of NH<sub>3</sub>, the primary source is agriculture (livestock) and there may not be determinable differences in isotopic signatures from different livestock operations (i.e. same source-type but different source locations). He asked if we should be hopeful of isotopic techniques advancing enough to prove useful in source attribution of Nr?

Amanda acknowledged these points but noted that the difference in source locations would still be resolvable via traditional source apportionment techniques and that the isotopic signature would be complementary to these. The effectiveness of the technique may be dependent on the specific location of interest. For example, it would be effective in an area where deposition is influenced by both urban (mobile or EGU) and rural (i.e. livestock) sources. In reference to the large ranges on the source profiles/signatures, some of the spread (shown in the Elliott et al., 2019 figure) is due to the averaging of data collected over multiple seasons. More analyses can be done to further separate out patterns and reduce the error bars on the N source profiles.

### Preferred source apportionment techniques for decision makers and stakeholders

Chris Clark (EPA ORD) had a question regarding the use of source apportionment models from a perspective of a scientist that is interested in using these techniques and interpreting their results. The techniques all appear reasonable, but is there any recommended approach that stakeholders or policy decision makers should use? And is there a recommended technique suited for a resource-constrained budget? His example was a prospective project of building in source apportionment tools in the Critical Loads Mapper Tool.

Daven Henze responded that the question will ultimately dictate which technique is most applicable for each situation. He mentioned that Particulate Matter Source Apportionment Technology (PSAT) is an excellent technique in locating origins of pollutants. If the researcher is investigating how pollutant levels at a receptor site may change due to short-term emissions reductions from control strategies, then a perturbation technique is a better tool to use. There are many variables to consider with the perturbation strategies including precursors that act as limiting reagents and short-term vs. long-term effects.

### **Closing Comments from Panel**

The panelists were asked to give a summary of their most important research needs for Nr modeling and source apportionment. Viney Aneja started off with the improvement of Nr emissions inventories for agricultural operations. He also mentioned that the US Nr deposition modeling framework should be compared with the EU framework as there are differences (e.g. longer transport distance of NH<sub>3</sub> predicted in US framework).

Amanda Cole agreed with Viney's point on comparing modeling frameworks and reported that ECCC's air quality model, the Global Environmental Multiscale – Modelling Air-quality and Chemistry (GEM-MACH) as well as CMAQ are included in the Air Quality Model Evaluation International Initiative (AQMEII-4) study which is focused on comparison of dry deposition models and investigating specific dry deposition parameters in Europe and North America.

Daven Henze thought that models are currently 'stove-piped' or streamlined (via inputs and algorithms) to simulate an individual process or system. Research is needed to make models more dynamic using techniques such as machine-learning, data assimilation, or sub-grid modeling. This could be used, as an example, to assimilate observations from an episodic emission event into the model. A precedent for this could be a weather forecasting model coupled with a biomass burning event. Sub-grid modeling of deposition could help resolve processes on scales that are not possible with current CTMs.

Jesse Bash thought that the greatest need from a modeling perspective was more observations, including satellite, in-situ, and especially controlled laboratory measurements (i.e. soil and vegetation experiments). He added that upcoming comparisons of deposition models will help us understand which species are currently well-modeled and which are lacking. The results will provide some insight into what processes are driving the results and will allow for prioritization of areas needing the most improvement.

Mike Barna added that the US Nr deposition modeling efforts largely employ two models, the Comprehensive Air Quality Model with Extensions (CAMx) and CMAQ. In his opinion, CMAQ is further ahead, particularly in its NH<sub>3</sub> bidirectional flux capabilities, but a strength of CAMx is that it is simpler and easier to run. An intercomparison should be performed between these two models to characterize differences and uncertainties.

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### Session 2: Emissions of reactive nitrogen

Peter Adams (Carnegie Mellon University): Process-based ammonia emissions inventories from livestock – status and needs

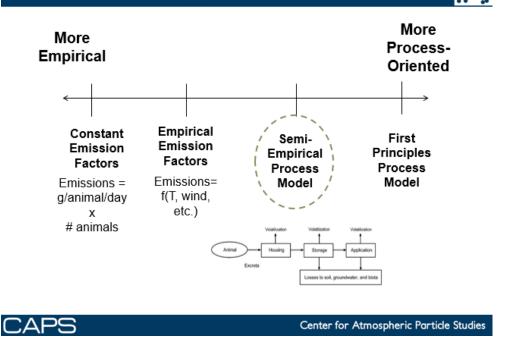
### Introduction

Livestock production is the primary source of atmospheric ammonia (NH<sub>3</sub>) in the U.S. (U.S. EPA, 2014). Accurate estimates of NH<sub>3</sub> emissions are needed to simulate air concentrations of NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup> aerosol, and wet and dry deposition of NHx (NHx = NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup>) in atmospheric models. Emissions estimates are also needed to apportion sources and develop mitigation strategies for inorganic PM<sub>2.5</sub> and nitrogen deposition. This talk briefly summarizes the process-based NH<sub>3</sub> emissions modeling that currently underlies NH<sub>3</sub> emissions from livestock in the U.S. Environmental Protection Agency's National Emissions Inventory (NEI). Emphasis is placed on some of the gaps and challenges associated with: a) incorporating measured emissions factors as constraints on the emissions model and b) characterizing diverse farming practices on a national scale.

### State of the Science

Methods for estimating NH<sub>3</sub> emissions vary widely in complexity from relatively simple emission factors, in which emissions are constant and are calculated by applying an emission rate (g/animal/day) to an activity metric (i.e., number of animals), to complex models in which emission processes are described using first principles. A schematic representing these different approaches to estimating emission factors is shown in Figure 1 and arranges these based on the level to which they are empirical or more process-oriented.

### **Versus Other Approaches**

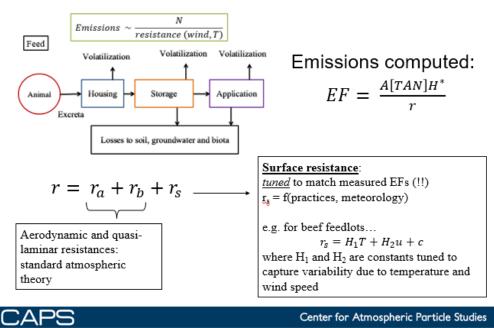


### Figure 1. Comparison of different approaches for estimation of emission factors based on degree of dependence on measurement or process

Empirical emission factors that incorporate drivers of emissions such as temperature or wind and semi-empirical process-based models represent an intermediate level of complexity, attempting to explain more variability in emissions than a simple constant emission factor while minimizing computational intensity and the need for extremely detailed input data that may not be readily available (e.g., first-principles model).

The goal of process-based models is to reproduce and capture as much real-world variability in emissions as possible. An example of a semi-empirical process-based model for NH<sub>3</sub> emissions from a confined animal feeding operation (CAFO) is shown in Figure 2. Emissions are estimated for each management component of the facility, including housing, manure storage (e.g., lagoon), and manure application. For each component, emissions are estimated using mass balance and mass transfer principles. The semi-empirical process-based model (McQuilling and Adams, 2015) shown in Figure 2 is used as the basis for the NEI. This approach simulates emission factors without too much bias by tuning parameters (e.g. surface resistance,  $r_s$  in Figure 1) to match measured emission factors. Furthermore, the model is not overly detailed so it can be applied at the national scale. Emissions depend on variables such as emission surface area, total ammoniacal nitrogen in the manure, and atmospheric resistances (e.g., aerodynamic ( $r_a$ ) and boundary layer ( $r_b$ ) resistances). Surface resistances ( $r_s$ ) are empirical, depending on factors such as temperature and wind speed and their corresponding regression parameters and regression functions are tuned to match literature emission factors.

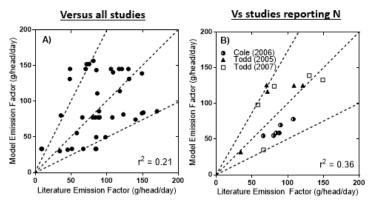
## Methods: Details



### Figure 2: Example of a process-based model of NH<sub>3</sub> emissions from a Confined Animal Feeding Operation (CAFO) (McQuilling and Adams, 2015).

While semi-empirical process-based models are an improvement over constant emission factors, they require additional measurements beyond just the emissions themselves. This requirement makes process-based models more complicated to apply in inventories. Traditionally, emission factors were used directly in inventories and little additional contextual information regarding farm management practices or measurement conditions was needed. Semi-empirical process-based approaches still require emission factors for model fitting but also require supporting measurements of key drivers of emissions such as the amount of nitrogen in animal feed and manure, temperature, and wind speed. Figure 3 illustrates the improvement in model performance that can be achieved when supporting measurements of key processes or nitrogen pools are available (i.e., contextual information). In this example, the semi-empirical process-based model explains more variability in emissions when considering only those studies reporting the amount of nitrogen in the feed or manure. While these additional measurements may increase the cost and complexity of emission studies, process-based models allow interpolation and extrapolation of limited emission factor measurements to other conditions and evaluation of alternative manure management practices.

### **Role of "Contextual Information"**



- Not all studies report all required input parameters (e.g. feed or manure nitrogen)
- Measurements need to report feed N, other practices, and meteorological conditions to put results in context and be useful to process-based models and inventories

CAPS

Center for Atmospheric Particle Studies

### Figure 3: Comparison of literature versus modeled (semi-empirical model) emission factors.

Inclusion of contextual information in emission studies is key to improvement and wider application of process-based models in inventories. Lack of emission measurements for specific animal types is another limitation to improvement of national emission inventories. For example, much less is known about emissions of NH<sub>3</sub> from pasture cows relative to cattle feedlots. While the emission factor for feedlot cows is as much as 8 times higher (CAPS model estimate) due to higher nitrogen intake, denser housing conditions, and lower soil infiltration rates, pasture cows represent a much larger fraction of the cattle and calf inventory. Scaled up to the national level, pasture cows may contribute ~ 40% of total NH<sub>3</sub> emissions from cattle, though more direct measurements of NH<sub>3</sub> emissions from pasture cows are needed to reduce the uncertainty in this estimate.

Knowledge of the distribution of farming practices at the national scale is another key data gap in improving emission inventories. Figure 4 summarizes aspects of uncertainty in the national NH<sub>3</sub> emission inventory for dairy cows (Pinder et al., 2004a). In this analysis, the National Practices Model (NPM) is used to estimate the frequency distribution of farm types in each county of the U.S., where a farm type is the set of farming practices that describes how manure is managed. The process-based Farm Emission Model (FEM; Pinder et al., 2004b) is then used to calculate monthly emission factors, given a set of farming practices and a temporal profile of meteorological inputs. As shown in Figure 4, uncertainty associated with knowledge of farming practices (NPM) is larger than uncertainty in the emission factors generated by the FEM. In addition to data on farming practices, access to accurate livestock population data is critical to development of national inventories.

### **Farming Practices!**

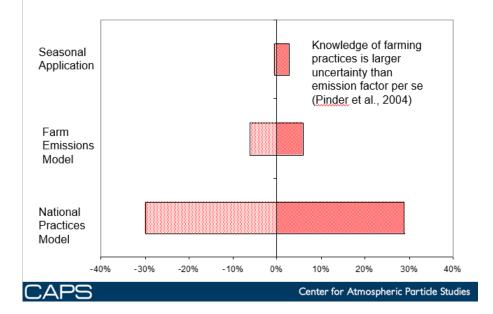


Figure 4: Sources of uncertainty in the national inventory for NH<sub>3</sub> emissions from dairy cattle (Pinder et al., 2004a).

### **Future Directions**

Improvement and wider use of process-based models in national NH<sub>3</sub> emission inventories is limited by several data and knowledge gaps. From a measurement standpoint, future emissions studies must include measurements to characterize nitrogen pools and processes as well as the physical and chemical drivers of emissions. Availability of data on farming practices and animal populations is insufficient to significantly advance current county-scale emission inventories or to develop a facility-scale emission inventory for any animal category at this time. Greater access to these data will likely require closer collaboration with commodity groups to collect information at the local level rather than relying on current national databases. Finally, the ability of atmospheric models to accurately simulate particulate matter formation and atmospheric deposition is strongly dependent on the quality of the underlying emission inventories. Evaluation of chemical transport models against ambient monitoring data is an important component of understanding the impacts of changes in emission inventories. With respect to constraining emissions of NH<sub>3</sub>, expanded monitoring of total NHx (NH<sub>3</sub> gas + NH<sub>4</sub><sup>+</sup> aerosol) or collocated NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> is needed.

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Kang Sun (University at Buffalo - SUNY): Observational data-driven constraints on the emissions and lifetimes of reactive nitrogen

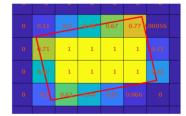
#### Introduction

Reactive nitrogen compounds visible to satellite-based sensors include nitrogen dioxide (NO<sub>2</sub>) and ammonia (NH<sub>3</sub>). Currently, tropospheric column abundance NO<sub>2</sub> is measured by multiple sensors on orbit, including OMI, GOME-2, OMPS, and TROPOMI. Ammonia is currently measured by the Infrared Atmospheric Sounding Interferometer (IASI) onboard the MetOp-A (2008-current) and MetOp-B (2013-current) satellites, as well as the Cross-Track Infrared Sounder (CrIS) onboard the Suomi National Polar-orbiting Partnership (S-NPP) satellite (2013-current). As illustrated in this presentation, satellite observations can be used to develop "top down" estimates of emissions to help inform the accuracy of "bottom-up" emission inventories. Satellite observations can also be used to characterize the lifetime of reactive nitrogen species in the atmosphere.

#### State of the Science

Satellites provide global-scale coverage of NO<sub>2</sub> and NH<sub>3</sub> observations. However, the spatial and temporal variability of satellite observations can be problematic for many applications. These limitations can be reduced by averaging satellite observations over time at a grid size smaller than the size of the satellite pixel itself. This procedure, known as physical oversampling, is a useful tool for processing satellite retrievals so that spatial and temporal features are more visible.

#### Physical oversampling satellite data



Grating spectrometers, e.g., OMI, TROPOMI

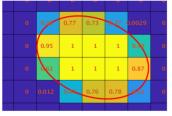
Weight

$$C(j) = A(j)/B(j),$$

where

$$\begin{split} A(j) &= \sum_{i} \frac{\Omega(i) S(i,j)}{\sigma(i)^p \sum_{j} S(i,j)} \\ B(j) &= \sum_{i} \frac{S(i,j)}{\sigma(i)^p \sum_{j} S(i,j)}. \end{split}$$

Zhu et al. ES&T 2017 Sun et al. AMT 2018

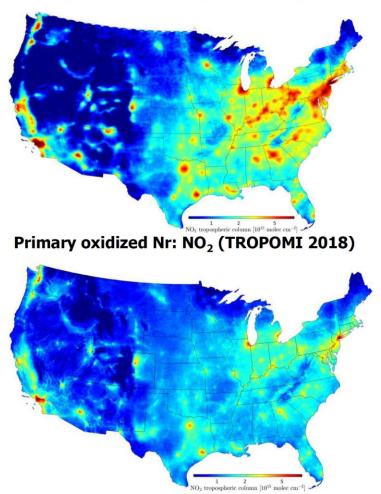


Fourier Transform Spectrometers, e.g., IASI, <u>CrIS</u>

- $\Omega(i)$  is the satellite retrieval at pixel *i*; C(j) is the result at grid point *j*
- *S(i,j)* is the spatial sensitivity of pixel *i* at grid cell *j*
- $\sum_{j} S(i,j)$  normalizes the sensitivity of pixel *i*
- $\sigma(i)$  is the retrieval uncertainty for pixel *i*

# Figure 1: Examples of oversampling geometry and weighting for grating and Fourier transform type spectrometers. Satellite pixels appear in red. Destination grid cells (squares) are 5 km x 5 km, and the overlapping areas are normalized by the grid cell area (25 km<sup>2</sup>), as labeled in each grid cell.

Physical oversampling produces a weighted average retrieval at a user-defined grid resolution. In the approach outlined in Figure 1 (Sun et al., 2018; Zhu et al., 2017), the contribution of each satellite observation to a given grid cell is weighted by the normalized sensitivity of the observation on that cell and inversely weighted by the observational uncertainty. In Figure 1, C(j) is the oversampled result for grid cell *j*.



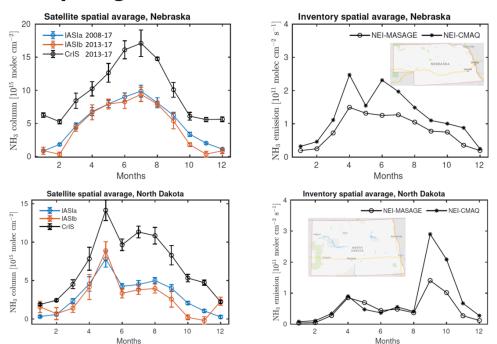
#### Primary oxidized Nr: NO<sub>2</sub> (OMI 2007)

#### Figure 2. Oversampled results for NO<sub>2</sub> from OMI (2007) and TROPOMI (2018).

Figure 2 shows an example of the spatial and temporal resolution of NO<sub>2</sub> that can be achieved with oversampling. A dramatic reduction in NO<sub>2</sub> column concentrations is observed between

2007 and 2018 resulting from reductions in NOx emissions. Urban areas are highly visible and major roadways can also be seen (e.g., in the Southwest).

Oversampling also allows for detailed comparisons of satellite retrievals in specific locations or domains. As shown in the left-hand plots in Figure 3, while IASI and CrIS column NH<sub>3</sub> concentrations show consistent temporal patterns, the absolute concentrations can differ substantially. Comparison of the satellite NH<sub>3</sub> concentration time series (left) with the temporal pattern of NH<sub>3</sub> emissions derived from the U.S. Environmental Protection Agency's National Emissions Inventory used in the Community Multi-scale Air Quality Model (NEI-CMAQ) and emissions derived from the Magnitude And Seasonality of AGricultural Emissions (MASAGE) model (Paulot et al., 2014) shows some disagreement in the timing of peak satellite NH<sub>3</sub> concentrations and "bottom-up" emissions.



#### **Comparing satellites with inventories**

Figure 3. Monthly time series of IASI and CrIS NH<sub>3</sub> total columns (left) and monthly NH<sub>3</sub> emissions derived from the U.S. Environmental Protection Agency's National Emissions Inventory used in the Community Multi-scale Air Quality Model (NEI-CMAQ) and emissions derived from the Magnitude And Seasonality of AGricultural Emissions (MASAGE) model.

Satellites also provide an opportunity for "top-down" estimation of NH<sub>3</sub> and NO<sub>2</sub> emissions for comparison to "bottom up" inventories. One approach relates an emission rate (Q) to an integrated mass enhancement (IME) and atmospheric lifetime ( $\tau$ ) as detailed in Figure 4. The IME, which is used in the methane research community, represents the satellite total column concentration integrated over a specified source area (Varon et al., 2018; Jacob et al., 2016). Enhancement refers to the difference between the emission impacted column concentration and the unimpacted background concentration.

#### Data-driven mission/lifetime estimations

• Integrated mass enhancement (IME) over a source region

$$IME = \int \Delta \Omega dA = \langle \Delta \Omega \rangle A$$

• <> is the spatiotemporal averaging operator. Emission rate Q:

$$Q = \frac{\text{IME}}{\tau}$$

• For conservative tracers like CH<sub>4</sub> or CO, the residence time is only determined by wind dispersion (L = length scale, U = wind speed):

$$\tau = \tau_d = L/\langle U \rangle$$

• Add chemical lifetime ( $\tau_c$ ) for reactive species like NO<sub>x</sub> or NH<sub>3</sub>:

• Hence:

$$\tau = \frac{1}{1/\tau_d + 1/\tau_c} = \frac{1}{\langle U \rangle / L + 1/\tau_c}$$
  
IME =  $\langle \Delta \Omega \rangle A = \frac{Q}{\langle U \rangle / L + 1/\tau_c}$  Two unknowns:  $Q$  and  $\tau_c$   
One parameter to calibrate:  $L$ 

### *Figure 4. Summary of integrated mass enhancement approach for estimating emissions using satellite retrievals.*

As indicated in Figure 4, knowledge of the lifetime of the species of interest resulting from dispersion ( $\tau_d$ ) and chemical reactivity ( $\tau_c$ ) over the source region is also needed. Wind speed (*U*) and length scale (*L*) of the source region are required for estimation of ( $\tau_d$ ). Figure 5 illustrates an example in which emissions of NH<sub>3</sub> over the Southern California Air Basin (SoCAB) are estimated by separating the IME over a range of *U*, which enables fitting the basin-scale emission (*Q*) and the chemical lifetime ( $\tau_c$ ) simultaneously. In this example, IME derived emissions are with the range of emissions estimated by Nowak et al. (2012) from aircraft measurements.

#### Fitting NH<sub>3</sub> emission/lifetime

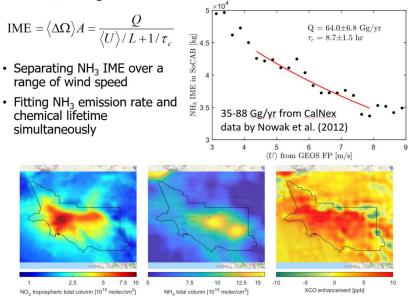


Figure 5. Example of NH<sub>3</sub> emission estimation for the Southern California Air Basin using the Integrated Mass Enhancement approach.

#### **Future Directions**

Physical oversampling and column-based emission estimates provide great opportunity for more detailed analysis of the spatial and temporal behavior of sources and sinks of reactive N species. However, additional work is needed in key areas to advance the use of satellite measurements in this regard. Additional work is needed to improve NH<sub>3</sub> column retrievals and their validation to reduce the discrepancies between among satellites observed here. Furthermore, more accurate estimates of the source region length scale and atmospheric lifetimes for reactive chemical species are needed to improve the accuracy of the IME emissions estimation method. The availability of CO observations from TROPOMI makes it possible to directly constrain the atmospheric dispersion lifetime for each air basin, using CO as a relatively conservative tracer. Leveraging the existing long record of NO<sub>2</sub> and NH<sub>3</sub> satellite observations will help improve our understanding on the temporal trends of emissions and chemical regimes over the critical reactive nitrogen source regions.

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#### Panel Discussion: Peter Adams (CMU), Kang Sun (UaB), April Leytem (USDA ARS), Marc Houyoux (EPA OAQPS), Ian Rumsey (EPA ORD)

The summary of the panel discussion is grouped into the following categories: audience questions for the presenters, panel discussion led both by moderator-posed questions and audience questions. Relevant questions and responses are grouped and summarized for clarity.

#### **Follow-up Questions for Presenters**

#### Process-based emission models

The audience questions and comments for Peter Adams (Carnegie-Mellon University) centered on the comparison of process-based models to emission factors. Viney Aneja (North Carolina State University) commented that the process-based model is being calibrated with emission factors, and asked how the model differs from emission factor and how is the process-modeling approach more useful? The response was that process-based models better explain variability in emissions than emission factors (EF). They are valuable as a way of interpolating between locations and to capture variability in emissions driven by factors such as temperature and windspeed. Overall, the model is unbiased with respect to EF because it is tuned to them, but the simple process-based models can explain the variability (i.e. scatter) around the mean emission. Process-based models are a step-forward, but not a "magic bullet" and there are still limitations that persist.

Bret Schichtel (NPS) commented that in Colorado a coordinated effort is being made to modify agricultural activities to limit N emission during upslope meteorological events that transport nitrogen into Rocky Mountain National Park. To evaluate the success of these alternative practices, we need to know how agricultural management activities affect emissions, particularly short-term emission "pulses". Process based models allow us to capture this important short-term variability in emissions and to incorporate management strategies into models. Emission factors do not allow this.

#### Satellite resampling methodology

Audience questions for Kang Sun (University at Buffalo) focused on the technical details of the resampling procedure presented. Yanxu Zhang (Nanjing University of China) noted that the retrieval methodology is highly technical and asked how the grid resolution for resampling is decided and has the sensitivity to the choice of resolution been tested? Kang Sun distinguished between the "resolution" and the "sample size grid". The resolution is an inherent property of the instrument and is fundamentally decided by the size of satellite pixel. It. Satellites can have different resolution but can be resampled on the same grid size (1km, 10km, etc). An example of this is that TROPOMI has a better resolution than OMI, but both can produce the same size sampling grid. The sample size limit is based on objectives of the dataset and what answers you

seek. One criteria is that you want to sample everything on the same grid size to maintain spatial consistency. The integrated mass enhancement (IME) approach (demonstrated in his talk) uses oversampling (lots of averaging and smaller grid size) which minimizes the differences in grid resolution of the resampling procedure because the data is averaged.

Daven Henze (UC Boulder) asked about the impact of instrument sensitivity and the retrieval algorithm on the uncertainty of the resampling procedure and the column concentrations. In one of the plots presented, IASI and CrIS column concentrations differ by factor of 2, which illustrates that what the instruments are seeing is fundamentally different. How does the IME method account for instrument sensitivity and its impact on the uncertainty of column concentration? Kang Sun responded that, within the simple framework presented, there is no way to consider vertical resolution of the averaging kernels. All averaging kernels will have some uncertainty, i.e. vertical distribution of sensitivity, which is not evenly distributed across the troposphere. The impact of lightning NOx on NO<sub>2</sub> is an example. The averaging kernel is more sensitive to the higher part of the troposphere. In the framework presented, this will be included as systematic uncertainty of the column concentration.

#### **Panel Discussion**

### The panel was asked to give their opinion on the largest uncertainties in emission inventories for reactive nitrogen.

#### Spatial and temporal allocation of emissions

Marc Houyoux (EPA OAQPS) began the discussion by noting that we have emission estimates at county level, but we need better spatial allocation at sub-county scale. A critical data gap here is the location of animal facilities. Perhaps satellites can help in this regard. Mark Barna (NPS) reiterated that we struggle with spatial allocation. For example, cows are dispersed throughout a county and we need to know where they are to properly allocate the emissions. A nationally coordinated method of developing a database of CAFO facility locations is critically needed. April Leytem (USDA ARS) noted that Google Earth is helpful. However, this may not be a practical approach for regional or national scales. Ian Rumsey (EPA ORD) noted that North Carolina State University has developed a facility-scale database using Google Earth for North Carolina and that perhaps facility identification could be automated. B.H. Baek (University of North Carolina) suggested that this kind of GIS information (i.e., shape files of NC CAFO locations) could be combined with satellite observations to refine spatial allocation at the sub county-scale.

Peter Adams presented a slightly different perspective, acknowledging that satellites will help with spatial allocation but that for some applications or questions sub county-scale spatial resolution in emissions is not required. For example, transport and receptor scales are on the order of hundreds of kilometers. Moving a single animal production facility 10 km in a county will not strongly affect chemical transport model predictions of PM<sub>2.5</sub>, for example. We need to make sure, though, that the animal populations at the county scale are accurate. Marc Houyoux

agreed that sub-county allocation is not an issue for some processes (regional transport) but is important for local issues/local planning.

Marc Houyoux noted that satellites can also help with temporal allocations. We now have monthly temporal allocation in inventories. As illustrated in Kang Sun's presentation, temporal variations observed by satellites at the monthly timescale show different patterns than temporally allocated bottom-up emissions in some areas. Peter Adams commented that this brings up the important question of how we combine bottom-up with top-down emission approaches. With current inventories and satellite products, are we at a point where satellite data and bottom-up emission inventories can be reconciled? The satellite is like having an emission factor measurement repeated over time but we need to better understand where the two correlate poorly or well and why.

#### Farm management practices and activity data

Marc Houyoux noted that in Peter Adams' presentation we heard the importance of knowing manure management practices. Knowledge of farm management practices is important to do the best job of creating emission estimates for localities. Regarding the importance of accurate activity data, Marc gave an example that before 2017 the dairy populations used in the EPA National Emissions Inventory (NEI) were far too low. In 2017, the NEI process better represented the populations and emissions increased and improved, especially in southern California. April Leytem followed up by agreeing that lack of activity data for facilities is a critical gap at the larger scale but at the facility scale knowing the differences in emissions and their processes between the housing and manure storage (i.e., management practices) is very important. Limei Ran (EPA ORD) commented that knowledge of manure application practices is an important uncertainty in the integrated air/soil/water models that EPA uses to understand agricultural impacts. For example, in the Soil and Water Assessment Tool (SWAT) we need to know manure application amount and timing, which we often don't know, to assess impacts of fertilizer runoff at the watershed scale. Ensuring that processes are properly integrated across models is also important. When we configure the Environmental Policy Integrated Climate (EPIC) model, we minimize manure application to avoid double counting of ammonia emissions from fertilizer in the bidirectional ammonia flux module within the Community Multi-scale Air Quality Model (CMAQ). We need to understand these uncertainties and limitations when using integrated models. Ian Rumsey reiterated that there are important uncertainties in emissions from manure applications, which, for example, can be as much as  $1/3^{rd}$  of total emissions from swine production facilities.

#### Approaches to improving activity data and challenges

John Walker (EPA ORD) noted that the need for improved activity data is key data/knowledge gap the keeps coming up in the discussion. This seems to be an example where engagement with agricultural stakeholders could help, and asked April Leytem what is a potential first step in pushing that question forward and which groups should be working together? April responded that it will be different for each industry. For example, poultry and swine are run by large integrators and they know all the details of on-farm activity and management practices. Management will be consistent from farm-to-farm and information could therefore be relatively easy to obtain. The beef industry is another example where the management practices are not very diverse, at least for feedlot operations, potentially making it easier to collect information from farmers. For example, if you know what the cows are eating you have a good idea of the emissions. They lose approximately 50% of the nitrogen they ingest on a feedlot. Emissions from backgrounding cattle (e.g., rangeland and pasture) are more difficult to characterize because there can be much more uncertainty in the amount of forage consumed and the forage quality. Accurate management information may therefore be more difficult to obtain. Furthermore, backgrounding cattle are a large percentage of total emissions from beef cattle. Dairy is more challenging because management may be specific to the individual dairy facility. Working at the state or regional level with individual dairy commodity groups, such as Idaho Dairyman's Association, would be one approach. Every state has a dairy commodity group that you could make contact with. For all of the sectors (i.e, dairy, beef, poultry, swine), developing relationships and building mutual trust with industry groups will be key to improving activity and management data.

### The panel was asked to discuss the role of ground-based monitoring in reducing uncertainties in emission inventories

Regarding the comparisons of bottom-up and top-down measurements, Marc Houyoux suggested that focusing an intensive ammonia measurement campaign in a specific area to link ground measurements to process models would be very useful for comparison with satellites. Viney Aneja asked Ian Rumsey if he could comment on the National Air Emissions Monitoring Study (NAEMS). Specifically, how do emissions measured under the NAEMS study differ from other emision data? Ian responded that those comparisons have not yet been developed but did recap the basic details of the study, noting that emission and ancillary measurements of ammonia, hydrogen sulfide, volatile organic compounds and PM were conducted over a period of two years at poultry, swine and dairy operations. Ian noted that while NAEMS is the most comprehensive emissions study to date, it did not include emissions from manure applications. Peter Adams remarked that NAEMS has been a significant step forward, particularly the fact that emissions were measured throughout the year. Winter emissions are important for PM modeling, even though emissions themselves are lower during winter compared to summer. NAEMS has more information and documentation on ancillary parameters like manure pH that other studies do not have. It is a good foundation to build on for future emissions studies.

#### Session 3: Spatial and temporal patterns of reactive nitrogen deposition

Jeff Collett (Colorado State University): Optimizing a ground-based reactive nitrogen monitoring strategy with respect to needs and resolution in a limited resource world

#### Introduction

Current U.S. monitoring programs have diverse objectives such as wet deposition, dry deposition, regional haze, and PM<sub>2.5</sub>. To optimize getting the most benefits for the scientific community, programs should be designed to be complementary and synergistic. This presentation summarizes existing monitoring networks and research needs for Nr deposition and explores how such optimization may be achieved in a limited-resource world.

#### State of the Science

Current network monitoring efforts provide precipitation concentrations and wet deposition fluxes (NADP/NTN), rural PM<sub>2.5</sub> and NO<sub>3</sub><sup>-</sup> (IMPROVE), urban PM<sub>2.5</sub>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> (CSN/STN), gaseous NH<sub>3</sub> (NADP/AMoN), and PM NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>and gaseous HNO<sub>3</sub> (CASTNET). Benefits of each network include the long-term record and good spatial coverage of NADP/NTN, the good spatial coverage with 24-hour time resolution of CSN and IMPROVE, and the gas measurements of CASTNET and AMoN. All of the networks have different objectives but there is some commonality in how the data are used, including source apportionment, model validation, and testing of air pollution control strategies. An important question is, how can we take existing networks and morph them to make them more complimentary and synergistic to better address broader objectives?

To investigate this question further, the networks were first examined in more detail, then some overall research needs for the field of Nr deposition were identified and discussed. Lastly, a few potential options for expanding monitoring network strategies to help address some of these needs were proposed.

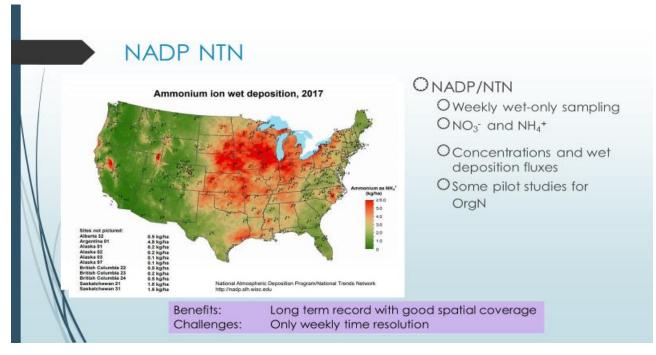
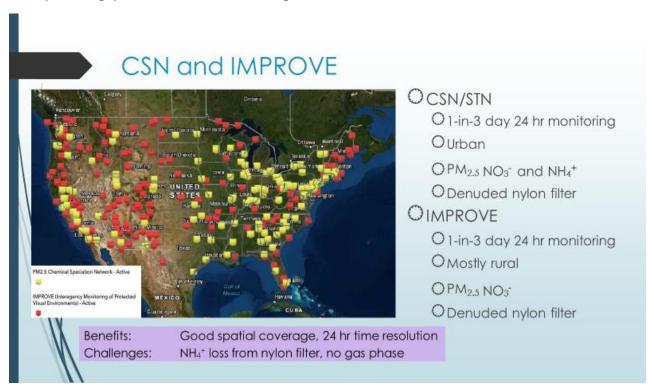


Figure 1. Location of NADP/NTN Sites in the contiguous United States

The NADP/NTN network (Figure 1) for wet deposition has good temporal and spatial coverage but the weekly time resolution is not ideal for source apportionment, particularly when there are multiple precipitation events in a weekly sample. For reactive N, organic species represent an important gap in the routine monitoring.



#### Figure 2. Location of CSN and IMPROVE Sites in the contiguous United States

CSN and IMPROVE focus on  $PM_{2.5}$  (Figure 2) and sample at higher time resolution. They have good spatial coverage but typically no collocated gas phase measurements and nylon filters are known to lose  $NH_4^+$ .



Figure 3. Location of CASTNET and AMoN Sites in the contiguous United States

CASTNET and AMoN (Figure 3) are referred to here as the dry deposition networks. They include gas phase measurements, which is a benefit, but have longer sampling durations than IMPROVE and CSN. Biweekly sampling makes source apportionment of AMoN samples particularly challenging. With respect to dry deposition, CASTNET NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> measurements are not size selective, thus the contributions from fine versus coarse fractions to deposition cannot be assessed. Volatility of NH<sub>4</sub> NO<sub>3</sub> on the CASTNET primary Teflon filter is a known issue.

The needs for reactive Nr monitoring were categorized under wet Nr deposition, dry Nr deposition, PM<sub>2.5</sub> speciation, source attribution, model validation and satellite validation. Three of these categories were further examined.

For dry Nr deposition, no network has a robust measurement of  $NH_4^+$ . We need to better understand the contribution of coarse  $NO_3^-$ , which we currently cannot assess from the CASTNET measurements. Gaseous  $NH_3$  is important but two-week integrated concentrations make determination of the bidirectional flux difficult. We need to understand how the ambient concentration, which is changing throughout the day, relates to the ecosystem  $NH_3$  compensation point. Hourly measurements are needed for bidirectional flux modeling. We need more measurements of organic N in both the particle and gas phases, recognizing that gases may have widely different deposition velocities.

Some of the same issues come into play for source attribution. Measurements of key species on relevant transport timescales are needed, which may vary by site. In some cases, 24-hour measurements may be sufficient while in other places hourly measurements may be needed. Changes in concentrations in Rocky Mountain National Park are associated with changes in transport from the west (downslope) versus the Front Range (upslope). Without high time resolution measurements, we may inaccurately apportion most of the NH<sub>3</sub> to sources west of RMNP since that is the predominant wind direction. In general, 24-hour measurements are more useful than weekly or biweekly measurements for source apportionment.

For model validation, collocation of gas and particulate species is needed. In a recent study, we were only able to identify 37 locations across the U.S. where collocated gas and particulate measurements were available. Measurement of the sum of gas and particulate species (e.g., total NH<sub>x</sub>) is also useful. Though you may not know the partitioning, at least you have the total for model validation. Filter based measurements of total NHx work well in the Rocky Mountain states but more recent measurements in the East suggest there may be artifacts under high humidity. Total gas + particulate measurements also represent an opportunity for cost savings where resources are not available for speciated measurements. As with source apportionment, the time resolution of the measurements should be consistent with the model process (e.g., transport, deposition, or particulate phase partitioning).

Figure 4 summarizes where we stand with respect to the strengths, weaknesses of current monitoring networks and future needs.

	Monitoring Need	Strengths	Weaknesses		
	Wet deposition	$\text{NO}_3^{\text{-}}$ and $\text{NH}_4^{\text{+}}$	No Org N Lack event resolution		
	Dry deposition	HNO₃	Lack size-resolved NO3 <sup>-</sup> Lack hourly NH3 Lack accurate NH4+ Lack Org N and NO2		
	PM <sub>2.5</sub> /regional haze	NO <sub>3</sub> -	Lack accurate NH <sub>4</sub> +		
	Source apportionment	24 hr NO3 <sup>-</sup>	Lack accurate NH4 <sup>+</sup> Lack <24 hr measurements of NH <sub>3</sub> , HNO <sub>3</sub>		
N	Model validation	Some co-located sites	Need more co-located sites Need accurate NH4+ Need better time resolution (<24 hr)		

Figure 4. Summary of the strengths and weaknesses of current monitoring efforts.

#### **Future Directions**

Addressing all of the current monitoring needs with limited resources will be difficult and there is no single solution. But there are some approaches that could move us in that direction, taking advantage of current monitoring infrastructure and existing and new technologies (Figure 5). For example, more sophisticated sampling strategies could be employed, such as event-based wet deposition sampling and denuder/filter pack samplers. Denuder/filter pack systems are labor intensive and expensive but can provide relatively high time resolution data (12 - 24 hour integrated samples) with accurate partitioning between gas and particulate phases. New technologies could be adopted for routine monitoring at higher time resolution using, for example, new generation aerosol mass spectrometers and continuous gas analyzers. These measurements are more expensive but the technologies have matured to the point that use for routine monitoring is feasible. Perhaps the most effective path forward is to build on existing monitoring infrastructure to create hybrid networks with fewer/simpler base network sites with collocated gas and particulate monitoring combined with multi-network super sites with a more sophisticated suite of high time resolution measurements to examine atmospheric chemistry and air-surface exchange processes in greater detail.

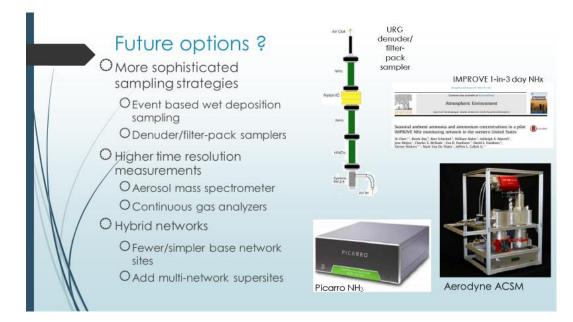


Figure 5. Possible future options for Nr monitoring.

Mark Shephard (Environment and Climate Change Canada): Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America

#### Introduction

With respect to reactive nitrogen (Nr), satellites currently provide measurements of ammonia (NH<sub>3</sub>) and nitrogen dioxide (NO<sub>2</sub>), which can be used to derive estimates of dry deposition. At the continental scale, deposition estimates from chemical transport modeling show that dry deposition contributes ~ 60% of total Nr deposition, with NH<sub>3</sub> and NO<sub>x</sub> dry deposition individually contributing  $\sim$  20% and 4%, respectively. These contributions from NH<sub>3</sub> and NO<sub>x</sub> translate to  $\sim$  32% and 6% of the dry deposited fraction of total Nr deposition. Based on these model estimates, satellites can help provide information on at least ~25% of the total Nr deposition, and ~40% of the more uncertain dry deposition component. The spatio-temporal coverage of satellites provides the opportunity to enhance our knowledge of atmospheric deposition through the fusion of the satellite observations with ground-based observations and air quality models. This presentation briefly describes the characteristics of satellite observations of NH<sub>3</sub> from the Cross-Track Infrared Sounder (CrIS) and NO<sub>2</sub> from the Ozone Monitoring Instrument (OMI) and recent applications combining satellite observations with modelled deposition velocities to derive atmospheric dry deposition of reactive nitrogen from these short-lived nitrogen species (Kharol et al., 2018; Shephard et al., 2020). The importance of satellite validation with ground-based monitoring is also discussed along with future research needs to improve satellite products for better understanding spatial and temporal patterns of Nr deposition.

#### State of the Science

In general, strengths of satellite remote sensing includes global spatiotemporal coverage, ability to fill in monitoring gaps (remote and inaccessible locations), the capability to re-process datasets with new algorithms (as technology improves), the detection of new sources and deposition locations, and transport between them, and time series analysis for trends and tracking of mitigation efforts. Some weaknesses include that not all surface pollutants of interest can be detected, temporal coverage is limited to 1 or 2 measurements per day and require cloud-free conditions, and vertical resolution is limited by the need to correlate total columns and surface levels with boundary layer values. In addition, the spatial resolution is ~ 5 to 10 km, and the data product is generally less precise than conventional monitoring. It is important to note that satellite remote sensing should be viewed as complementary to conventional monitoring and that its quality is dependent on robust validation with ground measurements.

The general characteristics of  $NO_2$  measurements from the OMI and  $NH_3$  measurements from the CrIS are shown in Table 1.

	Satellite	Spatial Resolution (km²)	Spatial Coverage	Temporal Coverage	Retrieved Quantity	Comments
NO <sub>2</sub>	Ozone Monitoring Instrument <b>(OMI)</b> (UV/VIS) (2004 – present)	13 x 24	Global	Daily ~13:30 (day)	Vertical column density (VCD) (molecs/cm <sup>2</sup> )	<ul> <li>ECCC has reprocessed NASA product to effectively improve resolution of VCDs (new AMF)</li> <li>McLinden et al., ACP, 2014</li> </ul>
NH <sub>3</sub>	Cross-track Infrared Sounder <b>(CrIS)</b> (2012-2038)	14 (diameter)	Global	Twice a day @ ~13:30 ( <b>day</b> ) ~1:30 (night)	Profile level volume mixing ratio values (ppbv)	<ul> <li>Limited vertical resolution</li> <li>Version 1.5</li> <li>Shephard et al., ACPD, 2019</li> </ul>

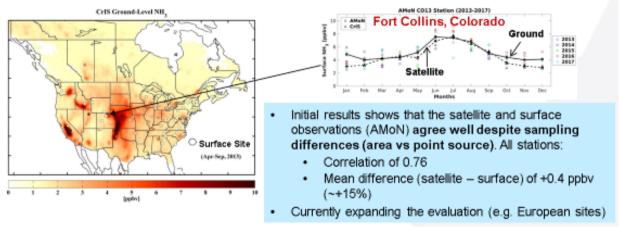
#### Table 1. General characteristics of satellite NO<sub>2</sub> (OMI) and NH<sub>3</sub> (CrIS) measurements.

In general, OMI NO<sub>2</sub> is a more mature product and is well validated. CrIS is a newer instrument that is most sensitive to NH<sub>3</sub> between 950 and 700 mb ( $\sim$ 0.5 to 3 km). Retrievals are not equally sensitive in the vertical and surface retrieved values are driven by sensitivity in the boundary layer. CrIS has a minimum detectability of  $\sim$ 0.5 ppbv under favorable conditions and random vertical column density errors generally range from 20 to 50%

Initial evaluation of results (Figure 1) comparing CrIS to 68 surface observations (AMoN & NAPS) shows overall good agreement (correlation of 0.76) despite the difference in sampling as approach (satellite is area average versus the specific location of a measurement point). Overall, there is a slight bias toward CrIS (~+15% or 0.4 ppbv), but at Fort Collins, CO, there is good agreement in the spring and summer and a bias towards the measurement in the winter months. NH<sub>3</sub> concentrations at Fort Collins are heavily influenced by local CAFOs, which may contribute to this bias. Agreement with surface observations tends to be stronger where emissions are spatially homogeneous over scales on the order of the satellite pixel and weaker where surface measurements reflect highly local sources.

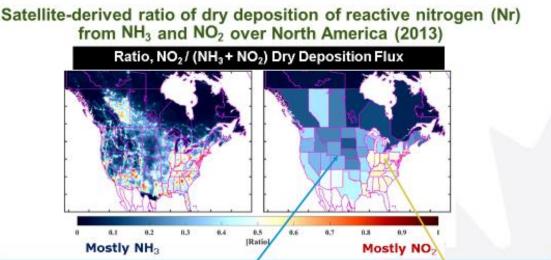
#### Satellite ammonia surface evaluation

- Satellite provides global coverage, but require validation
- North American surface networks:
  - 58 Ammonia Monitoring Network (AMoN) sites (USA) : bi-weekly
    - 10 National Air Pollution Surveillance Program (NAPS) sites (Canada): 3-day



#### Figure 1. Comparison of CrIS and surface observations of NH<sub>3</sub> concentrations.

To estimate dry deposition from satellite retrievals, the flux is determined by combining satellite derived surface concentrations with deposition velocities estimated from modeled meteorology and surface characteristics (e.g., big-leaf type deposition models, MODIS land-use data). Because CrIS provides a surface retrieval, the NH<sub>3</sub> concentration can be used directly. OMI provides a total column density of NO<sub>2</sub>, which must be combined with a vertical profile shape from an air quality model to determine the surface concentration. Results of the first application of satellite derived NO<sub>2</sub> (OMI) + NH<sub>3</sub> (CrIS) dry deposition for North America (Kharol et al., 2018) are shown in Figure 2.



- NH3 dry deposition flux peaks in agricultural and remote regions (e.g. Mid-West)
- · NO2 dry deposition flux dominates in urban regions (power plants) (e.g. North-East)

#### Figure 2. Map of the ratio of dry deposition of $NO_2$ to $NH_3 + NO_2$ across North America.

NH<sub>3</sub> hot-spots are mainly located over agricultural regions and NO<sub>2</sub> hot-spots are mainly located over densely populated cities and power plants. NH<sub>3</sub> is more dominant overall and will continue to increase in importance as NOx emissions continue to decline and NH<sub>3</sub> emissions are expected to increase. Examples of the use of satellites to characterize deposition hot-spots are shown in Figure 3.

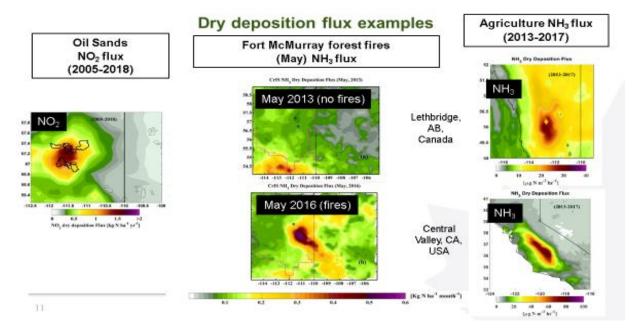


Figure 3. Examples of NO<sub>2</sub> and NH<sub>3</sub> dry deposition hot-spots derived from satellite measurements.

Large fluxes of NO<sub>2</sub> associated with NOx emissions in the Alberta Oil Sands region are observed as are emissions of NH<sub>3</sub> from the Fort McMurry forest fires. Broader analysis of NH<sub>3</sub> shows that northern latitudes affected by forest fires receive 2 to 3 times more dry deposition of ammonia relative to the local background. Examples of large NH<sub>3</sub> dry deposition fluxes associated with agricultural emissions in Lethbridge, Alberta (cattle) and the Central Valley of California (crop and animal production) are also shown.

Satellites are also useful for characterizing interannual variability and temporal trends in concentrations and dry deposition.

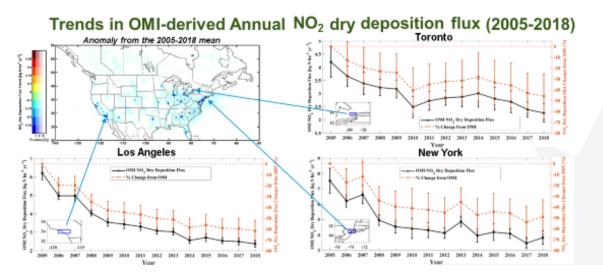


Figure 4. Trends in NO<sub>2</sub> dry deposition flux for Los Angeles, Toronto and New York derived from fourteen years of OMI data (Kharol et al., 2019, in preparation).

As shown in Figure 4, strong decreasing trends in  $NO_2$  dry deposition are observed over most of the eastern US and urban areas in response to regulatory efforts to reduce NOx emissions from vehicles and power generation over the past two decades.

#### **Future Directions**

More recent and future satellites will have increased measurement capabilities such as city scale and hourly observations over North America for NO<sub>2</sub>. The Tropospheric Monitoring Instrument (TROPOMI) was launched in 2017 and will provide daily (13:30 overpass) measurements of NO<sub>2</sub> and other gases at an improved spatial resolution of (5.5 x 3.5 km<sup>2</sup>). The Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument will be launched in geostationary orbit over North America in 2021. TEMPO will provide the first hourly observations of atmospheric composition from space, including NO<sub>2</sub>, and will have even higher spatial resolution (2 x 4.5 km<sup>2</sup>). Observations as well as increased spatial resolution for better characterizing deposition gradients around hot-spots.

Several examples have been presented that illustrate the potential of satellites to help derive estimates of dry deposition of Nr from NH<sub>3</sub> and NO<sub>2</sub>. The global spatial coverage is particularly valuable for remote regions that lack ground-based monitoring, and a consistent measurement approach can be used to track trends over various temporal scales. Initial satellite applications for Nr are promising, but the field is the still in the early stages of research and development. Future improvements to modelling, satellite observations, and validation data required to reduce uncertainties. These include:

• further refinement and harmonization of dry deposition schemes, which can yield flux differences of a factor of 2 or 3 for Nr species.

- more high-resolution surface measurements to create NH<sub>3</sub> diurnal concentration profiles in representative source areas over North America to convert satellite overpass measurement to daily average fluxes
- overall general need for flux validation data over various sources
- additional satellite retrieval improvements (e.g. NO<sub>2</sub> observations over fires, etc.)

Incorporating satellite measurements along with surface observations and output from chemical transport models in a measurement-model "data fusion" (MMF) framework for nitrogen deposition (e.g. NADP TDep MMF method; Schwede and Lear, 2014) should be investigated. Fusion of satellite data could be used in remote locations between surface monitoring locations and inform surface monitor "range of influence" concentration extrapolations needed in the TDep MMF method. Data could also potentially contribute to improvement of bias corrections between the measured and modeled values. Finally, satellites can also assist in the expansion of surface Nr monitoring by identifying key locations where additional monitoring would be most valuable.

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### Panel Discussion: Jeff Collett (CSU), Mark Shephard (ECCC), Mark Zondlo (Princeton University), Melissa Puchalski (EPA OAP), Randy Martin (USU)

The summary of the panel discussion is grouped into the following categories: audience questions for the presenters, panel discussion led both by moderator-posed questions and audience questions. Relevant questions and responses are grouped and summarized for clarity.

#### **Follow-up Questions for Presenters**

#### Optimizing ground-based reactive nitrogen monitoring studies

One of Dr. Collett's conclusions was to establish super sites as a means of optimizing existing networks. Jennifer Berry wanted a further description on the logistics of those super sites. Dr. Collett responded that super sites would have more flexibility (e.g. routine monitoring protocols) with respect to sites in existing networks (e.g. NADP, CASTNET). Typically, super sites are manned and could host more sophisticated equipment, which would allow for measurement of atmospheric chemistry and air-surface exchange processes at higher time resolutions (minimizing sample transformations during collection, storage, or shipment), and allow for greater speciation. There are many possibilities and the focus should be on getting the most information with the resources available. Justin Coughlin (USEPA, Region 9) noted that the NCORE network could be one of those possibilities as it is already established nationally and has both rural and urban aspects. Dr. Collett agreed that this would be a logical place to start.

Donna Schwede (US EPA, ORD) was interested in the sub-grid variability issue since modelers use area weighted averages for deposition estimates. However, a sampler, such as an AMoN sampler, is in one specific spot and a grid cell average may not be representative of what may be seen at the monitor. How would a super site address this issue? Dr. Collett was of the opinion that we need to do a lot more mobile measurements to get a handle on spatial variability. Mobile measurements are more critical in some regions such as those close to CAFOs. A network of samplers could be deployed in a relatively small area.

Melissa Puchalski commented that AMoN samplers had been deployed throughout Fort Collins and around the CASTNET Bondville, IL site. Spatial variability was not substantial within the grid cell at Bondville, which is surrounded by large areas of crop production, but strong variability was observed in Fort Collins related to emissions from CAFOs. Siting criteria need to be reevaluated for networks (CASTNET, AMoN, and NTN). Sites in these networks are traditionally intentionally located away from sources to characterize a regional signal, but NH<sub>3</sub> with its strong spatial variability was not considered in that siting criteria. Meetings have been held with the EPA OAQPS office to discuss deploying AMoN at NCORE and CSN sites.

#### **Panel Discussion**

#### Challenges of operating smaller networks

With high spatial resolution in mind, Randy Martin was asked to comment on the strengths and challenges of operating smaller sampling networks. Dr. Martin responded that when 40 Ogawa samplers were deployed along the Wasatch Front strong gradients were seen in all directions and it is important therefore to identify where these locations are. Since passive samplers do not capture diurnal variation, real time (i.e. high-time resolution) samplers should be deployed at select locations. Very strong diurnal patterns were seen across the network and the challenge is how to measure this, even seasonally. Dr. Martin sees development of new instrumentation as essential. Cheap sensors are coming online but do not have enough concentration resolution. He agreed that super sites are beneficial, but the issue of where to locate is significant. We need to look at areas were high spatial and time resolution would be needed.

Mark Zondlo was asked to further comment on advancements in high-time resolution measurement techniques. Dr. Zondlo responded there are a number of commercial sensors (e.g. Aerodyne, Picarro), and these have been deployed in mobile lab studies (e.g. San Joaquin valley in NASA-NSF joint study). Deployment of high- resolution instruments that measure in seconds or less is much needed for applications such as eddy covariance flux measurements. Zondlo's group (Princeton) has worked with open path NH<sub>3</sub> measurements, which have no sampling inlet and have found that open-path measurements averaged up to 30-min have matched Picarro data in terms of concentration.

The "stickiness" (i.e. propensity to adsorb onto sampling inlets) of NH<sub>3</sub> presents a challenge for accurate high-time resolution measurements, particularly below ten-minute sampling intervals.

An area where the NH<sub>3</sub> stickiness has presented a problem is to determine unbiased vertical profiles to validate satellite data. Picarro analyzers can be used in airplane measurements (e.g. made over Greeley, CO), but require short sampling durations and datasets suffer from biases. This issue was noted by both Mark Shephard, Mark Zondlo, and Kang Sun, and brought up in an earlier question from Gail Tonnesen.

This issue needs to be addressed with respect to deposition fluxes. An integrated approach will be needed. A lot can be learned from a combination of airborne measurements, mobile labs, flux towers, passive samplers, ground based measurements, and aqueous surface chemistry deployed at 10-100 km scales. Could be approached so the cost is not as large as bigger NASA/NOAA campaigns.

Leiming Zhang (ECCC) agreed that we need more data and super sites so models can be validated. The uncertainty in the measured flux data could be as large as that in the model. To illustrate the issue of uncertainty in the flux data, Leiming described a project comparing measured O<sub>3</sub> flux data at the Harvard Forest data (a long-term dataset from a flux 'supersite') among three flux measurement techniques (aerodynamic gradient, modified Bowen-ratio, and eddy covariance) and differences of a factor of 2 or 3 on a long-term scale. The point is that uncertainty in the flux measurements can be quite large. Because the models are developed from measurements, either directly or indirectly, we see a similar magnitude of uncertainty in model comparisons.

Leiming also noted the value in using aircraft flux measurements which integrate over much larger spatial scales compared to tower-based flux measurements. Although, if this level of uncertainty occurs at a ground-based site under conducive sampling conditions, what would the implications be for fluxes measured from an aircraft? The bidirectionality of NH<sub>3</sub> would also contribute to uncertainty.

Mark Zondlo mentioned that his group has investigated the bidirectionality issue at Duke Forest and with Jeff Collett at Rocky Mountain National Park, noting the value in time resolved measurements. At Rocky Mountain, Zondlo's group observed net deposition of NH<sub>3</sub> during upslope events that bring in NH<sub>3</sub> from the Front Range and re-emission back into the atmosphere during downslope events that bring in air with lower NH<sub>3</sub> concentrations from the west. Re-emission from dew was also observed by Jeff Collett's group. At Duke Forest, NH<sub>3</sub> uptake was seen in a hardwood forest canopy, with a strong diurnal cycle, while net emission was observed in an adjacent unfertilized grass field. There are different scales of fluxes, but these types of datasets are very helpful to improve bidirectional exchange models.

#### Prioritization of additional measurements. Where to start?

The moderator changed the topic to prompt a discussion on prioritization of monitoring needs of Nr referring to a slide presented by Jeff Collett listing some strengths and weaknesses of each (Figure 4, Collett summary).

Jeff noted that he did not rank the needs, which is challenging because of the different problems and considerations of importance from different perspectives (i.e. come things are easier to implement, some are important to NADP, others to other networks). Prioritization might be dictated by who leads the effort to prioritize? This question should be posed to the modelers as well. What is most important to them? As an example, is understanding the gas/aerosol partitioning of NH<sub>4</sub>NO<sub>3</sub> important, or will just using NHx be more helpful? We need to go through extensive analysis and debate to get there.

Bret Schichtel's (NPS) priority is high time resolution NH<sub>3</sub> measurements. Filter-based measurements of NHx could be cost effective and provide 24-hour integrated observations. Measurement of bulk organic N in precipitation can also be cost effective. Bret did not note any routine or semi-routine flux measurements in Dr. Collett's presentation. He wondered if more

complete routine (or semi-routine) flux measurements can be conducted in a super site set up? How important would this be relative to getting more complete surface concentration coverage at higher time resolutions?

Mark Shephard added that from a satellite perspective, the flux measurements and dry deposition velocities are one of the largest uncertainties and therefore a high priority.

John Walker mentioned that his group (building on the work of the Europeans) is developing a low-cost conditional time-averaged gradient system for making Nr flux measurements. The goal is not for hourly time-resolution, which is still needed for understanding processes, but to develop seasonal and annual deposition budgets. We should push forward these lower cost routine flux measurements to quantify budgets and characterize spatial patterns, but also we need to have some sites that implement the latest technology to directly measure air-surface exchange (i.e. fluxes) for model development.

Dr. Collett agreed and these kinds of abilities should be implemented at super sites. Having such an infrastructure would allow to bring in additional and collaborative research efforts to build up more detailed information for process-level studies. May need to be moved around to different locations to study different ecosystems.

#### How do we use satellite data to inform more ground-based monitoring of NH<sub>3</sub>?

John Walker as the moderator postulated that using satellite maps of air concentrations to identify areas of interest (e.g. hot-spots or elevation changes) where ground-based monitoring should go may be easier than securing funding for such a site, since the issue is not specifically agency-driven. Mark Shephard made a point about satellite and ground-based monitoring comparisons, stating that comparisons are generally better when concentrations are more homogenous. If a sensor is halfway up a mountain or near a CAFO, then comparisons will be poorer, and a ground-based measurement would be more beneficial.

It was noted that we need to communicate the NH<sub>3</sub> issue better as far as where the hot spots or areas of high concentration variability are located and why new ground-based sampling would be needed there. Satellites can be a help in this regard in terms of identifying hot spots and optimizing ground-based monitoring locations. Melissa Puchalski mentioned that this could be an example case of why other stakeholders should care about this issue and that this may be a good opportunity to reach out to them.

### John Walker to Randy Martin: How did you decide where to put your high-resolution samplers?

Dr. Martin responded that the strategy used was based on knowledge of the study area (Wasatch front) from previous research and then described some specifics. He first noted that the area was urban and made a point not to discount NH<sub>3</sub> from urban sources as well as agricultural. Previous work had identified areas that might be of interest (likely to exhibit diurnal patterns, or with high PM<sub>2.5</sub> concentrations). The selected sites included a near-roadway

(to capture mobile emissions), a notoriously high PM<sub>2.5</sub> location, and a location to provide information on transport between two air sheds. At each site, one to two weeks of hourly averaged data were collected with cooperation from EPA by using a mobile van to identify the hot spots as well as events (e.g. manure application).

Mark Shephard added that satellites can be used to validate ground-based measurements if an accuracy of 20 to 30% is acceptable, which is valuable for some applications.

#### **Audience Initiated Questions**

In reference to Donna's earlier comment on quantifying the difference between deposition in the model to multiple land-use types. Have you thought of using long-range open-path sensors (e.g. FTIR, DOAS) over a range of different land types (not homogenous) for high-resolution, spatially integrated measurements for a number of species? Could this approach provide information about spatial heterogeneity and how different land-use types effect concentrations?

Mark Zondlo was interested in following up but had not considered the direct question. This was something they potentially had the capability to do but would be a research project at this point. There are no commercially available sensors at this time.

Jeff Collett wondered whether satellite data can be used as another surrogate to look at individual days to ascertain spatial variability? How representative are super site measurements at a certain time of day? The technique may be powerful and the information should be accessible.

Mark Zondlo has performed satellite (IASI) oversampling at AMoN sites. Specifically, the top 10% of NH<sub>3</sub> column probability distributions were termed 'hot spots'. Out of 100 AMoN sites, 5 sites were within 100 km of the hot spots. So satellite maps will have a huge role to play. Melissa Puchalski agreed that there is a need for filling in surface monitoring gaps using satellites. She added that thinking about the issue from a budget perspective, we need to do a better job of communicating the NH<sub>3</sub> issue and tying it to policy or regulations to get resources to build out a more spatially extensive network and to improve temporal resolution to address the needs previously discussed.

#### Question for Mark Zondlo: We are resource constrained. NAAQS network wants to test out low cost sensors. How do we validate low cost sensors for NH<sub>3</sub> and what is the 'gold standard' calibration method?

Mark Zondlo responded that commercial sensors have made a huge step forward in the past ten years. Appropriate sampling protocols need to be established for low cost sensors. Existing biases and sampling limitations (i.e. don't try to sample 1-second or 1-minute data for rapidly changing concentrations with low cost sensors) need to be quantified. Value of data accuracy should be considered and balanced with cost. There is an opportunity to calibrate low cost sensors with current measurement technology.

## What information can ecologists provide to deposition scientists on where potential effects from excessive Nr deposition are happening? This information may assist in site placement or the types of measurements needed.

Jeff Collett responded that sites should be located in places where people (scientists, public) are interested in the results. Understanding ecosystem sensitivity impacts is very important. Deposition scientists need the ecologist's help in understanding the ecosystem characteristics as that can help to determine the flux, particularly with bidirectional NH<sub>3</sub> exchange. We need to work together to optimize monitoring strategies.

John Walker agreed and considered the cost of direct flux measurements. Pairing them with ecological or biogeochemical measurements helps to make the atmospheric measurements as valuable as they can be.

#### **Concluding Remarks from Panel**

Panelists were asked to give a closing remark and some of their high priorities on the topic of understanding spatial and temporal patterns of Nr deposition.

Mark Shephard thought that a data fusion type of product is needed to incorporate all of the measurement information available. Each method has its strengths. If we integrate all approaches, we will get the best result. Jeff Collett agreed and expanded the thought saying that merging the perspectives of different scientists, communities and networks will be needed to move forward.

Mark Zondlo also agreed and thought that funding agencies should work together in a more synergistic and collaborative way. His example of a data fusion project could involve satellites (NASA), agriculture (USDA), air quality and PM<sub>2.5</sub> (EPA), and process level research (NSF). The "one-off" efforts have been great and we've learned a lot, but there is a need to piece the whole picture together.

Another point pertaining to the supersites was made that we have co-located data available already that can be used. The example given was that there are ~25 sites situated away from hotspots with collocated gas  $NH_3$  (AMoN), gas  $HNO_3$ ,  $NO_3^-$ , and  $NH_4^+$  (CASTNET). That information could be used in  $PM_{2.5}$  modeling to determine if controlling NOx or  $NH_4^+$  would reduce  $PM_{2.5}$  more effectively.

Melissa Puchalski added that additional measurements for flux model parameters (e.g. soil samples, pH, NH<sub>4</sub><sup>+</sup> in vegetation) should be co-located at the sites of the atmospheric measurements.

Randy Martin also agreed and reiterated the need for more measurements with better time and spatial resolution. He thought that low-cost sensors should be considered, and while they require extensive and frequent calibrations, they can be valuable. The monitoring community should not give up on them.

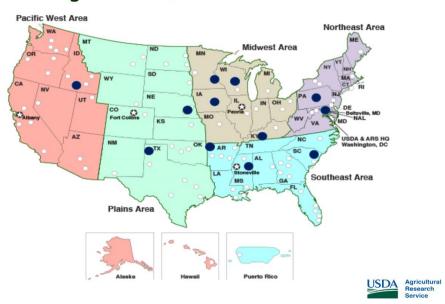
#### Session 4: Federal stakeholders

Peter Vadas (USDA ARS): Ammonia research in USDA-ARS: Objectives, accomplishments, and future directions

#### Introduction

As the National program leader for Land and Air for U.S. Department of Agriculture Agricultural Research Service (USDA ARS), Peter Vadas gave a broad overview of the agency with a focus on the type of NH<sub>3</sub> research that is conducted and welcomed feedback on how ARS could meet the need of stakeholders, including NADP.

The ARS is the principal research agency of the USDA and has four national research programs including Animal Production, Crop Production, Natural Resources, and Nutrition and Food Safety. The agency is distributed across 90 locations across the U.S. with 12 locations that focus on NH<sub>3</sub> research (Figure 1).



#### NH<sub>3</sub> Research – 12 locations

Figure 1. USDA ARS locations (white dots) distributed across the US (in five regions) with research locations primarily focused on agricultural NH<sub>3</sub> research highlighted (blue dots).

Most of the research relevant to the TDep Agricultural Workshop is in the Soil and Air research program (a sub-program of the Natural Resources national program), though there is a lot of integration and collaboration across programs. The agency's research is organized into 5-year research cycles, with a new cycle beginning in 2020. This presents an opportunity to evaluate

the needs of our stakeholders (including this workshop) moving forward and formally incorporate them into research objectives and experiments for the next 5-year cycle.

#### State of the Science

Peter Vadas then summarized current research objectives and experiments conducted on agricultural NH<sub>3</sub> emissions. ARS is mindful of supporting national emissions assessments as well as producer needs. Research objectives were broadly defined as 1) quantify NH<sub>3</sub> emissions and fill emissions data gaps with real-world information, 2) develop practices and technology to reduce emissions, and 3) support deposition measurements for national assessments. More specific projects were identified to measure emissions and deposition from animal production facilities and manure application methods, evaluate technologies to capture NH<sub>3</sub>, assess the impacts of animal diets on manure emissions, develop field to farm scale modeling, and assess the accuracy of emissions measurement methods.

The first highlighted research was in Florence, SC on whole farm swine emissions where a manure treatment system was found to reduce emissions from 0.8 to 2.64 g s<sup>-1</sup> to < 0.06 g s<sup>-1</sup> (approximately a factor of 10 to 40). Techniques to recover NH<sub>3</sub> include the usage of gaspermeable membranes in poultry barns and vegetative buffers surrounding farm operations to disperse and reduce emissions (reported as 20 to 50%).

Locations in the Southeast (e.g. Fayetteville, AR; Mississippi State, MS, and Bowling Green, KY) are focused on poultry operations for monitoring farm emissions. These focus on the whole production system and attempt to allocate emissions to specific farm components (emissions originating from livestock houses, manure application, etc.). For example, poultry production in AR has ~1.2 billion broilers with an estimated 46 g NH<sub>3</sub> bird<sup>-1</sup> loss (80% from houses), resulting in annual emissions of ~50 million kg NH<sub>3</sub>.

Studies of emissions control strategies (e.g. alum application, acid scrubbers) are exemplary of agency objectives to improve animal health and improve productivity. Research is focused on "win-win" scenarios, especially. Many of these control strategies are effective but expensive, and the agency is looking for ways to reduce cost of implementation and thus make the approaches more applicable.

### **Animal Facilities**

#### Fayetteville, AR Mississippi State, MS Bowling Green, KY



Emissions from poultry production. 46 g  $NH_3$ /bird loss from house to manure application. 80% from houses. 1.2 billion broilers in AR = 50 million kg  $NH_3$  emitted each year.

> Alum application to litter reduces NH<sub>3</sub> emission ~50%.





Acid scrubbers to capture NH<sub>3</sub>. 55% capture, but not cost effective. Develop biological scrubber with nitrifying bacteria to reduce costs.

Other innovations: Ventilated flooring, use of biochars

#### Figure 2. Research on Animal Facilities occurring in the Southeast ARS research locations. Focus is on quantifying emissions and to assess the effectiveness and applicability of NH<sub>3</sub> emissions control strategies.

ARS research at Ames, IA is focused on emissions from swine and poultry facilities. Investigating N mass balances and the impacts on animal performance. A wet air scrubber was effective at reducing NH<sub>3</sub> (25 to 35%), dust (80 to 90%), and odors (30 to 70%). Studies focus on cattle (beef and dairy) at the Southern Plains ARS locations (Bushland, TX and Clay Center, NE). Here, it was found that ~50% of N in feed is lost as NH<sub>3</sub> and researchers are looking at the effectiveness of manure additives (e.g. alum, lime, urease inhibitors, humates, zeolites, etc) to help reduce those emissions. These have found to be effective, but cost may limit its applicability.

Studies in Kimberly, ID are focused on dairy emissions. Researchers are quantifying NH<sub>3</sub> emissions from the wide variety of facilities that exist (e.g. open lot, open free stall flush) and also attempting to attribute emissions and their temporal variability to specific on-farm locations (e.g. lots, waste ponds, compost, barns, etc.; Figure 3).

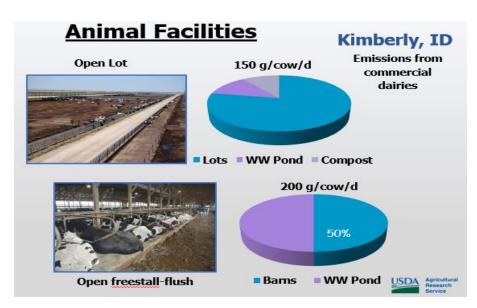
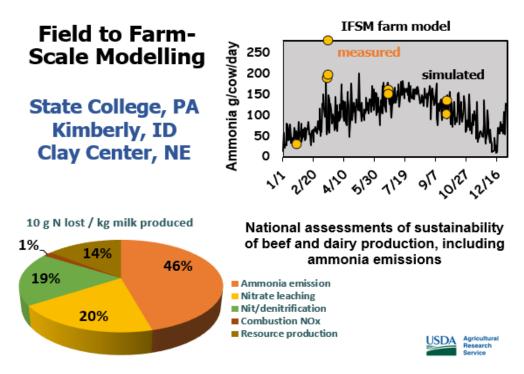


Figure 3. Research on different types of dairy cattle facilities in Kimberly, ID attempt to quantify NH3 emission and allocate them to specific source locations.

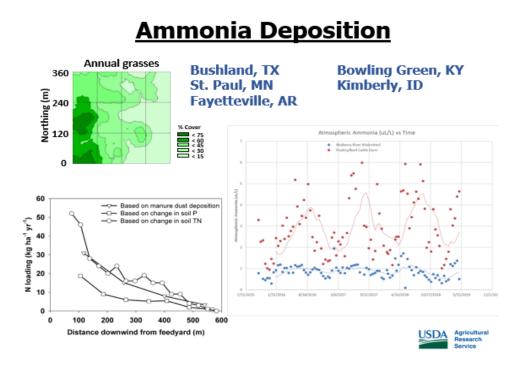
Studies also consider the impact animal diet formulations have on emissions. Small reductions in crude protein (CP) in feed can lead to large reductions in NH<sub>3</sub> emissions. A 3% decrease in CP lead to a 53% reduction in NH<sub>3</sub> emissions reported for dairy cattle. For swine and poultry diets, each 1% decrease in CP lead to a 10% reduction in NH<sub>3</sub> emissions. Reduction in feed and NH<sub>3</sub> represents a cost-savings to the farmer as well.

There is a lot of focus on manure application methods, the objective is get the manure under the soil surface to reduce volatilization. Looking at effects of different methods on different land types (pastures, crop fields, etc). Injection and incorporation of manure greatly decreases NH<sub>3</sub> emission, but there can be tradeoffs, so there is an attempt to consider the whole production system. The goal is to find an optimal balance of issues. For example, we need to understand how reducing NH<sub>3</sub> emissions and keeping it in the soil will affect N content in the soil, nitrate loss, or N<sub>2</sub>O emissions. There may be a trade-off of benefits. ARS also considers the impacts of NH<sub>3</sub> reduction efforts to other conservation issues such as reducing erosion.



#### Figure 4. Example analyses of outputs from the ARS Integrated Farm System Model (IFSM)

ARS also works on modeling farm systems. The Integrated Farm System Model (IFSM) was developed for national assessment of the sustainability of beef and dairy production. The model simulates the whole-farm system and considers NH<sub>3</sub> emissions, N and P cycling throughout different farming operations (e.g. feeding, animal housing, manure application). ARS experimental data is used to test and validate the model (Figure 4). Figure 4 also shows an example of analysis of modeled output data to estimate the relative pathways of N lost per unit of farm productivity.



#### Figure 5. Plots showing highlighted results of N deposition studies conducted by the ARS

Ammonia deposition is also researched at ARS, two examples were highlighted. The first was a study in Bushland, TX, looking at N deposition downwind of a beef feedyard (Figure 5, lower left plot) and impacts to vegetation cover in the surrounding prairie (Figure 5, upper left plot). The second is a study assessing NH<sub>3</sub> deposition to the Mulberry River watershed in AR (Figure 5; right plot).

Lastly, the ARS often collaborates to research for National Assessments. Examples given were the work on development of the NADP AMoN  $NH_3$  network, and the contribution of passive  $NH_3$  sampler monitoring data in the Cache Valley, UT atmospheric monitoring intensive.

#### **Future Directions**

ARS research will continue to focus research on livestock NH<sub>3</sub> emissions, as they represent a large source and are inadequately characterized especially at the farm-level. "Real-world" experiments on diverse, working farms will continue to quantify magnitude, variability, and origin of NH<sub>3</sub> emissions. Research efforts will continue to understand where the NH<sub>3</sub> produced ultimately deposits within and downwind of the region.

Efforts will be made to utilize measurements in models. Specifically, measured data on variability in management, weather, and soil that can impact emissions is needed. There are many research needs, some mentioned in the prior session of this workshop, that are good

opportunities for collaboration and ARS research can be helpful. Models include the larger scale models that EPA uses to estimate emissions, deposition, and reaction of NH<sub>3</sub> in atmosphere, but also include the smaller farm-scale models which help provide farmers real-world data and options and cost/benefit estimates for emission reductions and better production.

More research will be targeted at the development and evaluation of emissions reductions strategy and technology. Many effective techniques are known, but expenses are currently too high to be feasibly deployed on working farms. Research is planned to develop cost-effective emission reduction technologies.

Lastly, the accuracy and cost of emissions measurement techniques remains a challenge and new techniques and methods will continue to be assessed.

Jim Cheatham (NPS): Partnership to reduce nitrogen deposition impacts in Rocky Mountain National Park

#### Introduction

Jim Cheatham (National Park Service; NPS) presented an overview of the collaborative Rocky Mountain National Park (RMNP) Initiative to address air pollution and its impacts on the Park. RMNP is known as the "land above the trees". Approximately three-quarters of the 415 square mi park is above 9000 feet where high elevation subalpine and alpine tundra ecosystems developed under low nutrient conditions, with typically thin soils and low rates of plant growth, are more vulnerable to excess reactive Nitrogen (Nr) deposition than other ecosystems in the US or other countries (Figure 1).

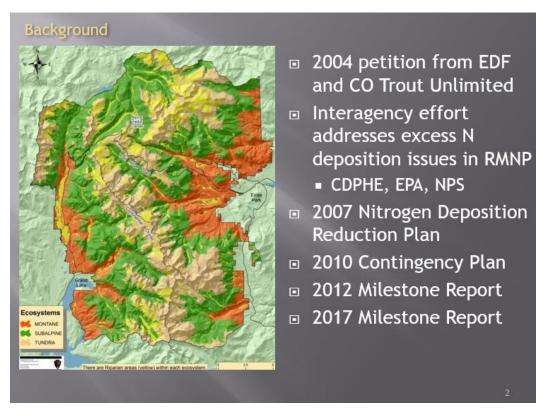


Figure 1. Map of Rocky Mountain National Park (RMNP) showing the subalpine and alpine tundra ecosystems which are sensitive to excess nitrogen deposition. The outline is a rough timeline of the RMNP Initiative.

The RMNP Initiative was formed in 2005 and resulted from a 2004 petition to agencies (NPS, Environmental Protection Agency; EPA, Colorado Department of Public Health and Environment; CDPHE) from the Environmental Defense Fund and Colorado Trout Unlimited. The agencies have a Memorandum of Understanding to pursue review of air pollution in RMNP and a course of action to address them. A rough timeline of the RMNP Initiative is shown in Figure 1, the 2007 Nitrogen Deposition Reduction Plan (NDRP) was endorsed by the Colorado Air Quality Control Commission along with the 2010 Contingency Plan (CDPHE, 2007). The Contingency Plan outlines corrective measures that will be implemented in the event that interim deposition goals described in the original NDRP are not realized. There have been two Milestone reports in 2012 and 2017 (CDPHE, 2014; CDPHE, 2019). While impaired visibility and elevated ozone levels are air quality concerns at RMNP, the Milestone Report focuses only on nitrogen deposition. Ozone and regional haze are addressed through other Clean Air Act processes. However, these processes do contribute to nitrogen oxide (NOx) reductions, reducing nitrogen deposition in RMNP as well. The NOx reductions that will occur in the future because of these programs are considered in the report and will be addressed by the MOU agencies in the future.

#### State of the Science

Nitrogen deposition happens everywhere but the Park's high elevation resources are especially susceptible due to their evolution under low nutrient conditions and limited capacity to buffer the chemical effects of excess nitrogen. Greater precipitation in the mountains also means more nitrogen is deposited by rain and snow. Thin soils and slow plant growth can't use all of the nitrogen which then runs off into mountain streams and lakes.

Figure 2 shows the observable ecosystem impacts (x-axis) that are expected to occur at specified Critical Loads of wet or total N deposition. The y-axis indicates the deposition loads at different levels (background, target, current). The solid grey box indicates those impacts which have already been observed in RMNP based on the current N deposition of 3.3 kg N ha<sup>-1</sup>yr<sup>-1</sup> (wet) or 4.9 kg N ha<sup>-1</sup>yr<sup>-1</sup> (total). The dotted grey box indicates the impacts which could occur at higher nitrogen loads, but are not currently occurring. The 'Target deposition' load of 1.5 kg N ha<sup>-1</sup>yr<sup>-1</sup> wet (~2.3 kg N ha<sup>-1</sup>yr<sup>-1</sup> total) has been adopted and endorsed by participating agencies as a resource management goal for 2032. This target load will be based on measurements from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) Loch Vale site.

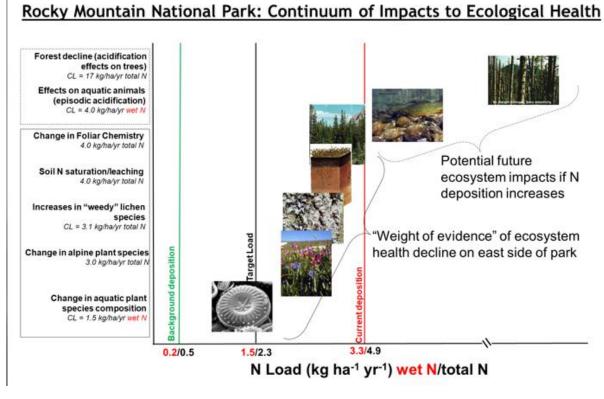


Figure 2. Continuum of observable ecosystem impacts expected for Critical Loads (CL) of N deposition (specified as wet and/or total loads).

The MOU agencies compared trends in wet nitrogen deposition data from Loch Vale to other NADP/NTN sites on the eastern slope of the Front Range that are exposed to similar emissions: a lower elevation site in RMNP (Beaver Meadows) and three sites located outside of the Park. The sites at Niwot Saddle and Sugarloaf are located in the mountains southeast of Loch Vale and complement each other as high/low elevation sites like Loch Vale and Beaver Meadows in RMNP. The site at Pawnee is much lower in elevation and provides additional regional context. The annual wet N deposition (grey bars) observed at Loch Vale and the 5-year rolling average (red) is shown along with the measured precipitation (blue shade) and the target RMNP Initiative glidepath (green) in Figure 3.

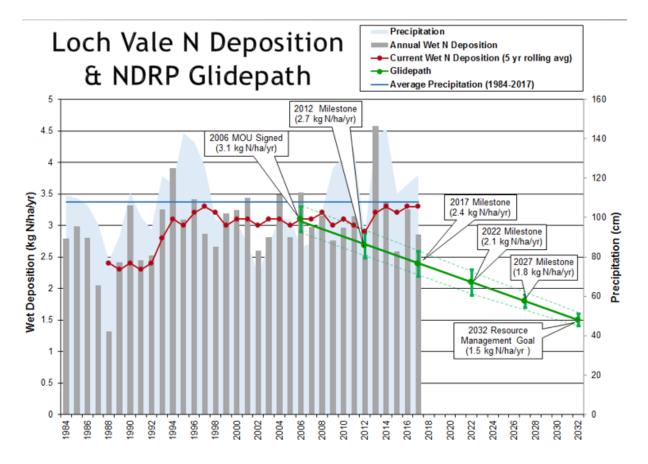


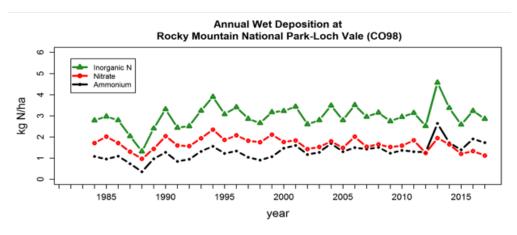
Figure 3. Time-series of annual wet N deposition and precipitation measured at Loch Vale, CO from 1984 to 2017. Five-year rolling average and targeted RMNP Initiative glidepath for annual wet N deposition is shown in red and green, respectively.

A significant increasing trend was noted for the 1984-2000 period for the Loch Vale site. With the addition of 2010-2012 data, this trend was no longer statistically significant at Loch Vale. This remains the case during the 2012-2017 time period. While this does not meet the goal of reaching a decreasing trend in wet nitrogen deposition, or hitting the milestone exactly, it is a shift in the right direction.

A notable feature of Figure 3 is the departure of the 5-year rolling average from the glidepath which remains a challenge. Also notable is the abnormally high wet deposition value for 2013 (4.6 kg N ha<sup>-1</sup>yr<sup>-1</sup>), which was attributed using back-trajectory analysis to a high percentage of transport over high emission source areas in Northeast CO.

Another issue is the ability to accurately estimate N deposition in high-elevation areas including at the Loch Vale site. Extreme weather conditions can result in 30 to 40% of the precipitation data being invalid, and the NADP approach is to replace missing data with an annual mean concentration of valid samples. Schichtel et al., 2019 found that this approach leads to high-biased annual deposition rates due to seasonal and precipitation-based dependencies and developed an alternate data substitution method which accounts for this dependence.

The speciation of the Loch Vale N deposition is shown in Figure 4 and reflects NADP/NTN network wide patterns (Li et al., 2016) that reduced N (NHx) levels are increasing while oxidized N (NOx) levels are decreasing. As of 2012, the majority of the inorganic wet N deposition is from reduced N (NH<sub>4</sub><sup>+</sup>).



Mean Annual Precipitation Weighted Concentration at Rocky Mountain National Park-Loch Vale (CO98) 2 Nitrate 5 neq/l 9 ŝ 1985 1990 1995 2000 2005 2010 2015 year

# Figure 4. Annual wet deposition of total inorganic N, nitrate N, and ammonium N at Loch Vale, CO NADP/NTN site from 1984 to 2017 (top). Mean annual precipitation-weighted concentrate at Loch Vale, CO NADP/NTN site (bottom)

The 2014 National Emissions Inventory (NEI) from the Colorado Denver Metropolitan area/North Front Range area (nine counties) shows that the largest sources of NOx are highway vehicles (38%), electricity generation (15%), off-highway vehicles (13%) and oil and gas production (8%). NOx emissions from all sectors are declining except oil and gas related activities.

The NEI data for the same area indicate the largest sources of NHx are livestock waste (70%), fertilizer (10%), and highway vehicles (8%). Statewide, livestock percentages are higher than national estimates with fertilizer being the second greatest contributor to ammonia emissions. Since ammonia emission inventories are being continually assessed and analyzed, it is important to focus on the largest source categories rather than the specific percentages. All of the emission inventories agree that the significant source categories are livestock and fertilizer.

The inventories do not necessarily agree on the smaller categories at this point, but these categories contribute a minimal percentage of emissions. They do include highway vehicles, other fuel combustion, fire, and waste disposal methods.

In addition to the NEI data, important demographic trends need to be considered. The Denver Metro population is increasing (including vehicle miles travelled). Data from the National Agricultural Statistics Survey show that the total head of cattle, sheep, and swine have remained fairly stable since 2008 although Colorado's total slaughtered live weight cattle and milk production have increased. The agricultural industry can claim that this represents an efficiency, but the beef and milk production can relate more directly to ammonia emissions.

Jim stated that the facility-specific emission factor (the difficulty in accurately estimating this was discussed in the morning session) would be very helpful to better understand what is going on, but without that information, the strategy is focused on what the producer can do as emissions can be tempered by the best-management practices (BMP) that the facility is implementing.

The RMNP Initiative strategy with Colorado agriculture is to assess the agricultural NH<sub>3</sub> BMP use for livestock and crop production. With limited resources, the focus is on Larimer and Weld counties (highlighted green in Figure 5) due to their high agricultural production, proximity to RMNP, and meteorology that is conducive to transport to RMNP. The assessment begins with a recommended list of BMPs for NH<sub>3</sub>, followed by surveying producers on their current BMPs and comparing their results with the recommendations. Identifying potential areas for improvement can help both agency and agricultural efforts to overcome barriers to broader BMP implementation, and finally to repeat the process over time to track BMP progress.



*Figure 5. Map showing RMNP and warning frequency from the CSU Early Warning System (EWS).* 

A specific example of a recommended BMP is the early warning system (EWS) which is managed by CSU. Under the right meteorological conditions for transport into RMNP, warnings are distributed to agricultural producers via emails or texts to suggest that they temporarily curb or avoid high emitting activities until those transport conditions pass (usually for 1-2 days). Assessing NH<sub>3</sub> BMPs is part of the agricultural industry's 5-year strategy spanning from 2019 to 2023 and consists of the EWS, research, monitoring, and outreach.

#### **Future Directions**

The collaboration of the RMNP Initiative agencies and the CO agriculture industry (shown in Figure 6) is an excellent example of a cooperative strategy to meet the common goal of reducing N deposition from agricultural sources to RMNP. It is important to gauge what outcomes are necessary in order to determine if that strategy will be successful. The first needed outcome is that the collaboration between the agencies and agriculture is on-going. That collaboration should fill data gaps in assessing BMPs and the increase and broad implementation of BMPs (e.g. EWS) should be evident. It is important that the agriculture industry's 5-year strategy is implemented. If all these outcomes are achieved, then then the

overall goal of reducing N deposition in RMNP (particularly from agriculture) to target levels is achievable.



## Figure 6. List of collaborating agencies and Colorado agriculture organizations participating in the RMNP Initiative.

There will be additional hurdles in reaching target levels from the increased NOx emissions in the Denver Metropolitan Area resulting from increasing population and vehicle miles traveled (expected 37% increase from 2016 to 2040) trends. There are numerous state and federal NO<sub>x</sub> emission control programs that have been recently implemented or have been scheduled to be implemented over the next 20 years, including controls for existing and new engines, Regional Haze controls on specific sources, of which the majority are power plants, federal on-road vehicle standards, and an enhanced inspection and maintenance program in the Front Range. CDPHE plans to address the increase in oil and gas NOx emissions through ozone SIPs and the next round of Regional Haze planning.

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Gail Tonnesen (EPA Region 8): Need for improved ammonia emissions and ambient monitoring data for modeling PM<sub>2.5</sub> and regional haze

#### Introduction

Ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) aerosol, which forms when NH<sub>3</sub> reacts with nitric acid (HNO<sub>3</sub>), is the largest component of PM<sub>2.5</sub> in Utah and California PM<sub>2.5</sub> non-attainment areas and is an important contributor to anthropogenic visibility impairment. Air quality planners and modelers require accurate NH<sub>3</sub> emissions inventories for modeling of PM<sub>2.5</sub> and regional haze to meet Clean Air Act and National Environmental Policy Act (NEPA) planning goals. For example, states are required to submit model attainment demonstrations that identify emissions reductions needed to attain PM<sub>2.5</sub> National Ambient Air Quality Standards (NAAQS). States must also submit Regional Haze State Implementation Plans (SIPs) in 2021, and regularly thereafter, that show reasonable progress in reducing visibility impairment at Class I areas. Furthermore, as required under NEPA, Federal Land Managers use air quality models to assess air quality impacts, including atmospheric deposition, caused by new development on federal lands. This presentation outlines several examples of research and monitoring needed to improve Chemical Transport Models (CTMs) used in these assessments.

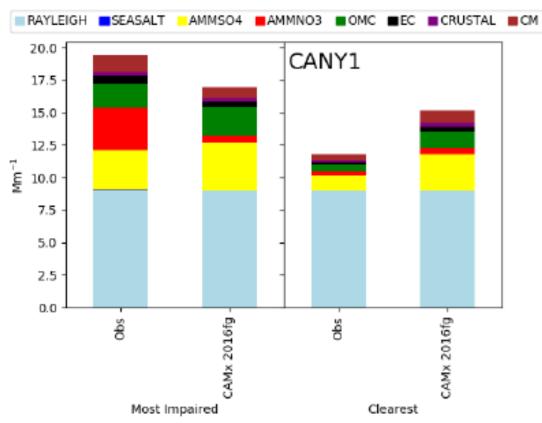
#### State of the Science

Ammonia is a key precursor to NH<sub>4</sub>NO<sub>3</sub> and affects aerosol acidity, which can affect conversion rates of sulfur dioxide to sulfate. It is particularly important to identify conditions in which NH<sub>4</sub>NO<sub>3</sub> formation is NH<sub>3</sub> versus NOx or oxidant limited. This requires accurate estimates of emissions for both oxidized and reduced forms of nitrogen, which is problematic in many areas due to uncertainties in NH<sub>3</sub> emission inventories. For example, the state of Utah has conducted modeling for the winter PM<sub>2.5</sub> SIP for the last 10 years. Results suggest that NH<sub>3</sub> emissions in the Cache Valley may be underestimated by a factor 10. These emissions must be artificially added back to the model to simulate winter NH<sub>4</sub>NO<sub>3</sub> episodes during inversions.

In addition to accurate specification of emissions, models must accurately simulate the other processes (meteorology, chemistry, deposition) that govern atmospheric concentrations of NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup>. Evaluation of model performance is challenging due to the need for collocated measurements of both NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup>. Datasets of continuous hourly measurements are relatively limited for both species. Monitoring networks employ methods that integrate air concentrations over periods of 24 hours to two weeks. However, many of the processes that drive ambient air concentrations are important at the hourly time-scale, making it difficult to interpret differences between models and measurements that average variability over days to weeks. Re-emission of NH<sub>3</sub> during morning evaporation of dew (Wentworth et al., 2016) is an example of a process that imparts diurnal variability in NH<sub>3</sub> concentrations. Hourly NH<sub>3</sub> concentrations in environments where this process is important. Satellites provide NH<sub>3</sub> data at shorter time-scales (during overpass) and with better spatial coverage than monitoring

networks but lack sensitivity at low NH<sub>3</sub> concentrations relevant to regional haze planning. Current NH<sub>3</sub> satellite products may therefore be less useful in Class I areas where concentrations are often low.

Results from the 2011 Western Air Quality Study model performance evaluation (<u>http://views.cira.colostate.edu/iwdw/</u>) illustrate the difficulties in modeling NHx in the western U.S. Across the 12 NADP/AMoN sites within the modeling domain, CAMx and CMAQ showed normalized mean biases of -70.3% and -62.2%, respectively, relative to observations of NH<sub>3</sub> air concentrations (mean observed = 1.22 ppb). Monthly statistics showed that the negative modeling bias persisted throughout the year in both models. Underestimation may be driven by a combination of bias in emissions and uncertainty in other model processes such as deposition.



Results from a 2016 EPA model evaluation of CAMx for regional haze shown in Figure 1 illustrate some of the challenges in speciating  $PM_{2.5}$  within CTMs.

Figure 1. Observed and modeled (CAMx) light extinction (Mm<sup>-1</sup>) at Canyonlands National Park (UT) on the 20% most impaired and 20% clearest days.

At Canyonlands National Park, the model substantially underestimates NH<sub>4</sub>NO<sub>3</sub> (red fraction in Figure 1) on impaired days though model performance is similar to observations on cleanest

days. It was noted that this is the site with the worst performance for  $NH_4NO_3$ . The model tends to overestimate  $NO_3^-$  over much of the U.S., so in these simulations the bidirectional flux of  $NH_3$ was turned off and the surface resistance to  $NH_3$  deposition was set to zero. This would result in higher deposition rates of  $NH_3$ , which leaves less  $NH_3$  in the atmosphere to form  $NH_4NO_3$ , thereby reducing the positive  $NO_3^-$  bias. However, this has the effect of making the negative  $NO_3^-$  bias worse at places like Canyonlands and throughout the Colorado Plateau. Part of the negative bias may be related to the model tendency to overestimate sulfate (yellow fraction in Figure 1), which is scavenging  $NH_3$  that would otherwise react to form  $NH_4NO_3$ . From the measurement perspective, it may be possible that a fraction of the observed  $NH_4NO_3$  at Canyonlands may in fact be  $CaNO_3$ , which is only now being added to the model. This uncertainty could be informed by speciated measurements of aerosol  $NO_3^-$ .

#### **Future Directions**

Data and knowledge gaps must be addressed in several areas to improve the ability of CTMs to:

- simulate NHx and PM<sub>2.5</sub> concentrations in support of SIP development and NAAQS assessments
- determine if PM<sub>2.5</sub> formation in urban and remote areas is NH<sub>3</sub> limited
- develop improved estimates of NHx deposition in Class I and sensitive Class II areas
- evaluate the air quality benefits of large-scale implementation of best management practices for mitigating NH<sub>3</sub> emissions.

More work is needed on the development of emission models and emissions inventories for NH<sub>3</sub>. Specifically, incorporation of fertilizer NH<sub>3</sub> emissions from the Environmental Policy Integrated Climate model (EPIC) modeling would improve this category in the EPA National Emissions Inventory (NEI). Additional collaboration with USDA on emissions and associated activity and management data will be needed to improve NH<sub>3</sub> emissions from animal sources in the NEI. Continued development of best management practices for reducing NH<sub>3</sub> emissions from agricultural sources is also needed.

More work is also needed to improve NHx deposition models, specifically bidirectional NH<sub>3</sub> flux parameterizations and estimates of dry deposition near sources. There is a need for more measurements of the diurnal profile and vertical profile of NHx, as well as flux measurements, to understand the roles of deposition and re-volatilization of NHx. Continuous time-resolved monitoring of collocated NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> is needed, if not in a routine mode then at least during intensive campaigns. Further development of source apportionment tools for atmospheric NHx is needed, as well as improved understanding of the influence of meteorological events on NHx deposition and aerosol processes.

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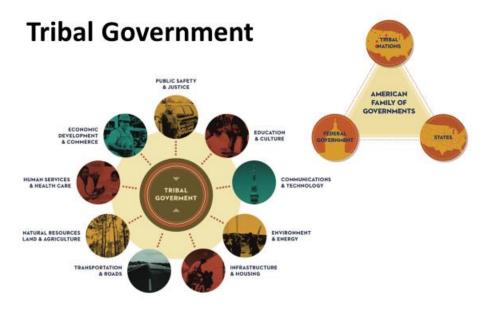
Daniel Cornelius (Intertribal Agricultural Council, University of Wisconsin-Madison): Native American agriculture and natural resources: Impacts relating to atmospheric deposition

#### Introduction

There are 573 federally recognized Indian tribes with 56 million acres in trust. 90 million acres were lost between 1887 and 1934. Tribal lands include some of the most diverse and ecologically sensitive habitats on the planet, creating opportunities for partnerships between Tribes, National Monitoring Programs, Federal and State Agencies, and Academia to assess environmental impacts of atmospheric deposition. Agriculture is a key aspect of tribal economies. Thus, tribal nations have an interest in better understanding the impacts of agriculture to tribal lands, including atmospheric deposition.

#### State of the Science

The diversity of tribal government functions is illustrated in Figure 1. Tribal governments are stretched in many directions but land and resource management is a key priority. Over the past 30 years much more capacity has been developed in that regard.



From NCAI Report - Tribal Nations and the United States, 2019.

#### Figure 1. Overview of tribal government functions.

Agriculture is an important component of tribal economies. According to the USDA 2012 Agricultural Census, there are 71,947 food producers farming approximately 57 million acres of tribal land. Sales of agricultural commodities are \$3.24 billion annually as livestock (\$1.8 billion) and commodity crops (\$1.4 billion). American Indians are large producers for commodity markets and any impact to productivity translates to immediate economic impact to operators. Thus, impacts to environmental quality may impact Native Americans more directly and severely than the general population. It is important to note that American Indian seed provides the foundation for modern agriculture and much of the seed stocks and diversity remain.

There are a number of tribal organizations that work on natural resource management across the country, the Intertribal Agricultural Council (IAC) being one. The IAC was founded in 1987 to pursue and promote the conservation, development and use of Tribal agricultural resources for the betterment of Native Americans. The Native Farm Bill Coalition (NFBC), which is a nationwide initiative to advance a common Native American agenda on the federal Farm Bill, is an example of linking policy and science. The Farm Bill is one of the largest pieces of legislation in the US, enacted by Congress every five years. The Bill addresses agricultural policies, food production and rural development. Over 170 Tribes and 14 intertribal organizations are represented in the NFBC, which resulted in a historic number of Tribal provisions in the 2018 Farm Bill. The NFBC is an opportunity for Tribal leaders to learn about environmental issues related to agriculture, which they can then advocate for in their communities.

#### **Opportunities for Collaboration**

The diversity of farming practices and the location of tribal lands in ecologically sensitive areas creates many opportunities for collaboration to better understand the patterns and impacts of atmospheric deposition. For example, wild rice is sensitive to climate variability and the chemical composition of the water. Better understanding the impacts of atmospheric deposition on wild rice is a research opportunity for partnership with Tribal Nations.

Existing partnerships between Tribal Nations and Land Management Agencies create opportunity for science and development of best management practices for agriculture. For example, the IAC works closely with USDA Natural Resources Conservation Service (NRCS) and with producers in the field to educate them on the scientific basis for resource management and how science can impact producers. There are also ongoing collaborations where tribes have built large capacity for monitoring and managing resources that could be further expanded. For example, the Great Lakes Indian Fish and Wildlife Commission (GLIFWC) manages the Ceded Territory where mercury monitoring is being conducted (Figure 2).

### **GLIWFC Mercury Concentration Data**

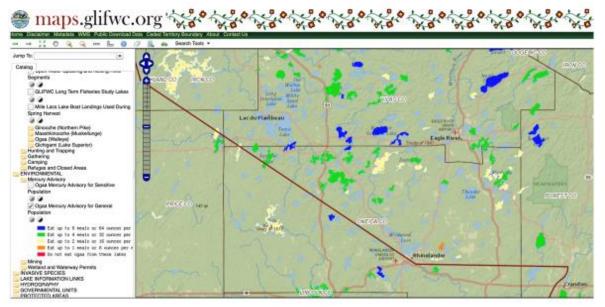


Figure 2. Example map of mercury advisories for sensitive populations. Colors indicate recommendations on fish consumption for lakes. Monitoring is conducted by the Great Lakes Indian Fish and Wildlife Commission (<u>http://maps.glifwc.org/</u>).

The GLIFWC mercury monitoring program is an example of how tribal nations partner with monitoring efforts that inform communities.

In addition to existing partnerships, tribal colleges and universities represent an opportunity for new monitoring and scientific collaborations.

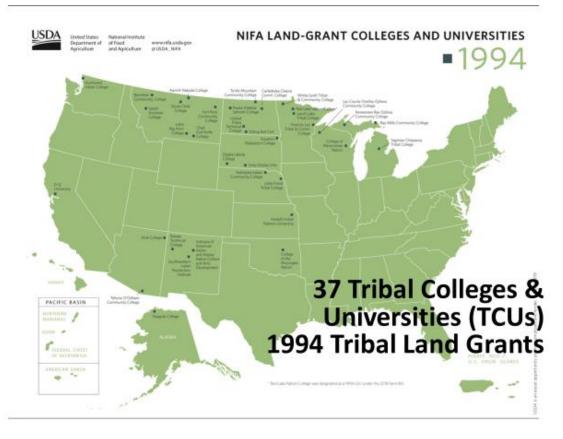


Figure 3. Map of tribal colleges and universities (TCUs).

As shown in Figure 3, the locations of tribal colleges and universities present opportunities to expand monitoring and research in areas that currently experience elevated rates of nitrogen deposition (Oklahoma, Kansas, Nebraska) and other areas in the Southwest and Northern Great Plains. Moving forward, the National Atmospheric Deposition Program and its stakeholders should pursue opportunities to engage more closely with Tribal groups to expand research and monitoring to better understand the linkages between agriculture and reactive nitrogen deposition and impacts to Tribal lands.

# Panel Discussion: Peter Vadas (USDA ARS), Jim Cheatham (NPS), Gail Tonnesen (EPA Region 8), Dan Cornelius (Intertribal Agricultural Council/University of Wisconsin-Madison)

The summary of the panel discussion is grouped into the following categories: audience questions for the presenters, panel discussion led both by moderator-posed questions and audience questions. Relevant questions and responses are grouped and summarized for clarity.

#### **Follow-up Questions for Presenters**

#### USDA ARS research on Best Management Practices (BMPs) for ammonia

Randy Martin (Utah State University) asked if the impacts to animal productivity (weight gain, etc) had been assessed in BMPs aimed at reducing crude protein in feed as a means to reduce ammonia emissions from animal manure. Peter Vadas responded that indeed that has been investigated. The idea is to see how much the feed protein can be reduced while maintaining animal productivity targets. For crops, USDA is investigating how lower crude protein in feed affects teh nutrient value of manure used for crop productions. They are investigating the full cycle. Viney Aneja (North Carolina State University) followed up by asking Dr. Vadas about the cost of BMPs, specifically, how much would the cost of the commercial product have to increase before the BMP was considered too expensive. Dr. Vadas pointed out that ARS does not generally work on the macroscale economics of BMPs but focuses more on the microscale. For example, in an assessment of the use of scrubbers to reduce NH<sub>3</sub> emissions from animal houses, ARS research would examine whether the value of the nitrogen recovered would exceed the cost of the acids used for scrubbing.

#### Measurements to inform aerosol processes

Gail Tonnesen mentioned in her presentation that some of the CAMx model underestimation for ammonium nitrate aerosol in Utah may be due to the fact that some of the "observed" nitrate may be non-volatile (e.g., calcium nitrate). Jesse Bash (EPA) commented that this was also the case in the San Joaquin Valley. Gail reiterated that measurements of non-volatile nitrate would be particularly useful if supersites could be established, referencing Jeff Collett's morning presentation in which the concept of hybrid networks including a limited number of supersites was discussed.

#### **Panel Discussion**

The panel was asked if current research is providing the information that Federal Agencies need to better understand the linkages between agriculture and reactive N deposition.

Perspectives on emissions and BMPs

Jim Cheatham noted that what we hear from agriculture is that you can't just use a single emission factor for cattle, given the diversity of producers, practices, etc and that at present it is difficult to develop estimates of facility scale emissions taking into account farm-specific management practices. The direction NPS is moving is to evaluate the available practices and BMPs and potential for individual producers to implement the recommended BMPs as a metric of performance within their focus area. This approach is currently used to assess the baseline for agricultural performance, with a goal of improving performance over time by implementing BMPs within the focus area. Additional research on the effectiveness of BMPs would be helpful in this regard.

Peter Vadas noted that one thing USDA/ARS scientists could use help on is developing the best methods for measuring emissions. Furthermore, ARS has a lot of stakeholders and our ability to meet their needs requires knowing what the needs are by attending these types of meetings. For our scientists, working on farms and engaging directly with growers is also very important, for example in understanding what practices need to be characterized and whether emission models are addressing realistic scenarios. Keeping lines of communication between producers and ARS scientists is key.

Donna Schwede posed a question to Dan Cornelius (IAC), noting that Dan Wildcat was the keynote speaker at the NADP meeting in Santa Fe a few years ago. He talked about how the unique perspective of the Tribal understanding of nature was different from ours. Are there particular agricultural management practices that tribal nations use that others should be using and that our models should be capturing? Dan noted that the Menominee reservation is an example where the land was managed differently from surrounding regions in that it was never clear-cut. Many researchers are investigating how this has affected things like soil quality. In terms of sharing knowledge, there are examples of BMPs being used and the knowledge being shared. However, sometimes there are hesitations in sharing that information with the broader outside community. It really depends on a case by case basis but building partnerships is key to working together.

#### Measurements to inform air quality and deposition modeling

Gail Tonnesen noted that air quality models do not perform well for vertical dispersion, particularly at night. Time integrated measurements (24 hour or longer) average over these nighttime conditions, making it difficult to use the measurements to assess model performance. Higher time-resolution (hourly) and vertical profile measurements are needed to evaluate and improve the models. Additionally, EPA Region 8 is concerned about visibility impacts in Class I National Parks and Wilderness Areas and some sensitive Class II areas. These areas often experience concentrations of NH<sub>3</sub> (0 to 0.5ppb), and sulfate and nitrate (0.1 to 0.5 ug/m<sup>3</sup>) that are very low compared to health-based standards but are significant for regional haze planning. This is a big challenge for models and emission inventories used to address visibility impairment issues in these areas. New measurements and research that allow for model evaluation at such low concentrations are needed.

#### **Opportunities for Cross-Agency and Stakeholder Engagement**

John Walker noted that there seemed to be considerable overlap in the presentations in terms of science needs that are common among agencies, representing an opportunity for agencies to engage more closely to advance the science more rapidly. The question was posed to the panel, what are the opportunities to communicate with other agencies and stakeholders during the planning stages of your programs?

Gail Tonnesen noted that the EPA Region 8 office works closely with the land management agencies and tribes through the Western Regional Air Partnership (WRAP). The WRAP is motivated by the Regional Haze Rule, the National Environmental Policy Act, and regional ozone planning so there is good integration and communication for planning in that regard.

Jim Cheatham noted as an example the Memorandum of Understanding between National Park Service, Colorado Department of Public Health and Environment, and U.S. EPA, which provides the basis for agencies and stakeholders to engage closely with respect to planning and implementation of the ongoing Rocky Mountain National Park Air Quality Initiative. As a final comment, Jim noted that he has heard throughout the day about the need to better understand facility-scale practices. It's critical that we stay on the cutting edge of effective practices and we need to keep the lines of communication open with producers and stakeholders to do so.

Peter Vadas described the USDA Listening Sessions which are an opportunity for stakeholders to describe their interests and research needs to USDA program managers. This process then informs the development of research action plans that ARS stations use to define their 5-year research projects. That's an example of a formal opportunity for stakeholder engagement. Furthermore, ARS scientists are constantly engaging with stakeholders at the project level, which is an important line of communication. As a final comment, Peter reiterated that these types of meetings are great for engaging with stakeholders. When you see needs that ARS can fill, please reach out.

Mike Olson (NADP) noted that NADP works closely with Tribal groups for monitoring, mostly in the air quality arena. Mike posed a question to Dan Cornelius: does the Intertribal Agricultural Council give feedback to the air monitoring groups and is there something NADP can do to foster that communication? Dan responded that his group (IAC) helps tribes achieve better access to USDA programs so they typically work more closely with USDA/NRCS than USDA/ARS. However, many tribes do participate in air quality monitoring. One opportunity for greater engagement with the air monitoring community is our IAC annual conference, which would be a place for these discussions, as well as the Native American Fish and Wildlife Society. Those are good opportunities to connect directly. At the more local scale, in Wisconsin we have Wisconsin Tribal Conservation Advisory Council. EPA used to come to those meetings more frequently but still does from time to time. These meetings would be an opportunity for air monitoring groups to engage with the Tribal agricultural community on a more local scale. As a

final comment, Dan noted that tribal lands are some of the most ecologically diverse and productive lands in the world and we appreciate the opportunity to have a place at the table when decisions are being made and for the science to be conveyed in a way that people are engaged and that they can understand.

### Session 5: Commodity groups and state agencies

Bill Hammerich (Colorado Livestock Association): Commodity group perspective - Colorado Livestock Association

#### Introduction

Bill Hammerich is the Executive Officer of the Colorado Livestock Association (CLA) and presented a summary of the organization and its perspective and activities supporting the Rocky Mountain National Park Initiative to reduce N deposition to the Park.

The CLA is an organization of farmers and agricultural producers that works to strengthen and unite animal agriculture in Colorado. Its headquarters are located in Greeley, CO and was formed in 1998 through a restructuring of the Colorado Cattle Feeders Association. The restructuring reflects the membership of the organization which includes producers of cattle and sheep, cow/calf, dairy farmers, swine operations, and agribusiness partners (CLA, 2019).

Much of the agricultural activity in northeastern CO is situated along the drainage of the South Platte River, which is a very productive area. As an example, Weld county is the 5<sup>th</sup> largest agricultural producing county in the U.S., though the county itself is very geographically large.

#### State of the Science

The CLA is an advocate for environmental protection, an issue which it takes seriously. That focus includes both air and water, but the focus for the workshop is on air. The CLA first became aware of the issue of N deposition to RMNP in 2005. Beginning in 2006, CLA and a diverse group of stakeholders including the Environmental Defense Fund came together to evaluate the issue. The focus eventually shifted to the agriculture industry and the RMNP Initiative agricultural subcommittee was subsequently formed.

At the beginning of the process, focus included the development of Best Management Practices (BMPs) and included technology companies with emission reductions ideas and products (e.g. compounds to mix with manure or spread on bedgrounds to bind NH<sub>3</sub> and prevent its emission and strategies for reduction of lagoon emissions). Over time, the cost and effectiveness of many BMP technologies usually dictate their applicability on operational farms (mentioned frequently in Peter Vadas' earlier presentation).

Some challenges encountered by the CLA, especially at the beginning of the process, included convincing its members that the issue with N deposition in RMNP was a real problem and one that the agricultural community could assist with. Free seminars were held to explain the science behind the issue, particularly the meteorology behind the 'up-slope events' (i.e. wind trajectories that carry agricultural emissions from the farms up into RMNP). The partnership with the NPS was helpful in this, as Jim Cheatham and other NPS officials frequently presented.

As discussions and deliberations between the partners progressed, trust was built. Bill felt that that trust was key to the success of the collaboration, and any similar efforts should focus on building trust.

In 2014, the RMNP Initiative partners were working with a doctoral candidate from Texas A&M University who envisioned the idea of the Early Warning System (EWS) to be used in northeast Colorado to provide a forecast of periods when upslope conditions are likely and BMPs at CAFOs on the Front Range would therefore be most effective. The EWS was patterned off a similar and successful system used in Kansas for prescribed pasture burning events in the Flint Hills. The EWS of northeast Colorado is housed at CLA with 63 participating members. For an average upslope event warning, there is a 38 to 45% response from producers to implement BMPs. The response is dependent on the time of year and other factors (e.g. severity of event). Prior to the next EWS cycle, there is a plan to re-evaluate the BMPs and the EWS, particularly for benefits that may not be obvious to some producers. CLA is hopeful that the BMPs review and outreach will help to improve response.

As part of the plan to assess BMPs, a survey of producers in Weld and Larimer county is conducted by CLA. The target for that survey is a 60% response and CLA is optimistic to reach that target in February 2020. The challenge in procuring the survey response goes back to the issue of trust. Agricultural people can sometimes be suspicious about government-related programs and want to know what the motivation is. CLA has observed that there is an age dependence in response which was hypothesized to result from generational attitudes on environment and trust of the government. The method of the survey distribution is important, as surveys distributed by U.S. mail (with return envelope included) received nearly triple the response compared to electronic distribution. The survey will be repeated in late November 2019, which may result in less of a response (due to the repeat) but needs to be done to preserve the confidentiality of the response which CLA regards as very important.

From the vantage point of CLA, the most important aspect of engaging the farm operators is effective communication and transparency. People need to know what we are doing, why we are doing it, and what the expected ultimate outcome is.

#### **Future Directions**

CLA will continue to partner in the RMNP initiative. There are plans in place to re-evaluate the BMPs and communicate their benefits to the producer. There are plans for additional producer surveys and continued participation in the EWS. CLA feels that a lot of the difficult groundwork has been completed and a sense of trust has been built between the participating agencies and the producers. CLA will continue to build this trust and improve communication to help meet the targets established in the RMNP Initiative and ultimately reduce the contribution of agricultural N deposition to RMNP.

#### References

Colorado Livestock Association. 2019. Colorado Livestock Association. <u>https://coloradolivestock.org/about/</u>. Accessed September, 2020.

#### Alan Blaylock (Nutrien): Fertilizer industry use of atmospheric deposition data

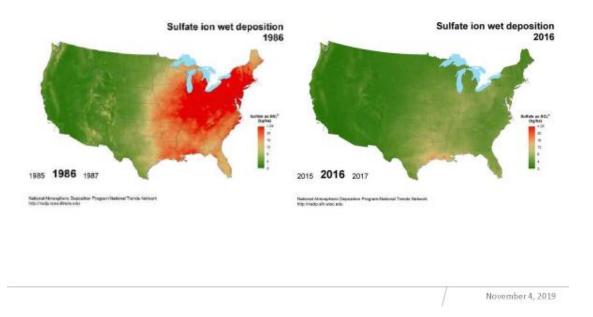
#### Introduction

Alan Blaylock, as a representative of Nutrien, provided an outlook on how the fertilizer industry utilizes atmospheric deposition data and develops and utilizes best management practices (BMPs) to reduce NH<sub>3</sub> volatilization from fertilizer application. Nutrien is the largest fertilizer manufacturer and supplier in the world, and has its US headquarters in Loveland, CO. Nutrien Ag Solutions, a subsidiary company, is the largest agricultural retailer in the world.

The fertilizer industry uses atmospheric deposition data in numerous ways. The first is determining the potential source of crop nutrients and several examples are discussed. A second way is using deposition data as an indicator of nutrients in the environment. This is done less frequently as it is often not clear where nutrients originate from and how they are dispersed. The industry considers the interpretation of the indicators of nutrients in the environment as too complex to be of much use. A third use of deposition data is for the early detection of emerging issues, and lastly, for understanding the impacts of management changes. Changes to management can be more difficult to apply given some of the communication and differences in application. For instance, atmospheric scientists quantify concentrations on the ppb level, whereas the soil science and agricultural industry use much larger scales. Scientists also apply models on a regional and/or national level and the agricultural industry manages issues on a much smaller scale (field/farm).

#### State of the Science

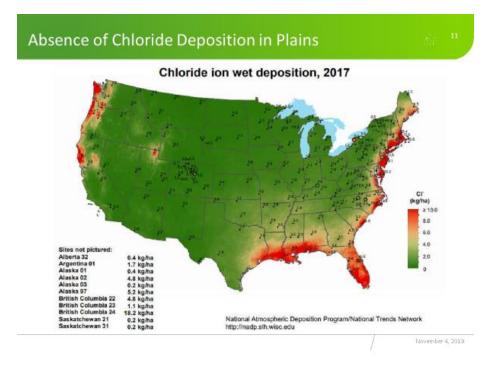
Two examples of how the fertilizer industry can use deposition data are described for sulfur and chloride. Greater crop yields have necessitated a greater demand for sulfur (S). However, the significant decrease in sulfur deposition across eastern North America has caused S deficiencies and crop responses in formerly unresponsive areas. Currently, there is an increased need for fertilizer S applications. The NADP sulfate ion wet deposition maps from 1986 and 2016 (Figure 1) illustrate the decrease in sulfate wet deposition.



### *Figure 1. Maps illustrating the change in annual wet sulfate ion deposition between 1986 and 2016.*

From 1986 to 2016 sulfate deposition in the eastern U.S. has decreased from levels above 20 kg ha<sup>-1</sup> down to single digits of kg ha<sup>-1</sup>. The deposition rate in the 1980's was more than sufficient for crop needs, but current deposition rates do not supply enough sulfur to meet crop demand and S deficiencies are now seen. Many of the soils in regions where sulfur has declined have significant sulfur supply from soil organic sulfur mineralization. Until the deposition declined sufficiently below the total crop sulfur budget as to justify fertilizer application, a change in practices was not needed in some areas. The increasing need for sulfur applications to crops has been followed for several decades based on the trends in the NADP deposition data, which have been a useful resource in this regard.

There is naturally low chloride deposition in the Plains states and Prairie provinces where most cereal crops are grown (Figure 2). Since these soils are high in potassium, little or no potash is applied. However, chloride is the second nutrient in potash and cereal crops have high chloride requirement and sensitivity.



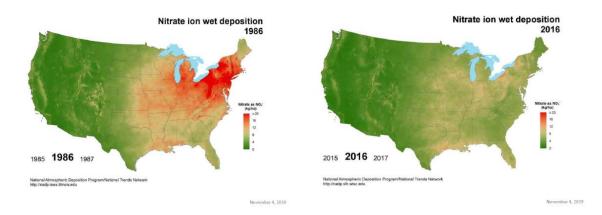
### Figure 2. Chloride wet deposition for 2017 illustrating the absence of chloride deposition in the Plains states where cereal crops are grown.

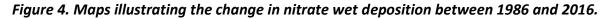
Soil test values show that the percent of soil samples with less than the critical level of 4 ppm water equivalent chloride concentrations are most prevalent in the Midwest. Low chloride levels have been identified as the cause of leaf spot syndrome in winter wheat (Figure 3). It was originally identified as a physiological spot, but it has been determined to be caused by chloride deficiency. This has necessitated chloride applications to crops as there is little or no chloride deposition in the Midwest, far from the oceans. Maps of chloride deposition help to inform such management strategies.



*Figure 3. Evidence of chloride deficiency in winter wheat.* Not all varieties of wheat show this damage.

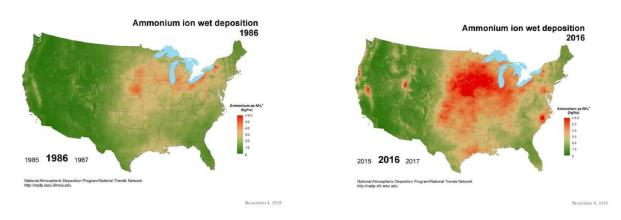
From the vantage point of agronomics, inorganic N deposition from the atmosphere is a very small portion of total crop N budget, especially when compared to the crop demand and other N sources to the system (e.g. fertilizer and mineralization of organic nitrogen). Nitrate wet deposition exhibits similar reductions from 1986 to 2016 as sulfur deposition (Figure 4).





While nitrate deposition has decreased since 1986, ammonium wet deposition has shown the opposite trend (Figure 5). Most of the ammonia emissions are attributed to agriculture. Much of the discussion in the morning focused on ammonia emissions from animal feeding operations, but not as much discussion about cropland. This trend in ammonium deposition could be explained by regional increases in NH<sub>3</sub> emissions and a shift in the gas-particle partitioning of NH<sub>4</sub><sup>+</sup> toward more NH<sub>3</sub> in the gas phase as concentrations of acidic nitrate and

sulfate decline (Warner et al., 2017). However, historical trends in total ammonia emissions developed from bottom-up national inventories contain large uncertainty, particularly for animal production and mobile sources (van Damme et al., 2017).

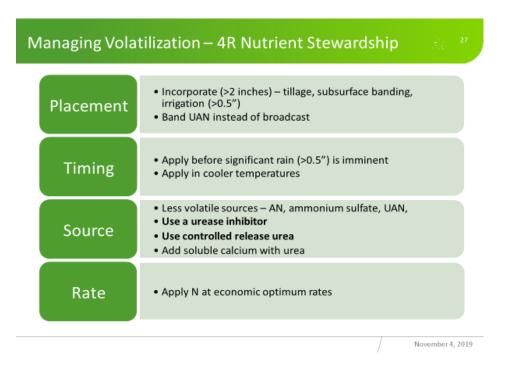




In addition to these reasons for the increasing ammonium deposition, Dr. Blaylock also noted that "global ammonia emissions have doubled over the past 70 years and are forecast to continue to rise, in large part because of growing demand for chemical fertilizers" (Plautz, 2018). He stated that, while nitrogen use has increased significantly in some parts of the world, the U.S. has seen relatively small increases in nitrogen fertilizer use, so total nitrogen use seems unlikely to account for the increase in atmospheric ammonia. In the U.S., nitrogen fertilizer use has increased from about 11.5 million tons in 1985 to about 13.0 million tons in 2015, with annual fluctuations caused by supply/demand and price changes (USDA, 2019a). Some of this increase can be attributed to increased corn acreage over this period. Average per-acre rates have remained largely unchanged over this period. A significant shift in fertilizer form has occurred over this period that could influence atmospheric ammonia as anhydrous ammonia and ammonium nitrate use has declined accompanied by a sharp rise in urea use (3.4 M tons in 1986 to over 7.0 M tons in 2015) (USDA, 2019b).

The fertilizer industry has developed and actively promotes Best Management Practices (BMPs). Nutrien's education program includes the '4 R Framework' which refers to the 'Right Source, Right Rate, Right Time, and Right Place' (Figure 6). For every fertilizer application decision, components of this framework are considered. These decisions are often very complex and a change in one component will impact the other components. Some of the BMPs for NH<sub>3</sub> include incorporating the fertilizer into the soil instead of surface application. Some changes have taken place with fertilizer use. In the Midwest, surface-applied urea is recommended to be applied when a significant rainfall forecast (Timing in Figure 7), which will help to get fertilizer into the ground, so it does not lay on the surface and volatilize. The

temperature of application has recently been called into question. A study in Montana showed high NH<sub>3</sub> volatilization rates of fertilizer applied to frozen soils in winter.



### Figure 6. The four cornerstones of minimizing ammonia volatilization due to fertilizer application

Another BMP is to use less volatile sources (e.g. ammonium sulfate) as urea or urea-containing solutions are the culprit in most NH<sub>3</sub> emissions. There is a need to enhance efficiency of fertilizers in order to decrease urea volatilization. Enhanced efficiency fertilizers use a urease inhibitor to slow the hydrolysis of urea. Lastly, fertilizer should not be overapplied.

There are other gaseous N emissions (e.g. N<sub>2</sub>O, N<sub>2</sub> gas, NO<sub>2</sub>) in addition to NH<sub>3</sub>. N<sub>2</sub>O is usually very small portion of the N budget, and not agronomically significant, however, N<sub>2</sub>O and N<sub>2</sub> gas emissions can be significant in denitrifying conditions.

#### **Future Directions**

In the red, high ammonium deposition areas (Figure 5), there has been a large conversion from plowed agriculture to minimum or no tillage practices which has resulted in an increased use of surface-applied fertilizer (likely urea, UAN). No tillage results in more surface nitrogen applications, thereby increasing opportunity for loss to the atmosphere. This is an unintended consequence of switching to reduced tillage for the sake of erosion prevention, soil health, and carbon sequestration. Also, much of the nitrogen fertilizers used to be applied before planting, but practices have switched to more split application, i.e. more in-season application, to reduce risk of loss and improve efficiency, which means application in warmer temperatures where

potential for volatilization maybe higher. A big question to consider is if we have exacerbated NH<sub>3</sub> volatilization by BMPs to address other areas of concern (nitrate leaching, soil health, carbon sequestration)?

There are also several important processes that are critical for ammonia emissions and may be critical questions for modelers to consider:

- 1. The majority of ammonia volatilization from fertilization occurs in the first 7-10 days after application. Do models account for these peak events?
- 2. Precipitation events that follow soon after fertilizer application play a large role in ammonia volatilization. Do models account for precipitation events?
- 3. Soil ammonium is generally a very small portion of soil nitrogen except for a short period of 1-2 weeks after fertilizer application. If you are not measuring during this period, it will not be observed, is this situation recognized in current models?
- 4. Ammonia may volatilize from crop leaves/biomass. Do models take into consideration this volatilization? How are emissions and depositions affected as plant productivity and biomass increases?

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Plautz, J. 2018. Piercing the haze. Science, 361, 1060-1063.

USDA. 2019a. <u>https://www.ers.usda.gov/data-products/fertilizer-use-and-price/summary-of-findings/.</u>

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Warner, J. X., Dickerson, R. R., Wei, Z., Strow, L. L., Wang, Y., Liang, Q. 2017. Increased atmospheric ammonia over the world's major agricultural areas detected from space, Geophys. Res. Lett., 44, 2875–2884, doi:<u>10.1002/2016GL072305</u>.

#### Additional Resources

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#### Greg Zwicke (USDA NRCS): USDA NRCS and reactive nitrogen

#### Introduction

The Natural Resources Conservation Service (NRCS) serves as the conservation agency within the United States Department of Agriculture (USDA). The NRCS is not a regulatory agency in that it cannot tell farmers what to do, nor is it a research agency. The agency relies on ARS, NIFA, FS, and other federal agencies and universities to conduct research. The NRCS works with agricultural producers to accomplish natural resource objectives through voluntary conservation efforts. To this end, a lot of effort is put into building relationships. The agency provides technical and financial assistance to help implement Best Management Practices (BMPs) and uses their conservation planning process to address resource concerns holistically. The NRCS works out how to take the science into their system and then out to the farmers. The disparity they experience with deposition research is that deposition data are collected on a broad scale and the NRCS works on the farm scale. The main problem to solve is how to take scientific information that reflects large-scale processes (i.e., landscape or regional) and apply it at the farm scale. The purpose of this presentation is to inform scientists of what NRCS does so scientists can then provide feedback on how the science can help with agricultural air quality issues.

#### State of the Science

The NRCS planning process is composed of nine steps that identify resource issues on farms associated with soil, water, air, plants and animals, and energy. There are 47 separate resource concerns within NRCS right now and five are related to air quality. The agency applies a suite of conservation practices to holistically address these resource issues without causing other issues on farms. Air emissions from agriculture include particulate matter (PM), ammonia, volatile organic compounds (VOCs), oxides of nitrogen (NOx), odorous sulfur compounds, methane, nitrous oxide, and carbon dioxide (CO<sub>2</sub>).

The five air quality resource concerns are:

- 1. Emissions of PM and PM precursors; involves ammonia
- 2. Emissions of ozone precursors; involves NOx
- 3. Objectionable odors; involves ammonia
- 4. Emissions of greenhouse gases; involves nitrous oxide, and
- 5. Emissions of airborne reactive nitrogen (Nr); involves ammonia and NOx

Emission of Nr is a relatively new concern that came about as a result of NRCS' participation in the Rocky Mountain National Park Air Quality Initiative. The agency did not have a mechanism to help with nitrogen deposition issues on farms which is why the 5<sup>th</sup> air quality resource concern was added. As can be seen from the list above, nitrogen is involved in all 5 air quality resource concerns.

The NRCS has nearly 170 existing, official conservation practices, about 50 of which have a specific air quality related purpose. Conservation practices are not specifically control technologies but can include the application of control technologies. Many of the air quality related conservation practices are related directly to PM and dust emissions, but several specifically target nitrogen. These are:

- 1. Air filtration and scrubbing (add-on technologies such as scrubbers to swine barns, etc.)
- 2. Amendments for treatment of agricultural waste (alum application)
- 3. Combustion system improvement (reducing emissions of NOx from engines like tractors)
- 4. Feed management (reducing nitrogen inputs into system)
- 5. Field operations emissions reduction (arose out of California agricultural practices and deals with dust and PM emissions and addresses NOx this year)
- 6. Nutrient management (reducing nitrogen inputs from animal and crop nutrients)

Congress has directed NRCS to administer various federal conservation programs. These programs are the vehicle that allows NRCS to allocate financial assistance to participating producers. The main programs among these are the Environmental Quality Incentives Program (EQIP), the Conservation Stewardship Program (CSP), and the Regional Conservation Partnership Program (RCPP). EQIP is the biggest of these three programs with an annual budget of approximately 1 to 1.5 billion dollars. EQIP promotes agricultural production and environmental quality through financial and technical assistance. Under EQIP is the National Air Quality Initiative (NAQI) which is designed to address air quality issues by helping producers meet air quality compliance requirements and by providing opportunities to install air quality conservation grants (CIGs).

The NAQI program was introduced in the 2008 Farm Bill at a budget of \$35 million per year to specifically address air quality issues and was originally targeted for PM non-attainment areas only. However, it became evident that non-attainment areas, other than California and a small portion of Arizona around Phoenix, did not have a big agricultural contribution to the non-attainment. The program was restructured in 2015 to start working with NRCS state offices to apply for funds to address various agricultural air issues and it is now open to all areas and states. NAQI was reauthorized in the 2014 and 2018 Farm Bills with \$37.5 million per year from 2019 through 2023. California is the biggest user of these funds having spent over \$120 million since 2008. Most of this money goes towards reducing emissions from diesel irrigation engines or tractor engines or to replace them with Tier 3 or Tier 4 engines (engines designed to operate with less emissions).

The Conservation Innovation Grants (CIG) program, also under EQIP, is a matching grant program to stimulate the development, demonstration, and adoption of innovative conservation technologies and approaches. There is no research conducted, rather the program is a mechanism to help get research over the "hump" from research to implementation by

performing on-farm demonstrations. By collecting data through application of new technologies on farms, the technology can be made more economically feasible. Through implementation of new technology, producers can also see if it is a viable option for them. At least 50% of the total project cost must be provided by the grantee with national and state-level grants being available. Several air quality projects have been quite successful in the past through such grants, however, there has not been much focus on air quality in the past few years. Instead, the focus has been on soil health, water quality, and nitrogen related water quality issues.

#### **Future Directions**

The main issue for the future is how can nitrogen deposition monitoring data be applied to inform questions of emission and local scale processes around farms. There are three main areas where NRCS needs input from the scientific community in order to apply nitrogen deposition data. These are:

- 1. How does a farmer know that there is a resource concern on the farm?
- 2. How should nitrogen emissions/flows at the farm level be addressed? Where are the areas and/or practices that can be tweaked in order to reduce emissions, and/or shifting such impacts so they are easier to control or eliminate? and
- 3. What practices/actions/techniques can be applied to reduce nitrogen emissions?

# Panel Discussion: Bill Hammerich (Colorado Livestock Association), Alan Blaylock (Nutrien), Greg Zwicke (USDA NRCS), Lisa Devore (CDPHE)

The summary of the panel discussion is grouped into the following categories: audience questions for the presenters, panel discussion led both by moderator-posed questions and audience questions. Relevant questions and responses are grouped and summarized for clarity.

#### **Follow-up Questions for Presenters**

#### **Best Management Practices**

Tim Sullivan (E&S Environmental Chemistry, Inc) noted that best Management Practices (BMP) for air quality is to fertilize in advance of rain. For water quality, it would be the opposite suggestion. How do you deal with these kinds of dichotomies?

Alan Blaylock responded that it is never as simple for the land owner/grower as we want to make it. In applying BMPs you must weigh the probabilities of success and reduce risk. The decisions can be complex, so we recommend identifying the greatest risk and attempt to mitigate for that. For many farmers the biggest risk of N loss maybe volatilization, in other areas (e.g. Indiana) leaching and denitrification may also be risks. Weather is a large factor in risk, it is a gamble because it is hard to predict the weather. Implementation of many BMPs ends up being all about risk management. Because of this, farmers are often encouraged to apply at multiple times to minimize the risk of fertilizer loss to the atmosphere.

Greg Zwicke noted that NRCS runs into this issue all the time. They try to take a holistic view of conservation planning. When looking at multiple resources (i.e., air, water, soil) at the same time, there will be conflicts. NRCS mainly focuses on water and there are conflicts between air and water. One set of BMPs is going to improve water quality but may harm air quality and vice versa. We first need to figure out how to determine if there are resource concerns and prioritize the concerns. If water quality is the biggest concern, then we may be OK with impacting air quality negatively and vice versa. The main issue is identifying the greater concern in order to deal with it.

Ryan McCammon (Wyoming BLM) posed a question regarding the RMNP deposition Early Warning System. Have you ever had any producers ask what changes were seen as a result of my actions? For instance, after a warning was issued have you ever been asked: I did all these things, what did it do to the Park as a result of what I did? What effect did the BMP have downwind from the EWS area in the Park?

Lisa Devore (CDPHE) commented that Aaron Pena, a graduate student at CSU, analyzed the early warning system to see if it was effective in reducing wet deposition in RMNP (Pina et al., 2019). Aaron found 13% reductions in wet deposition of NH<sub>4</sub><sup>+</sup> at the Beaver Meadows NADP/NTN site and lower reductions at the Loch Vale (approximately 6%) site. His analysis was

only for an 8-week period but reductions in deposition in response to BMP implementation were evident. We would like to look at a longer-term evaluation but need another graduate student for this. We are going on 5 years now for EWS.

Alan Blaylock noted that often these environmental impacts have been accumulating over decades, but practices are implemented based on regulations that specify relatively short clean up periods of 1 to 2 years. For example, new nitrogen management rules for Minnesota for water quality specify 3 years to clean up ground water and then regulations kick in. Sometimes early testing may indicate that BMPs were implemented but did not solve problems. The response time is slower than what is desired because natural systems sometimes do not respond quickly. We just don't know the effect of BMPs sometimes for many years. It is really tough to look at response times in the short term.

#### Sponsorship of NADP sites by agricultural stakeholders

Ryan McCammon posed the question, what about having/funding NADP sites as part of research on a farm?

Greg Zwicke responded that NRCS has not looked at funding NADP sites. Main reason being that all the funds under the NRCS programs are required to go directly to producer. There is some play in CIG's but mainly to demonstrate technologies that could be deployed on the farm.

Bill Hammerich noted that producers would be willing to have an NADP site. I know of 2 producers that have worked with CSU and Princeton.

Mike Olson (WSLH) followed up by asking, would producers be interested in having an NADP site? Maybe a change in N in soil systems related to deposition cannot be detected but we could actually see these reductions in air concentrations at an AMoN site after BMPs are implemented for some period of time.

Alan Blaylock noted that the fertilizer industry is confident a grower can reduce emissions from specific fields in hours or days with BMPs. If you are just monitoring near that field then air impacts can be seen right away. There has been lots of research on N management practices. There is good confidence in some BMPs that have been studied for decades.

Chris Clark (EPA) asked Greg Zwicke, the Soil Health Partnership consists of known networks of hundreds of farmers that coordinate activities. If NADP were to partner with a network like this and farmers were interested in hosting instrumentation, then would an NADP site be eligible for some funding?

Greg responded CIG or RCPP programs would be most applicable. Discussions have started on this. The air team has to interface with soil health people. Region 8 is putting together a soil health workshop to help bring their folks up to speed on soil health and how it could affect other media. NRCS is involved on both the water and air side, which has led to a discussion

within NRCS on how to promote co-benefits of BMPs for soil and water quality. NRCS is hoping to push some of that information out to the public soon.

#### Farmers as a source of data

Chris asked Alan, apparently on-farm soil pH data are not as abundant as we would like. Do you know of any existing network where farm soil pH data could be accessed? Alan responded that he did not know of a network that would have this data. However, farmers that test their soils would have this data, at field and even sub-field levels. Farmers have confidentially issues and are reluctant to give out this data. Analytical labs have the data but are not locally identified because of confidentiality issues. Would likely need to go to individual farmers for this data.

#### **Moderated Discussion**

### Building trust with the agricultural community: Lessons learned from the Rocky Mountain National Park Air Quality Initiative

John Walker noted that the RMNP Air Quality Initiative and Early Warning System is really a success story in terms of federal agencies and industry establishing partnerships and working together to address nitrogen deposition in the Park and to better understand the role of agriculture. What were some keys to developing this partnership that you think are broadly applicable that can be applied to other areas of the US?

Bill Hammerich responded that getting the agricultural community to understand the issue as well as being transparent and honest were key. It has taken awhile but farmers today are involved in the RNMP issue. They have come to grips with the fact that scientific research is not always going to give them the answer they want, but if it is credible, well done, and peer reviewed, it is what it is and they will learn to live with it.

Lisa Devore noted that, from the state perspective, we have been working on this issue for more than 12 years. At first, government and the agricultural community were coming from very different perspectives. Both had leaders and staff that worked very diligently and transparently together and would have multiple meetings to educate agricultural stakeholders to understand subject matters such as ecosystems in RMNP and relevant data. Agriculture stakeholders and producers have also been very good in turn at explaining their procedures and providing outreach opportunities to better understand the agricultural perspective. As a combined group, they have taken numerous field trips to both types of operations. They have been to the Loch Vale NADP site and also to feed lots, dairy farms and taken grain and crop tours, etc. These types of mutual outreach interactions were important in building relationships and could be effective in other parts of the country.

Jim Cheatham remarked that it took a decade to develop trust and rapport as Bill described and to consolidate information on what agricultural producers should be doing versus collecting the information on what they actually are doing. The latter could be done sooner to get on the

right track and would be a lesson learned for developing relationships with the agricultural industry in other parts of the country.

Alan Blaylock remarked that the Certified Crop Advisors (CCA) is an existing networking partnership administered through the American Society of Agronomy that could be engaged by the scientific community to build partnerships. Find those growers that are interested in working with you and open to doing research. Certain producers like to collaborate with researchers and they often work with retailer or crop advisor of the same mind set. During the EPA workshop on enhanced efficiency fertilizers this issue came up over and over from all sectors. There is a massive network of technical experts and semi-experts in the field that know the growers and which are bell cow growers (i.e. growers interested in research and cooperative projects). There is a trust in place. The Nature Conservancy (TNC) and cooperative extension services are in somewhat similar positions. TNC has done a lot of work in setting up these partnerships. A good place to start is by going to retailers and asking to collaborate with any bell cow growers who might be interested.

#### **Concluding Remarks from the Panel**

Alan Blaylock commented that after feeling out of place and like I did not belong in the morning, I learned a lot and really appreciated the invite. Fertilizer industry has huge stake in Nr in the environment and needs to be in this conversation. Fertilizer industry is a culprit in part of this and they are very interested in solving some of these problems. Our constituency, our customers and their buyers are telling us we must do some of these things. Food companies are telling farmers what to do, how to grow their food and what practices they need to use. I appreciate the invite and opportunity to interact with this group.

Lisa Devore noted that one thing that would be helpful for the future is NADP and all the products it offers from tools, graphs, etc. Also involves the national perspective and how it influences all other components of policy, planning, regulation. Data make a difference. People are looking at the data and this project has been highlighted a couple of times. It would be good for stakeholders to hear about other projects and what else is up and coming in NADP from the national perspective.

Bill Hammerich thanked the group for the invite. I also felt like I did not fit in with this group initially, but the takeaway is that I feel much better and more optimistic that we are working our way toward some solutions.

Greg Zwicke remarked that he appreciated all the work done to help characterize N emissions and the spatial allocation of these N emissions. Please stay in touch to try to help us (NRCS) figure out what to do with all this information.

#### References

Pina, A.J., Schumacher, R.S, Denning, A.S., Faulkner, W.B., Baron, J.S., Ham, J., Ojima, D. S., Collett, J. L. 2019. Reducing Wet Ammonium Deposition in Rocky Mountain National Park: The

Development and Evaluation of A Pilot Early Warning System for Agricultural Operations in Eastern Colorado. Environmental Management, 64. 626-639. https://doi.org/10.1007/s00267-019-01209-z

### Wrap-up and next steps

Below is a summary of the workshop discussion (led by John Walker) on potential workshop products.

A couple of the products from this workshop will be geared toward TDep to help us better understand how we can continue to engage with the agricultural community on N deposition issues. Besides a summary report which will include presentations and panel discussions, we would like to develop a Stakeholder engagement plan for TDep, specifically for the agricultural community. We will reach out to some of you to help us put together an outline as a road map for TDEP to begin doing more concrete things to stay engaged with the agricultural community.

Today we would like to get your input on the idea of a communication piece geared toward the agricultural community/producers. This would be framed by the needs of the stakeholder community and supported by some of the science discussed today.

What are some thoughts on developing a communication piece out of this workshop? Here are some of the science needs and here are some of the stakeholder needs. How do we bring these together? What outlet would there be for such a communication?

Jesse Bash suggested that we make it open access so that it can be shared with everyone with no fees or barriers.

What are some outlets for reaching the agricultural community?

Chris Clark echoed that trust is the main issue in reaching the agricultural community. There are existing networks that have this trust and those networks would be the route for reaching the agricultural community. There are folks here that have connections to these networks.

What is the vehicle these networks use?

Chris Clark continued that a journal article is not the best vehicle for reaching agricultural community.

Rich Grant noted that an article without a linkage to a trusting source is worthless. All states have various farming magazines, such as Prairie Farmer in Indiana. Extension services are also a very good outlet. A certain number of people from the extension service side have skepticism about the importance of Nr emissions/deposition. A workshop geared toward extensionists needs to be held, perhaps in agronomy society meetings where one can reach certified crop

#### consultants.

Randy Martin noted that scientists and engineers often have trouble communicating interests to general public. Might be good to bring other professionals (e.g., communications experts) on board that are better at doing this than we are.

Dan Cornelius noted that extension with NRCS has even more of an impact in working with producers. We have asked a similar question with hypoxia and runoff. What are the recommendations as to when to apply fertilizer? Looking at the bigger picture, there are relationships between these two issues and making connections with the right groups to address the issue is important. We are working with FWS and Landscape Conservation Cooperatives. Bringing these together with 15-20 different professionals from state and federal agencies is an example of an opportunity.

John Walker noted that at the core of this effort is NADP wanting to engage more closely with agricultural community. What data products can NADP provide? What types of monitoring data are useful to stakeholders? This would be a key part of moving forward.

Donna Schwede (EPA) asked if there is a role for the NADP Education and Outreach Subcommittee (EOS) committee in developing a communication piece on this workshop. The audience agreed that there would be. EOS has developed updated brochures in the past and this is a project they should consider doing something similar.

Carrie Furiness (NC State) agreed with Rich Grant that we should consider what our target audience (agricultural stakeholders) reads. This would include trade publications, commodity publications, newsletters.

An audience member asked about using social media as an outreach platform to connect to the agricultural community. Mike Olson noted that NADP is still getting a handle on how to effectively use the platform itself and illustrated the process that the subcommittees will funnel communication ideas through EOS, which will then pass messages for the NADP Executive Committee to post.

John Walker asked how do the commodity groups use social media? Bill Hammerich noted that he does not but his staff do and communicate topics regularly.

Selma Isil (Wood, Inc.) mentioned reaching out to NADP site operators could help to reach more of an audience. Site operators in the past have put on local seminars for different groups (e.g. Audubon, and 4H groups). There are opportunities at the local level that should be explored.

Alan Blaylock noted that there are outlets already in place that do a lot of this outreach like Fertilizer Institute, Agriculture Retailers Assoc., Certified Crop Advisors and they use social media. Keep us in the conversation. As long as you can bring the science back to an impact on the end user on the farm, the producer, cattlemen, etc. in terms of something that affects them, their system, etc. and something they can do something about then they will be interested. In your writing, relate science back to something in the real world and you will have a much better chance of developing an audience with the producers and people whose lives are impacted.

**Disclaimer:** Any opinions, findings, and conclusions or recommendations expressed in this publication are those of the workshop participants and do not necessarily reflect the views of the U.S. EPA, NADP program sponsors, or the University of Wisconsin-Madison.