Comparison of Aircraft Observed, Remotely Sensed, and Modeled Ammonia Concentrations in the American Midwest

Alexander Frie[1], Timothy Griffis[2], Matthew Erickson[2], Mackenzie Smith[3], Dylan Millet[2], Kelley Wells[2], Jonathan Bent[4], Xueying Yu[2], Stephen Conley[3], Randall Kolka[5], Mark Shepard[6], Karen Cady-Pereira[7]
[1]Minnesota Sea Grant College Program, [2]University of Minnesota Department of Soil, Water, and Climate, [3]Scientific Aviation, [4]Picarro, Inc., [5]United States Forest Service, Northern Research Station, [6]Environment and Climate Change Canada, [7]Atmospheric and Environmental Research

Ammonia in the Midwest

- Ammonia (NH_3) is the most abundant atmospheric gaseous base, and its emission is unregulated in the United States.
- The largest NH_3 sources are agricultural (e.g., fertilizer, animal waste).
- Emission reductions of acidic atmospheric species, land use changes, and increasing temperatures contribute to higher ambient NH_3 concentrations.

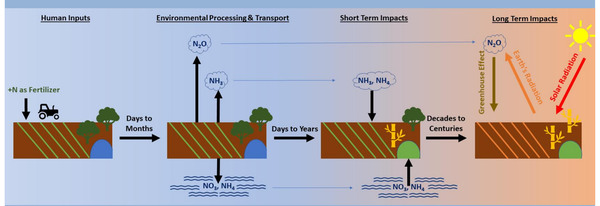

Figure 1: Graphic describing the many impacts of N pollution over various timescales.

Sparse Measurements, High Uncertainty

- Existing NH_3 emission inventories are highly uncertain.
- Observations of atmospheric ammonia are sparse, especially those with high spatial or temporal resolution.
- Model v. tall tower measurement comparisons have previously found models underestimated NH_3 in May in the Midwest.

Measurement, Modeling, and Remote Sensing

- Remote sensing provides high spatial coverage with high uncertainty.
- Aircraft measurements provide high temporal coverage and vertical resolution.
- Models provides high spatial and temporal coverage but rely on *apriori* emission, chemistry, and transport assumptions.

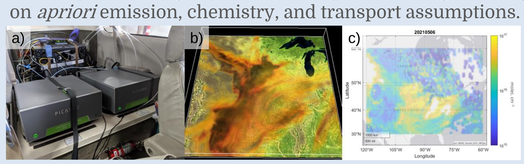

Figure 2. a) Picarro instruments mounted in the aircraft, b) example frame of NH_3 modeling results, c) example distribution of total column NH_3 retrieved by CRiS CRPR.

Measurement: Picarro G2103

Technique: Cavity ring down spectroscopy.
Uncertainty: ±0.19 ppb
Peak/trough time (10%-90%): ~85s
Target Region: IA, NE, SD, MN

Model: WRF-Chem

Domains: 3

Emissions: 2017 National Emissions Inventory (NEI), NCAR FiNN v1, Model of Emissions of Gases and Aerosols from Nature (MEGAN)

Mechanisms: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) and Model for Ozone and Related chemical Tracers (MOZART)

Target Regions: Iowa, Nebraska, South Dakota, Minnesota

Retrieved: CRiS CFPR

Data Source: Cross-Track Infrared Sounder Fast Physical NH_3 Retrieval (CFPR) v 1.6
Overpass Time: ~1 p.m. local time
Horizontal Resolution: 15 km

Target Regions

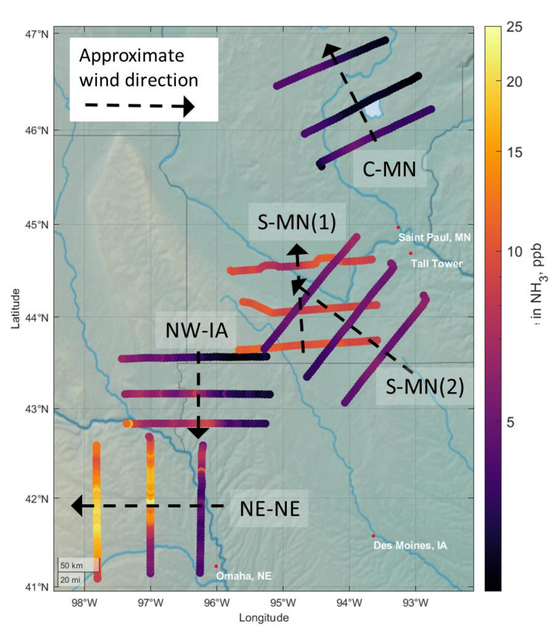

Figure 3: Parallel transects from flights, colored by aircraft observed NH_3 concentration. NE-NE(1) is not shown. Observations are within the boundary layer.

ID	Region	Flight Date
NE-NE(1)	Northeast Nebraska, Day 1	May 9, 2021
NW-IA	Northwest Iowa	May 10, 2021
NE-NE(2)	Northeast Nebraska, Day 2	May 11, 2021
S-MN(1):	Southern Minnesota, Day 1	May 16, 2021
S-MN(2):	Southern Minnesota, Day 2	May 17, 2021
C-MN	Central Minnesota	May 18, 2021

Table 1: Flight day IDs, target regions, and date of flight.

Land Use and Major Sources

NE-NE: Land use dominated by annual crop agriculture. High densities of livestock operations. Contains the county with the highest cattle production in NE.

NW-IA: Land use dominated by annual crop agriculture. High densities of livestock operations. Contains the county with the highest cattle production in IA.

S-MN: Land use dominated by annual crop agriculture. Some livestock operations. Significantly smaller cattle population than NE-NE or NW-IA.

C-MN: Transition zone between annual crop and forested/lake region. Some livestock operations. Significantly smaller cattle population than NE-NE or NW-IA. This region contains multiple lakes, including Lake Mille Lacs, MN (536 km²).

Satellite: Aircraft: Model Comparison

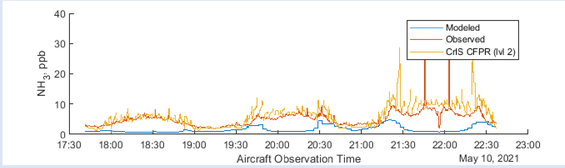

Figure 4. Retrieved, modeled, and measured NH_3 concentrations for NE-NE(2). A day that displayed strong agreement between the retrievals and measurements, but model underestimates. This is a point source dominated region.

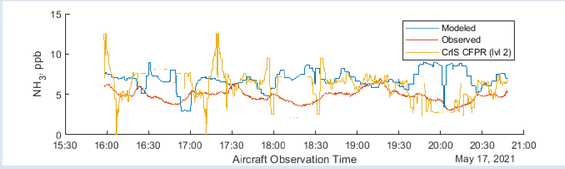

Figure 5. Retrieved, modeled, and measured NH_3 concentrations for S-MN(2). A day that displayed strong agreement between the three methods, on average. This is a non-point source dominated region.

Regional Averages

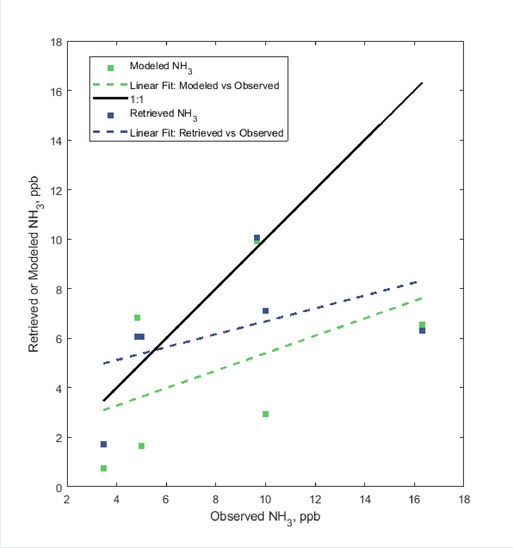

Figure 6. Average nearest neighbor (to flight tracks) CFPR (blue) and WRF-CHEM NH_3 (green) concentrations plotted against average flight concentrations.

	Aircraft ± SD	Retrieved ± SD	Modeled ± SD
NE-NE (1)	16.3 ± 6.5	6.3 ± 7	6.5 ± 2.8
NW-IA	5 ± 2.2	6.1 ± 3.5	1.7 ± 1.1
NE-NE (2)	10 ± 5.4	7.1 ± 1.6	2.9 ± 1.2
S-MN (1)	9.6 ± 1.3	10 ± 5	9.9 ± 2.7
S-MN (2)	4.8 ± 0.8	6 ± 1.8	6.8 ± 1.2
C-MN	3.5 ± 0.7	1.7 ± 0.8	0.7 ± 0.3

Table 2. Average flight concentrations and nearest neighbor (to flight tracks) CFPR and WRF-CHEM NH_3. Standard deviations represent variability in results, not uncertainty.

Mass Balance

Mass balance analyses were performed for regions between transects.

- NE-NE, NW-IA, C-MN regions yielded higher fluxes than those predicted in the model.
- These regions all contain cattle sources, suggesting underestimated cattle emissions in inventories.
- Downscaled mass balance reveals sources and sinks within target regions (Figure 8)

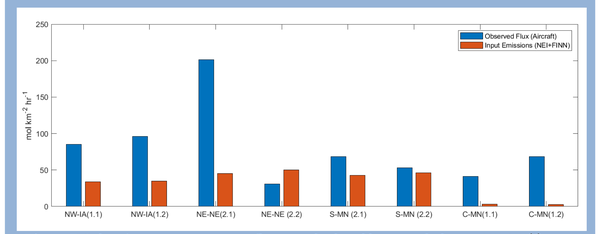

Figure 7. Select inter-transect mass flux as calculated by mass balance. NE-NE(1) and S-MN(1) are not shown due to inappropriate mass balance conditions.

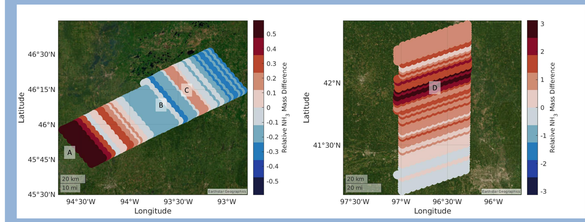

Figure 8. Relative NH_3 mass flux between parallel transects for C-MN(1.1) (left) and NE-NE (2.1) (right) labeled with A) an agricultural source region, B) Lake Mille Lacs deposition region, C) cattle source north of Lake Mille Lacs, and D) cattle source near Wisner, NE.

Implications

- CFPR retrievals capture the spatial disruption of boundary layer ammonia near strong point sources in NE.
- Modelled NH_3 underestimated observed NE-NE, NW-IA, and C-MN concentrations.
- NE-NE and NW-IA displayed both strong source character and model underestimates, suggesting they contain sources contributing to strong spring pulses.
- These regions contain high concentrations of cattle, suggesting underestimates of cattle emissions contribute to the spring underestimation in models.
- Underestimates of NH_3 in C-MN region suggest possible transport underestimates as well.

Acknowledgements

This work was supported in part by NSF, USDA, and Minnesota Sea Grant. We would like to thank Picarro Inc for supplying the G2103 instrument for the flights. We would also like to thank pilot Paolo Wilczak and the scientific aviation team. We would also like to thank the Minnesota Super Computing Institute for providing the super computing infrastructure for the modeling portion of this project.