Reduced Nitrogen in the Western US in Winter

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Sensitivity of $PM_{2.5}$ to NH_3

For dry particles, the presence of excess NH_3 allows semi-volatile salts to form, e.g. NH_4NO_3

For deliquesced particles (and liquid water clouds), NH_3 controls the pH of $PM_{2.5}$

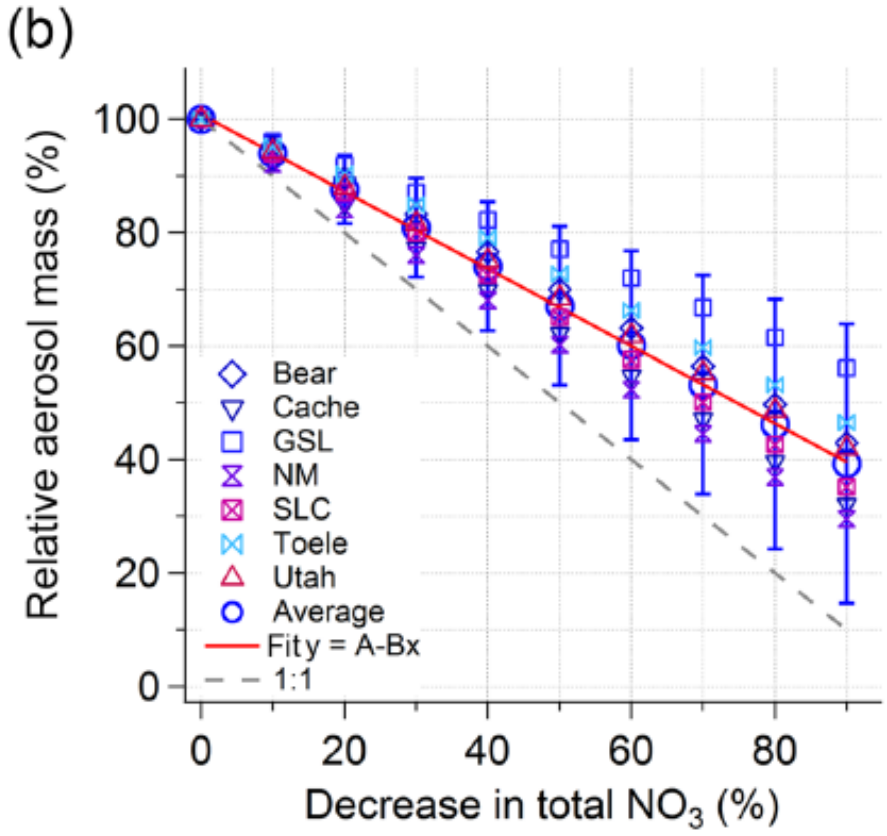
- partitioning of semi-volatile gases, e.g. HNO_3, HCl, oxalic acid

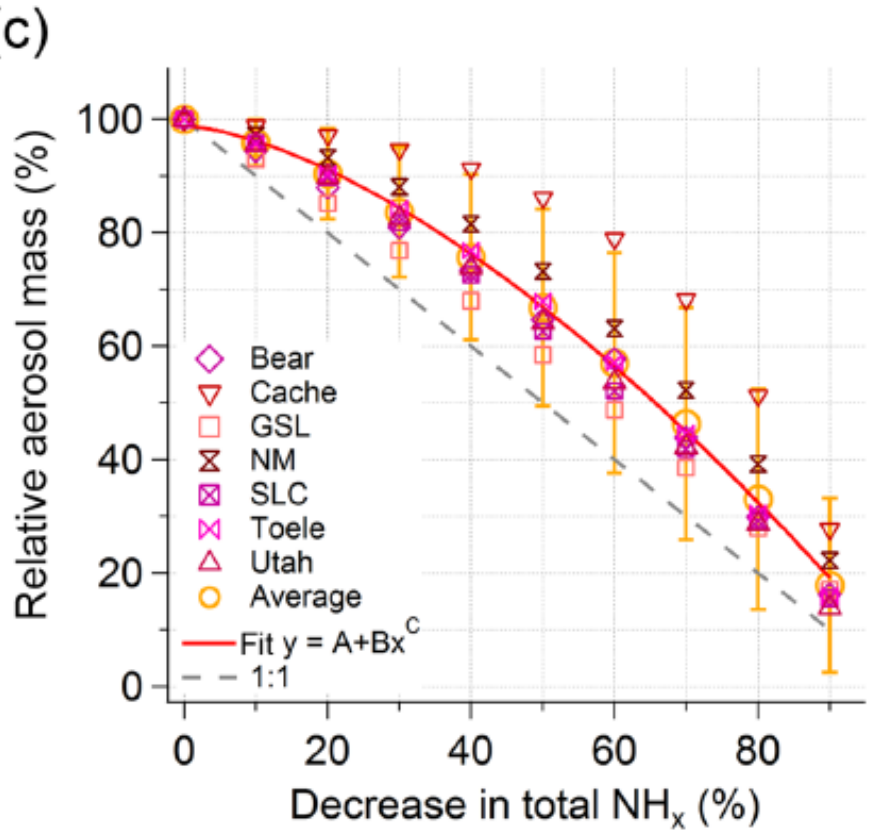
- rates of some aqueous phase reactions

NH_3 can also react with particle phase carbonyls to generate brown (light-absorbing) constituents

Deliquesced NH_4NO_3 is limited by NH_3 **and** NO_x

aircraft AMS, CIMS and QCL data from UWFPS 2017 in Utah





Franchin et al., *Atmos Chem Phys*, 2018

Sources of NH_3

Livestock

Fertilizer application

Residential wood combustion

Biomass burning

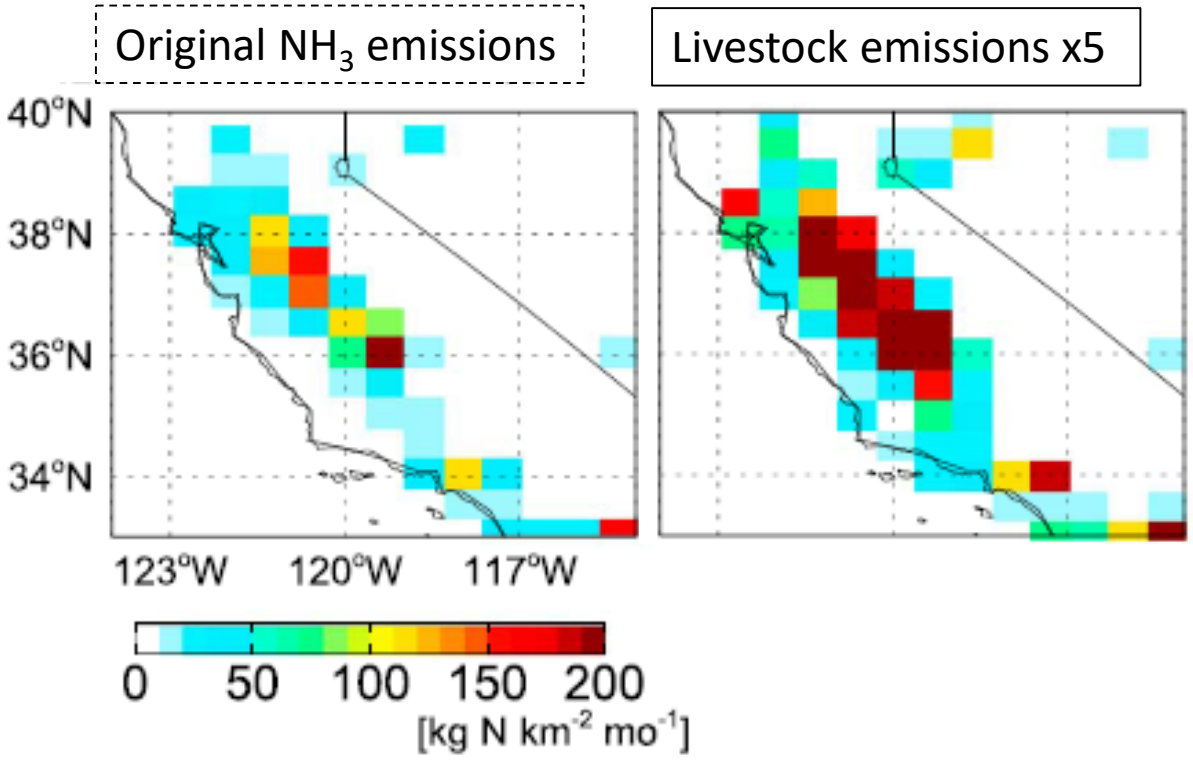
Mobile sources

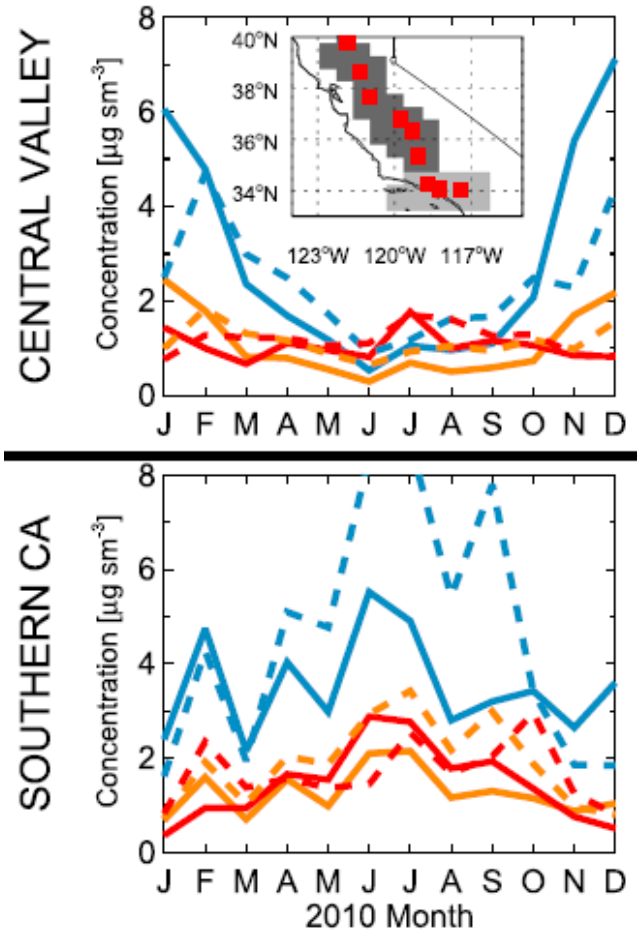
Industry

Bidirectional exchange

CalNex implied livestock NH_3 sources were significantly underestimated

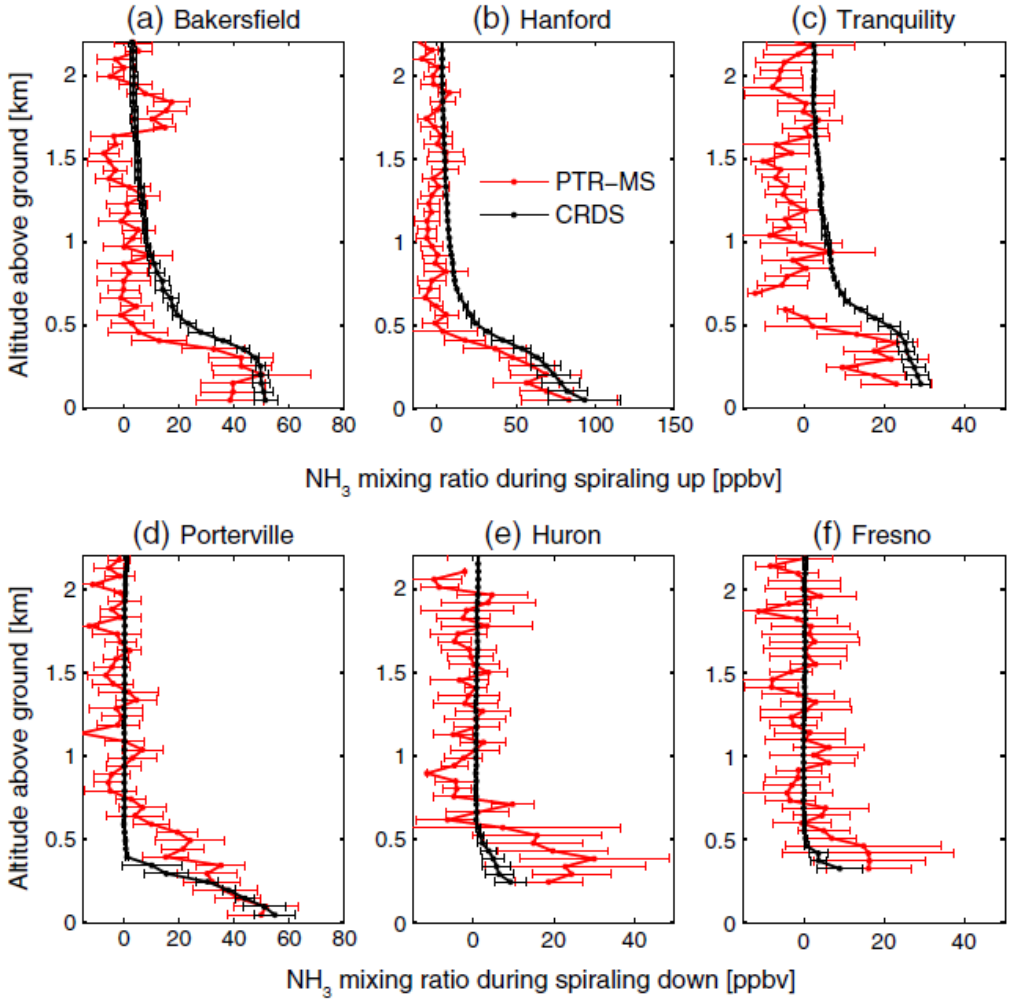
GEOS-Chem modelling using NEI 2005

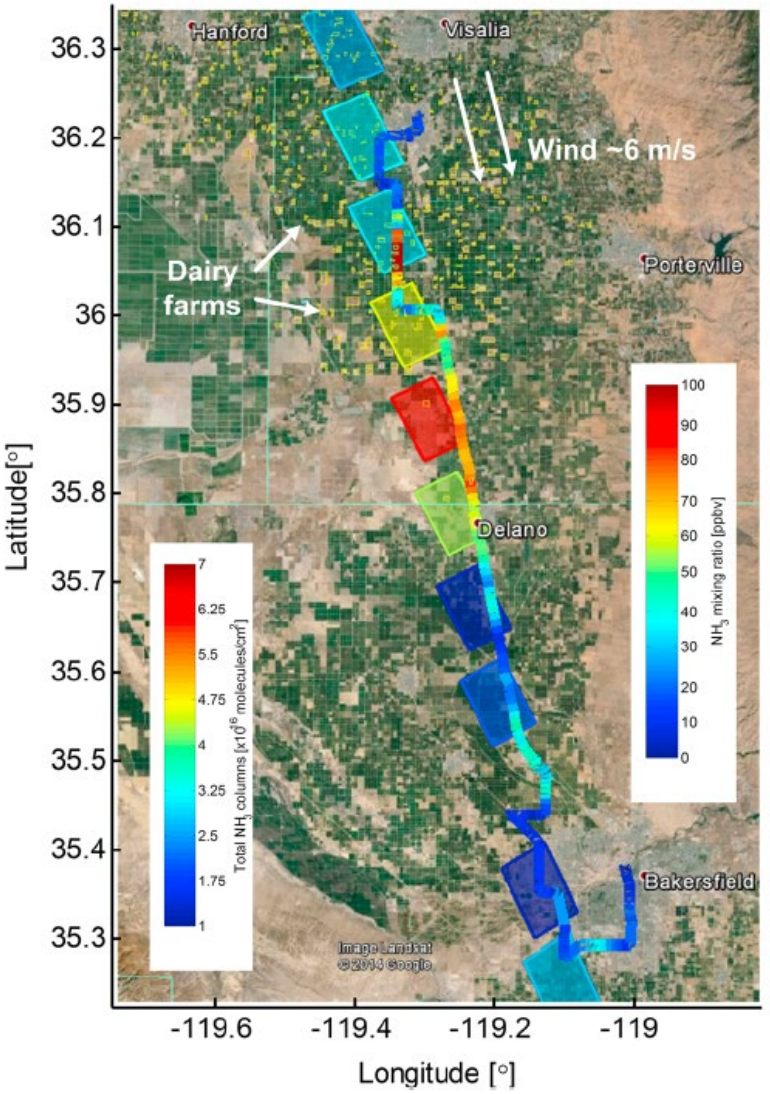




Schiferl et al., *JGR*, 2014

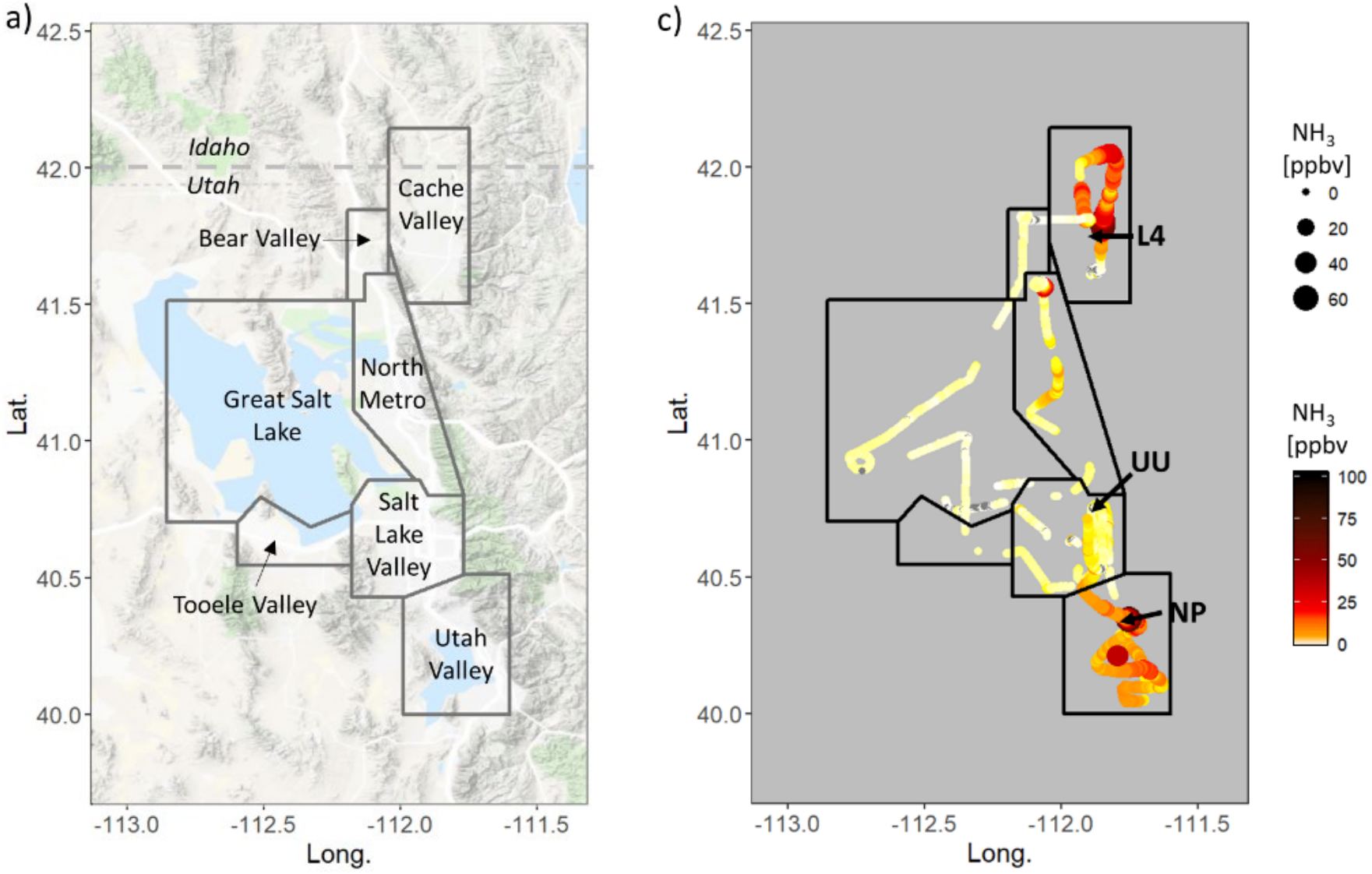
Satellite, Aircraft and Ground-based in DISCOVER-AQ





Sun et al., *JGR*, 2015

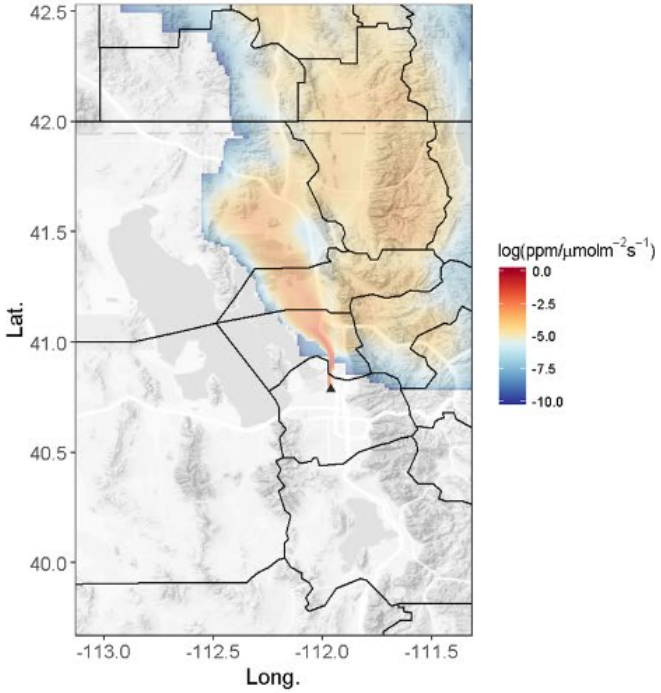
Spatial distribution of NH_3 in UWFPS

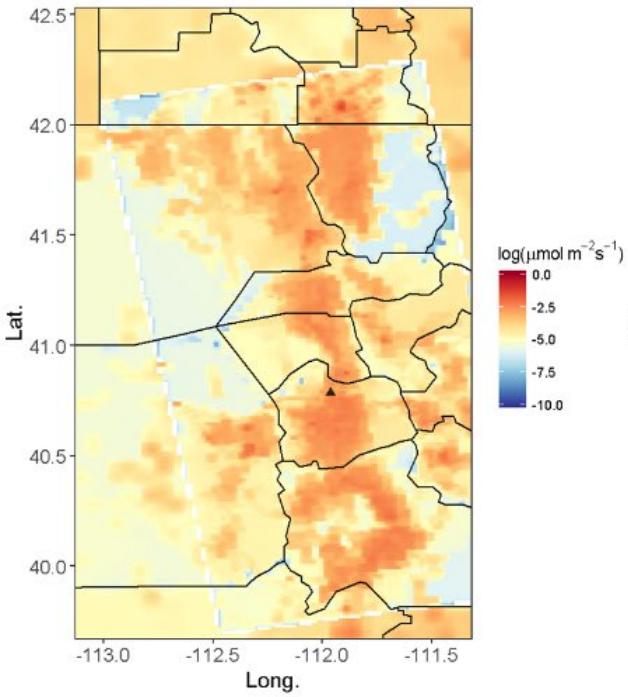


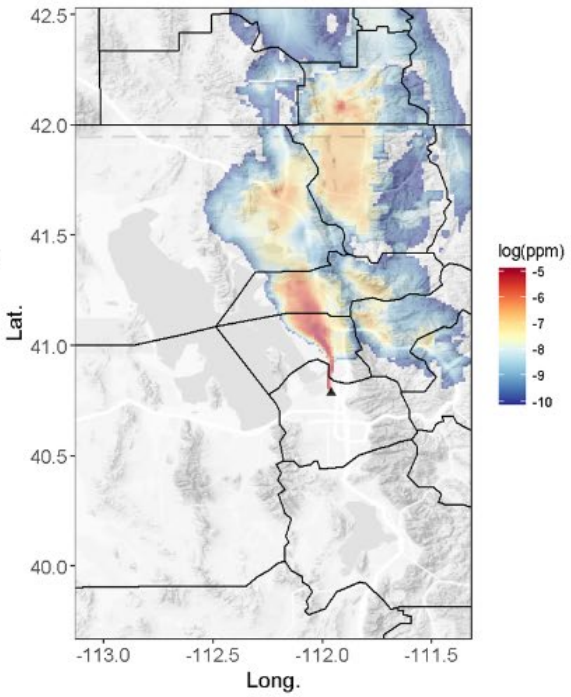
Moravek et al., *ACPD*, 2019

Constraining the NH_3 inventory

STILT footprint x NH_3 emission inventory = NH_3 contribution







Using HRRR met fields at 3x3 km

UDAQ based on NEI 2014

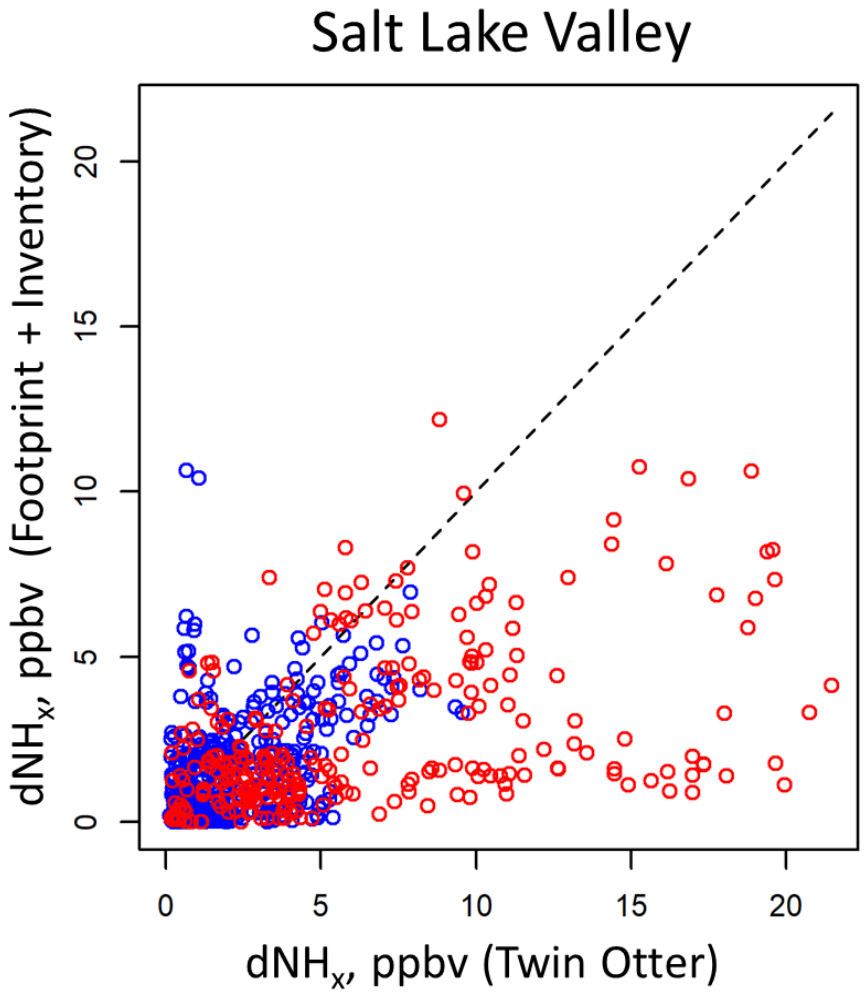
sum

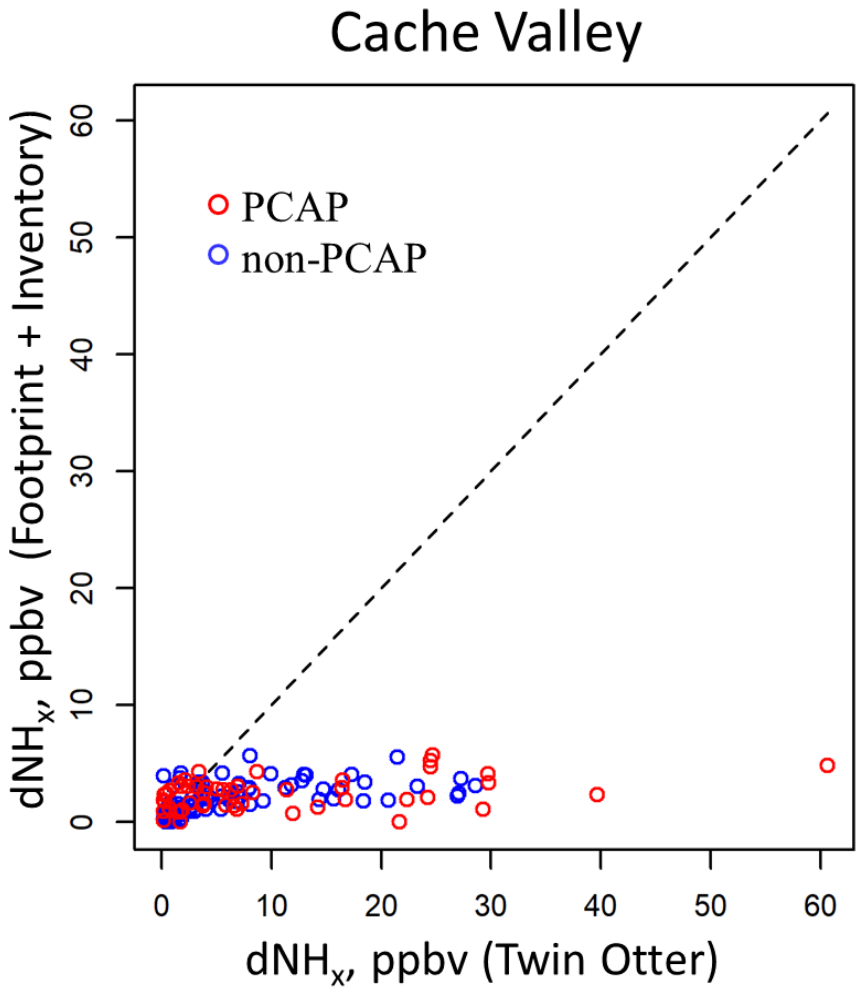
NH_3 mixing ratio at receptor (0.87 ppbv)

Compare to measurements

Moravek et al., *ACPD*, 2019

Underestimate of NH_3 emissions in Utah

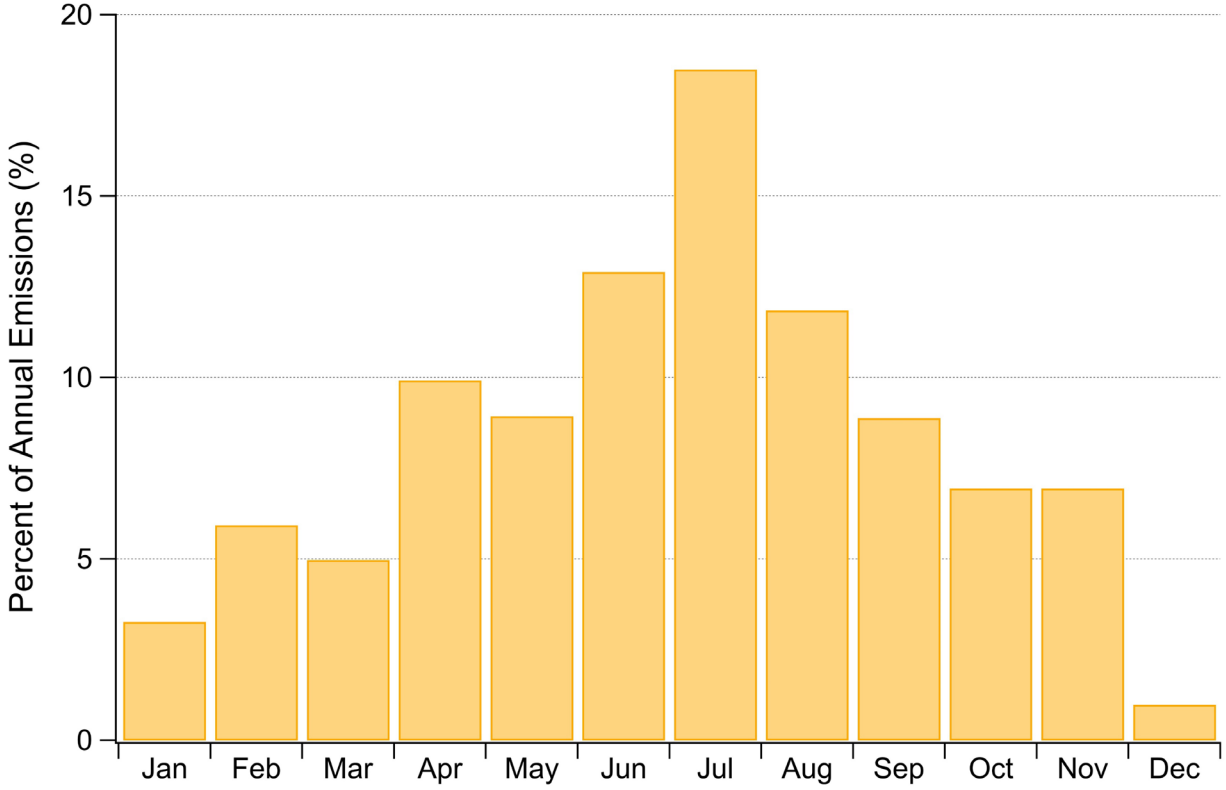




Wintertime emissions from animal husbandry 4x too low in 2014 UDAQ inventory

Moravek et al., *ACPD*, 2019

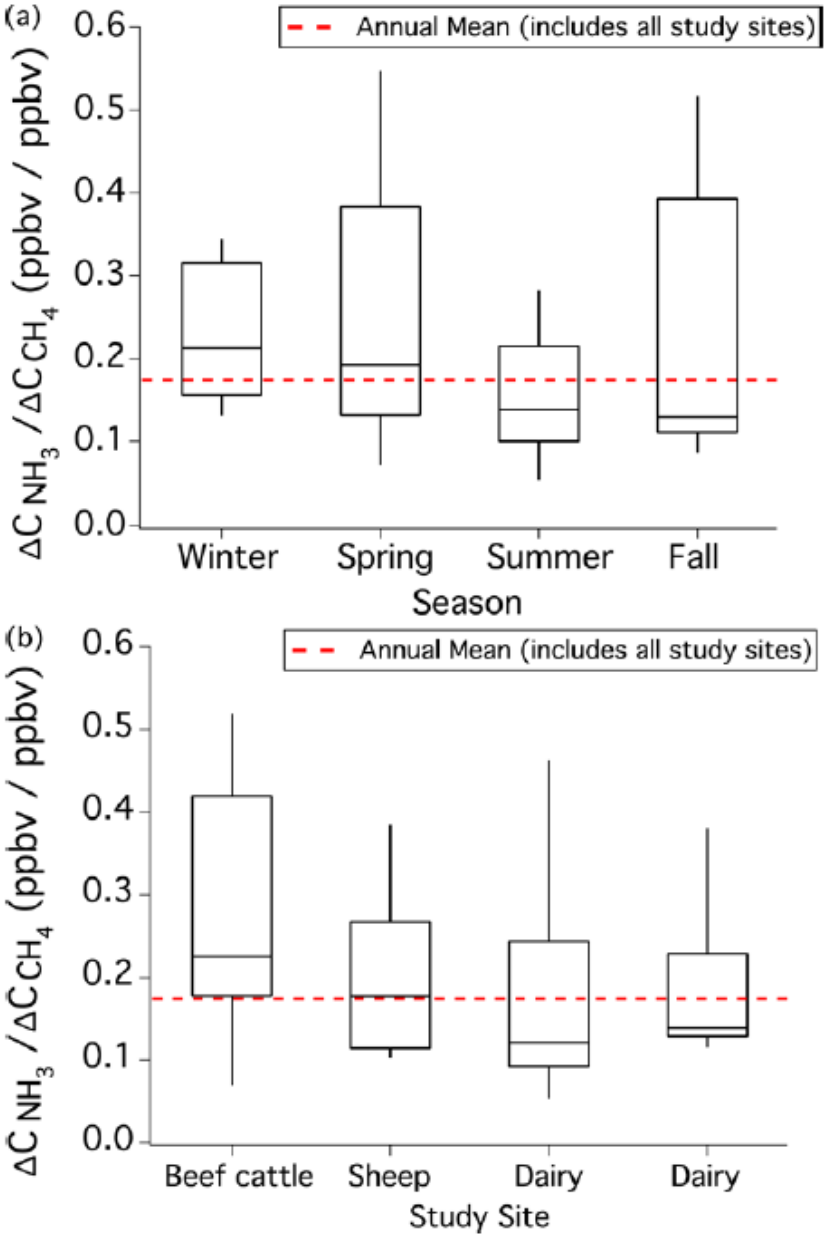
Large seasonality for livestock NH_3 emissions in UDAQ inventory



In contrast, USU inventory from Moore and Martin has equivalent emissions in July and January

Seasonal cycle imposed on annual emissions
Cycle was inferred through inverse modelling in Gilliland et al., 2006

CAFO survey in Colorado showed modest seasonality



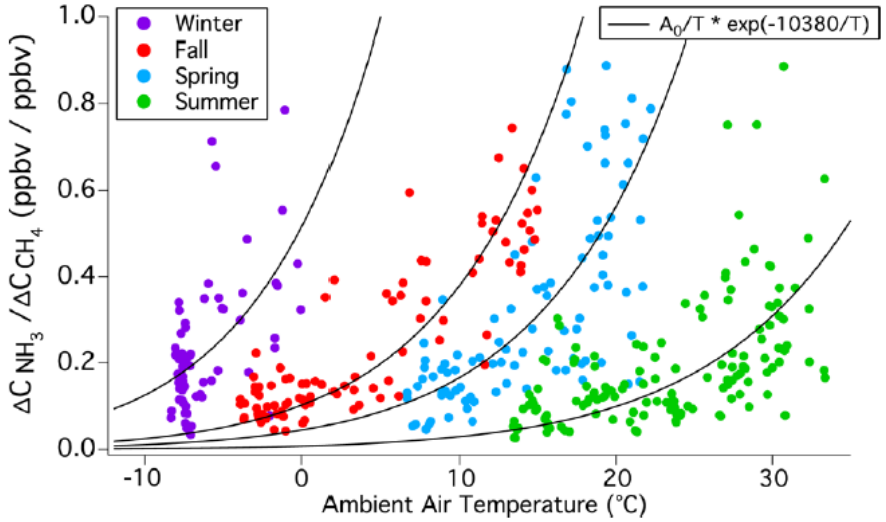
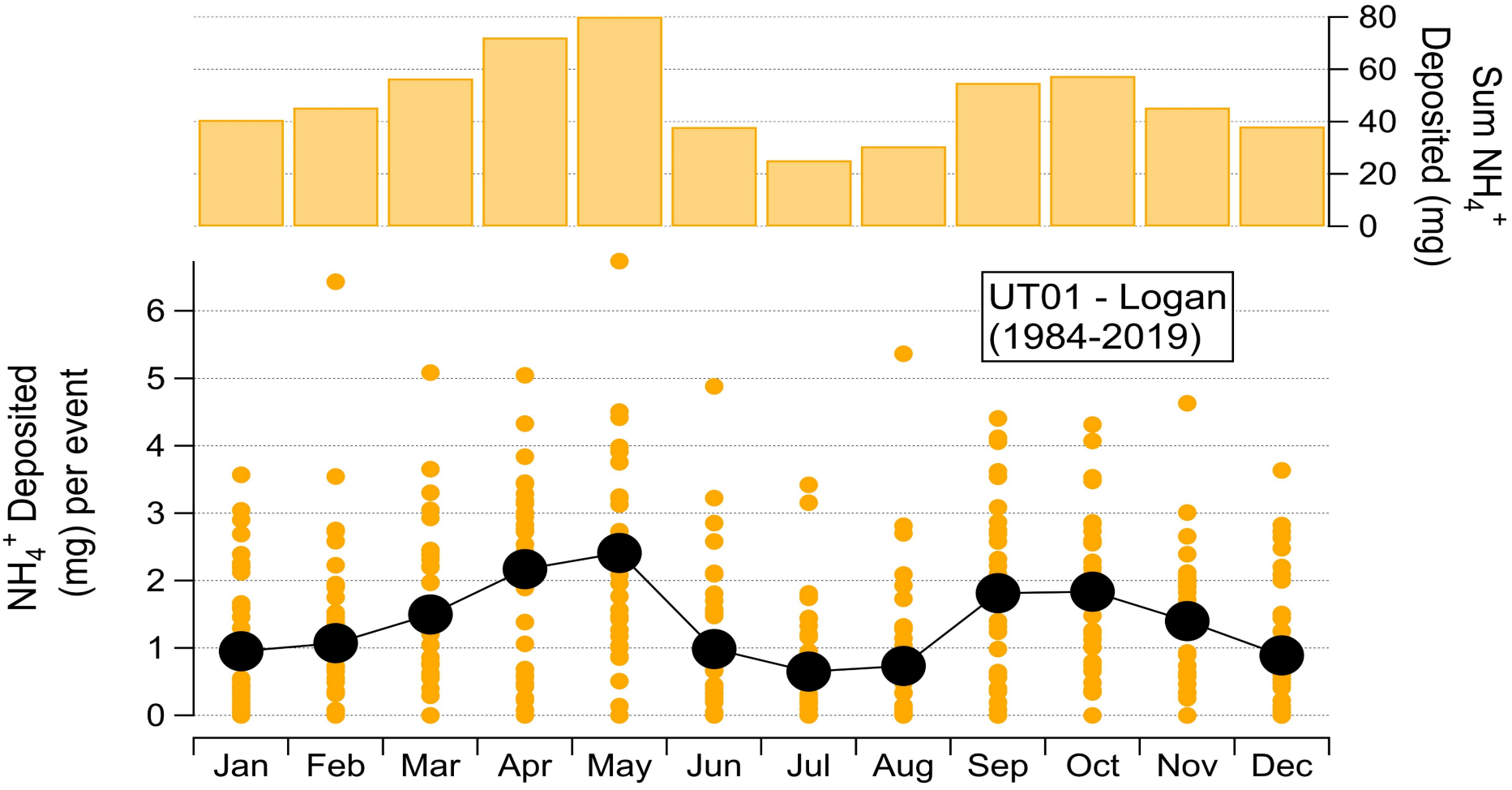


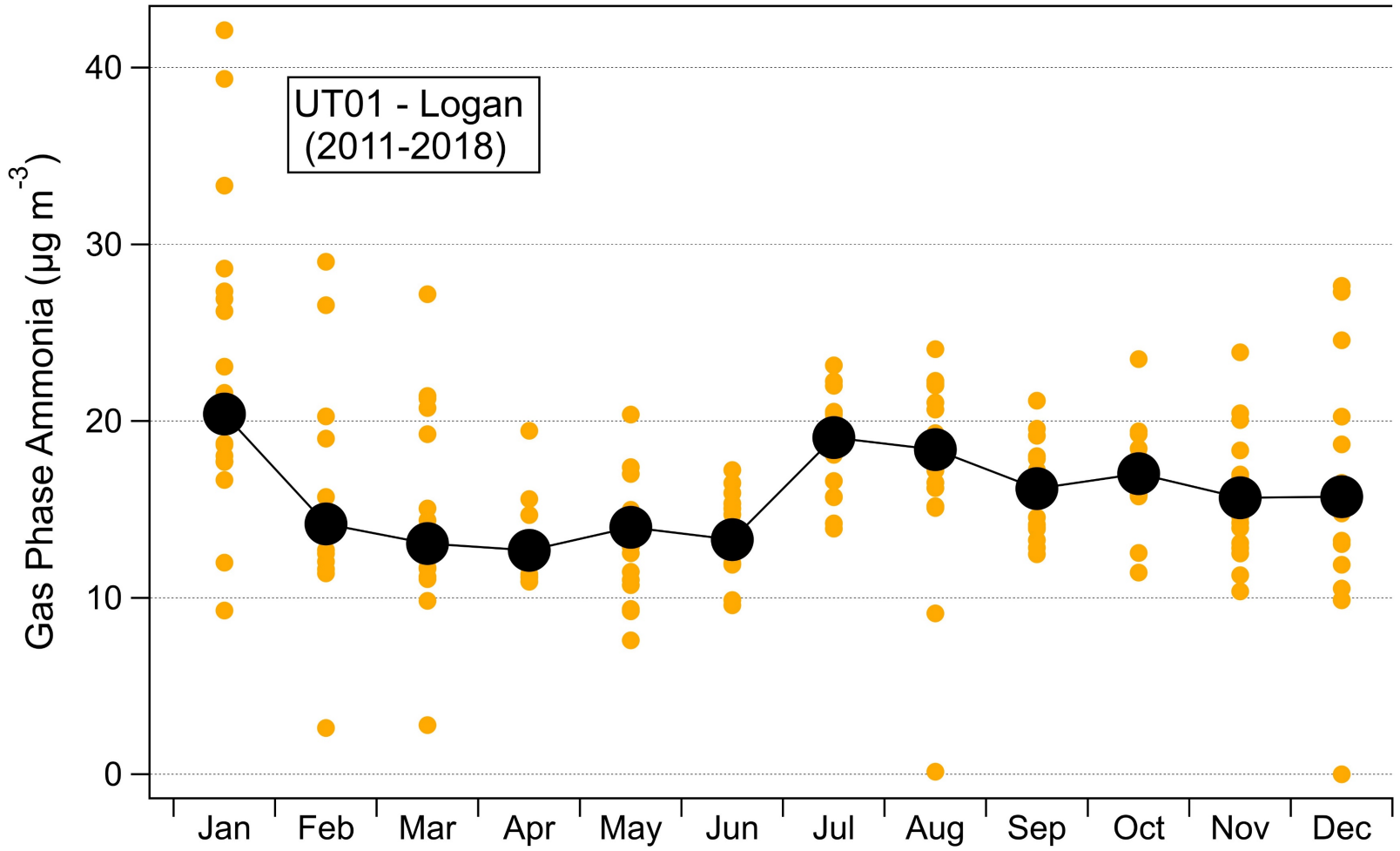
Figure 6. Temperature dependence of the $\Delta C_{NH_3}/\Delta C_{CH_4}$ enhancement ratio in each season. The black lines represent the temperature dependence of the ammonia volatilization process and are scaled by a multiplicative factor A_0 for each season.

Eilerman et al., *ES&T*, 2016

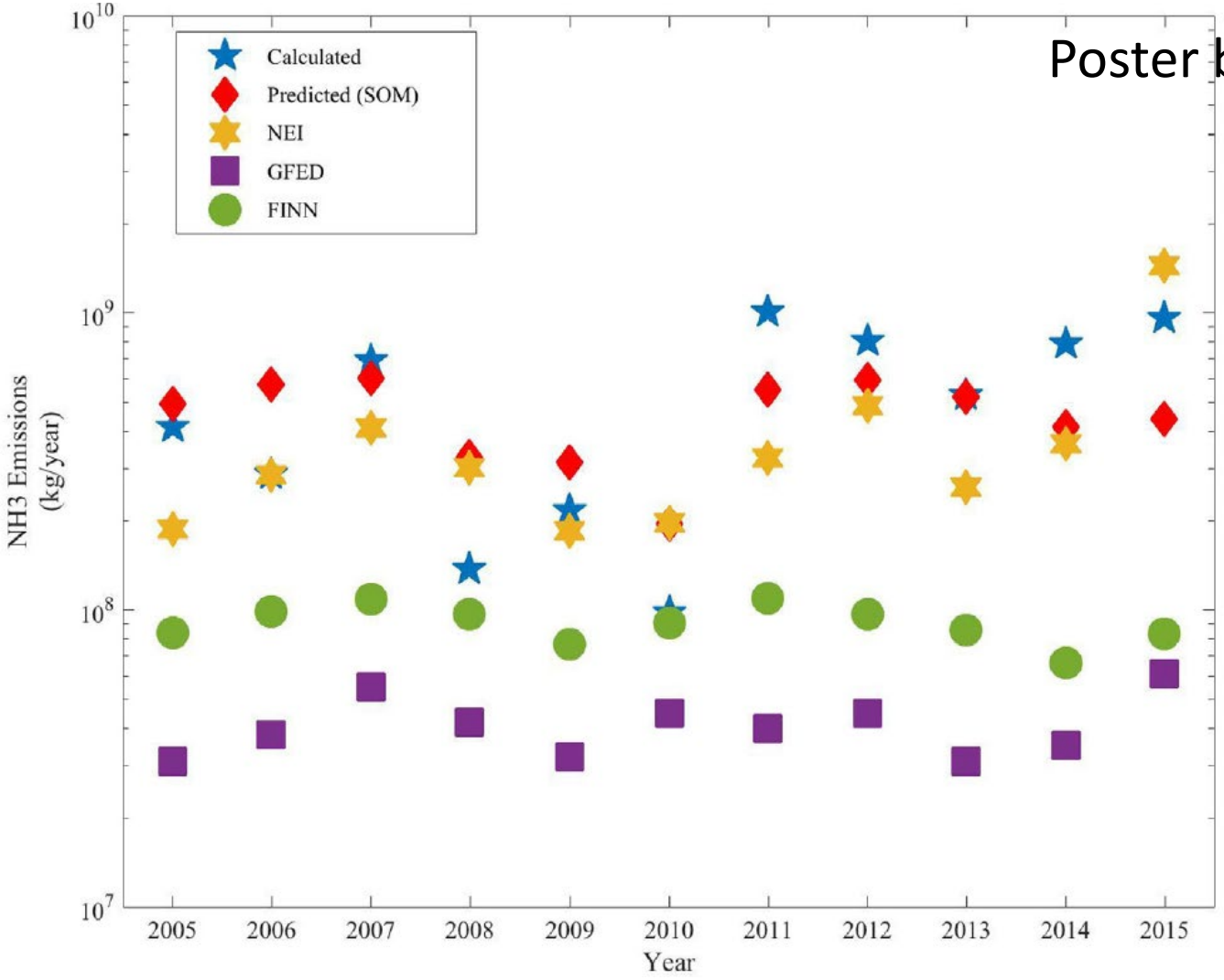
NH_x deposition at Logan (NADP)



NH_3 concentration at Logan (AMoN)



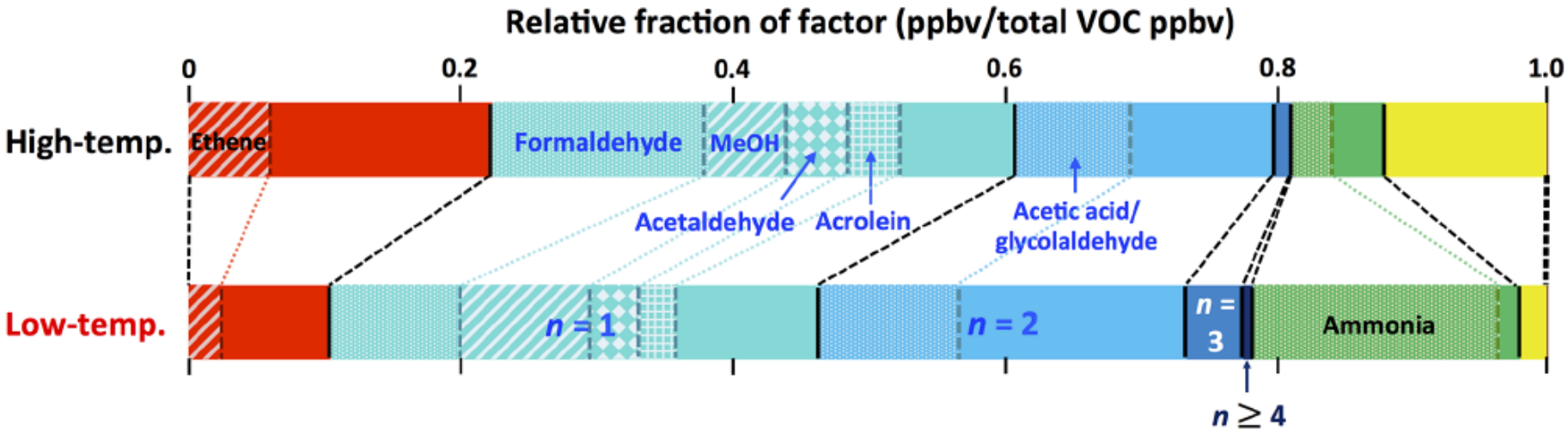
Biomass Burning Sources of NH_3



Poster by Viney Aneja

Bray et al., *Atm Env*, 2018

Biomass Burning Sources of NH_3



	High-T	Low-T		High-T	Low-T
Hydrocarbons	0.223	0.105	N-containing	0.067	0.196
Oxygenates (number of oxygen : *n*)			N and O-containing	0.119	0.019
n = 1	0.386	0.359	S-containing	0.001	0.001
n = 2	0.190	0.270	Others	0.000	0.000
n = 3	0.014	0.041			
n ≥ 4	0.001	0.008			

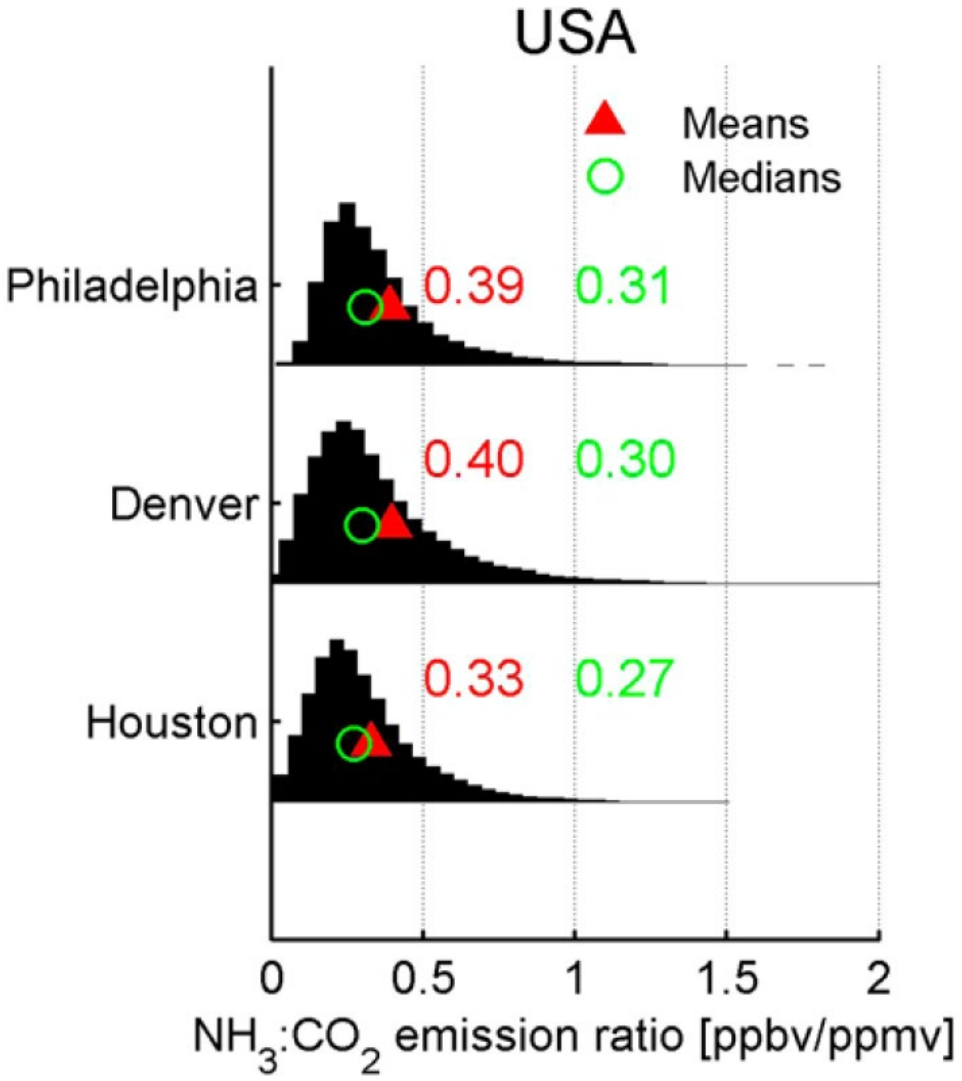
Sekimoto et al., *ACP*, 2018

Mobile Sources of NH_3

Can be diagnosed through emission ratios with CO, CO_2, NO_x

Gasoline vs diesel?

Differences in emission ratios during winter operating conditions?



Sun et al., *ES&T*, 2016

Key Questions

- How does the sensitivity of $PM_{2.5}$ to NH_3 vary
  - spatially across the western U.S. in winter?
  - through time over the course of an extreme episode?

Requires: extensive (ground-based, aircraft, satellite), high-time resolution (hourly or better) measurements of NH_3 and p-NH_4^+ (plus HNO_3 and other $PM_{2.5}$ chemistry)

- What are the emissions of NH_3 for each sector and how do they vary seasonally?

Requires: extensive (ground-based, aircraft, satellite), high-time resolution (hourly or better) measurements of NH_3 and p-NH_4^+ (plus co-emitted species CO, CO_2, CH_4…)

NH_3 concentration at SLC (AMoN)