Inverse Modeling to Estimate Seasonal NH_3 Emission Estimates

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Problem Statement:

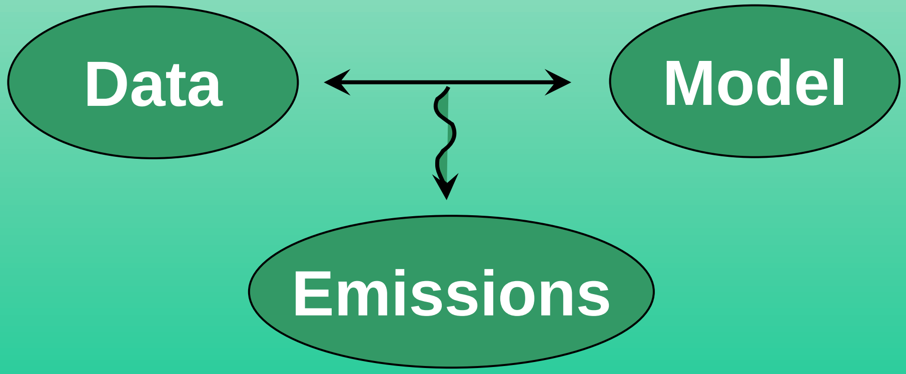
- NEI NH_3 Inventory provides annual total emission estimates, but
- We expect seasonality in emission rate because of source types
  - NH_3 emissions from animal waste are a function of temperature
  - Fertilizer application patterns are seasonal

1990 National Emissions Inventory (NEI): Annual U.S. Ammonia Emissions (thousand short tons)		
Livestock	3307	76%
Fertilizer	420	10%
On-road vehicles	192	4%
Chemical and Allied Product Manufacturing	183	4%
Other (Industrial Processes, Off-road vehicles)	229	5%
TOTAL	4,331	

USEPA, *Nat'l Air Pollutant Emission Trends (1990-98)*, EPA-454/R-00-002 [2000]

Objective and Approach:

- Inverse modeling to estimate monthly emissions for NH_3
- Compare NADP wet [NH_4^+] with air quality model results
- Inverse method estimates emissions that result in optimized model results for wet [NH_4^+]
- Using monthly emission estimates, re-run air quality model to consider impact on model predictions of related species



Inverse Methodology for this study

- Method follows existing approach *(Chang et al, 1996, Gilliland and Abbitt, 2001)*
- Kalman Filter nomenclature
- Applied recursively at 1 time increment *(iterations unnecessary due to linearity)*
- Similarities to optimal estimation via cost function minimization
  - *e.g., Palmer et al., 2003; Jacob et al., 2002*
  - *Contribution of emissions during the time increment is tested here while Palmer et al. and Jacob et al. consider accumulated contribution from an emission source*

$$E_t^{post}(\mathrm{m}) = E_t^{prior}(\mathrm{m}) + G(\mathrm{m}\times\mathrm{n})\left(\chi_t^{\mathrm{obs}}(\mathrm{n}) - \chi_t^{\mathrm{model}}(\mathrm{n})\right)$$

$$G = S_{\mathrm{a}}K^T\left(KS_{\mathrm{a}}K^T + S_\Sigma\right)^{-1}$$

Variance of error in the concentration (aka, Noise; observational uncertainty of 4%; add’l tests of model uncertainty)

$$K = \frac{\partial f}{\partial E};$$

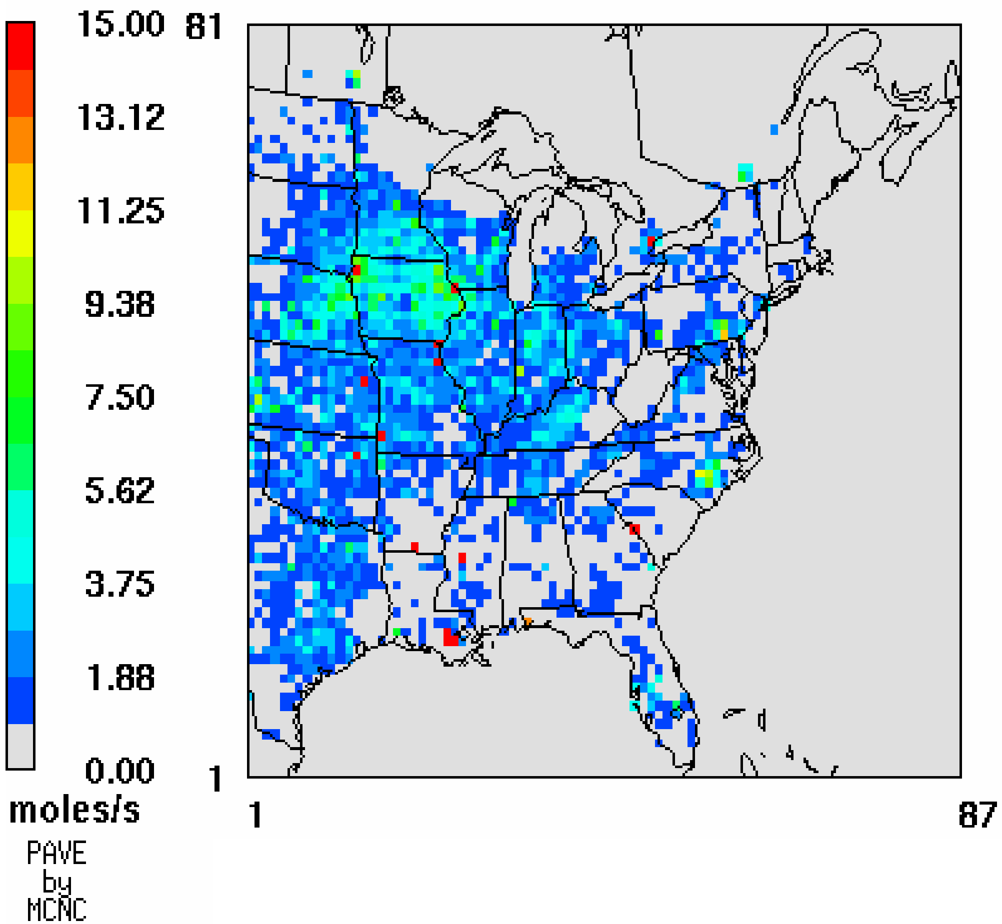
Variance of error in emissions (40% uncertainty estimated, Asman et al.)

$$\mathrm{y} = \chi_{\mathrm{t+1}} - \chi_{\mathrm{t}} = f(\ ) = AQModel$$

Community Multiscale Air Quality (CMAQ) Model

- Results based on 2001 CMAQ Release
- Regional scale domain
  - 36km horizontal grids
  - 21 vertical layers
  - Eastern US
- RADM2 chemical mechanism
  - Aerosol v.2
- Emissions
  - 1990 USEPA National Emissions Inventory
  - Mobile 5b
  - BEIS2

Average NEI NH_3 emissions



For inverse application, entire domain is treated as 1 source region *m*.

http://nadp.sws.uiuc.edu/

National Atmospheric Deposition Program

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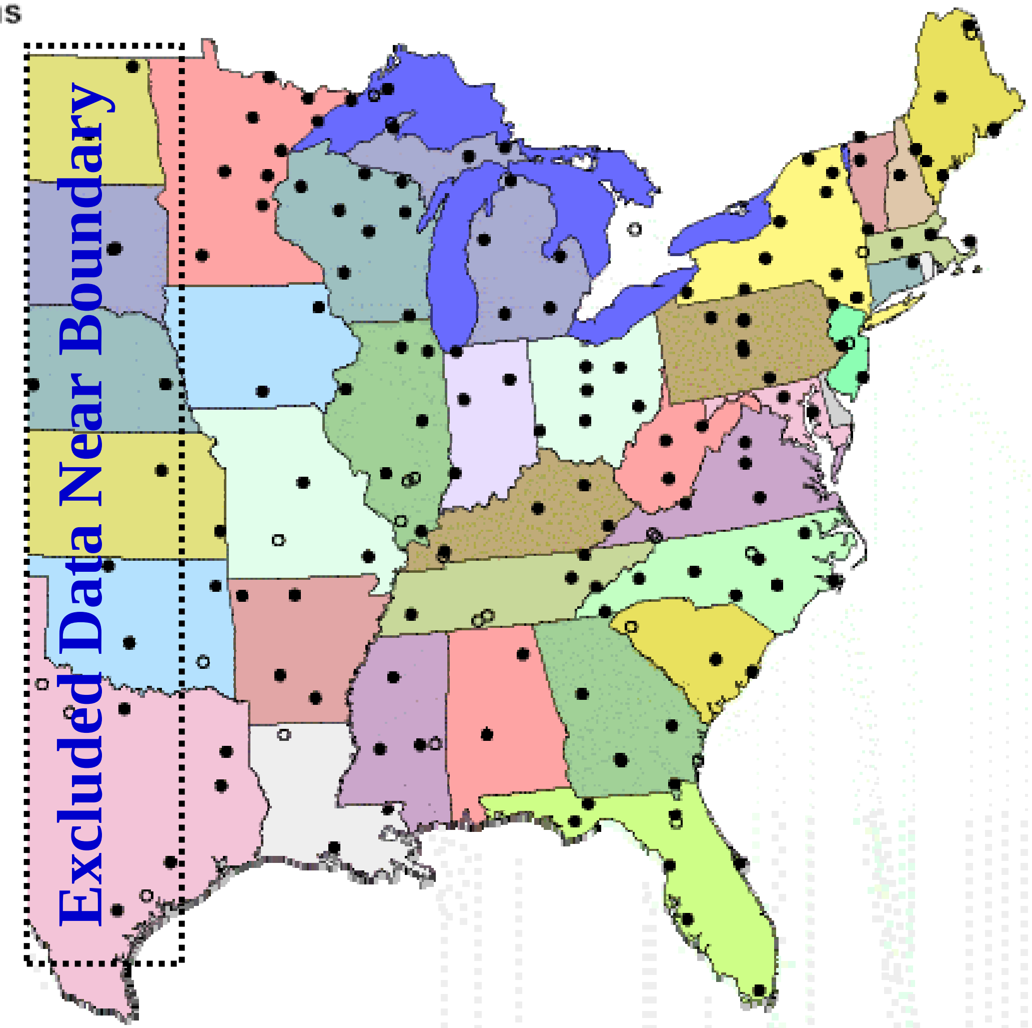
- $[NH_4^+]$ wet concentrations used in inverse modeling
- 15% bias

  [Butler and Likens, 1998; Gilliland et al., 2002]

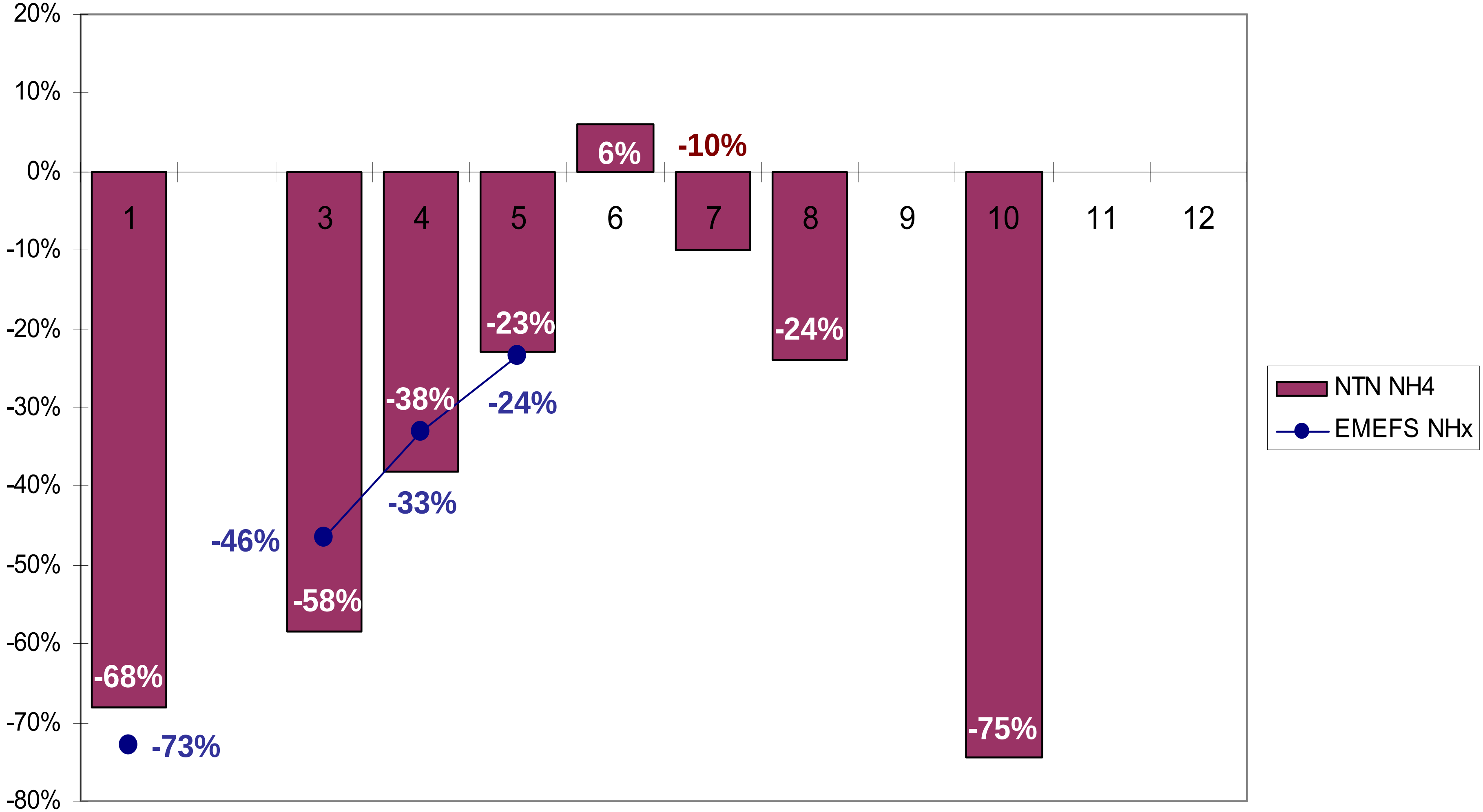


Also…

- *EMEFS $[NH_x]$*
- *CASTNET $[NO_3]$, $[NH_4]$, and $[SO_4]$*

SUMMARY of SEASONAL ADJUSTMENTS

(S_{Σ} based on 4% relative uncertainty * Obs)



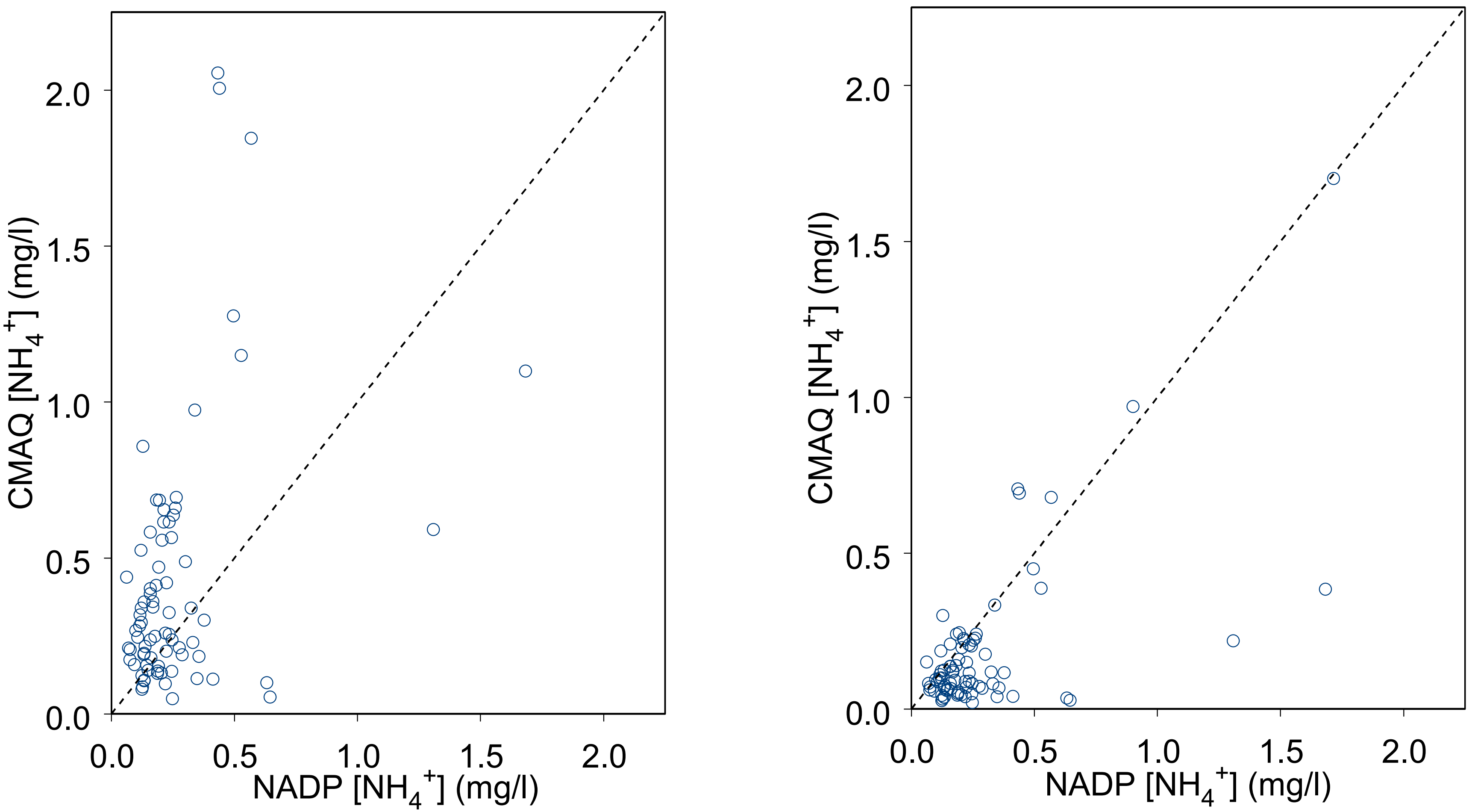
Jan 1990

NADP [NH_4^+]: Before Emission Change

RMSE= 0.52 mg/l, R= 0.65

NADP [NH_4^+]: After 68% Emission Decrease

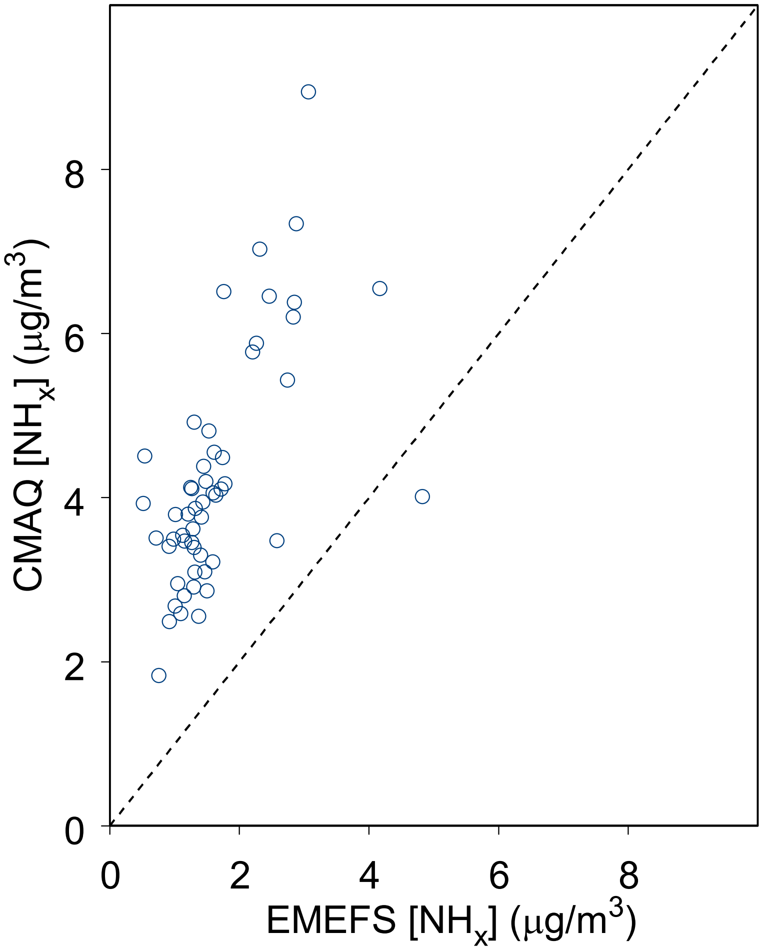
RMSE= 0.25 mg/l, R= 0.67



Jan 1990

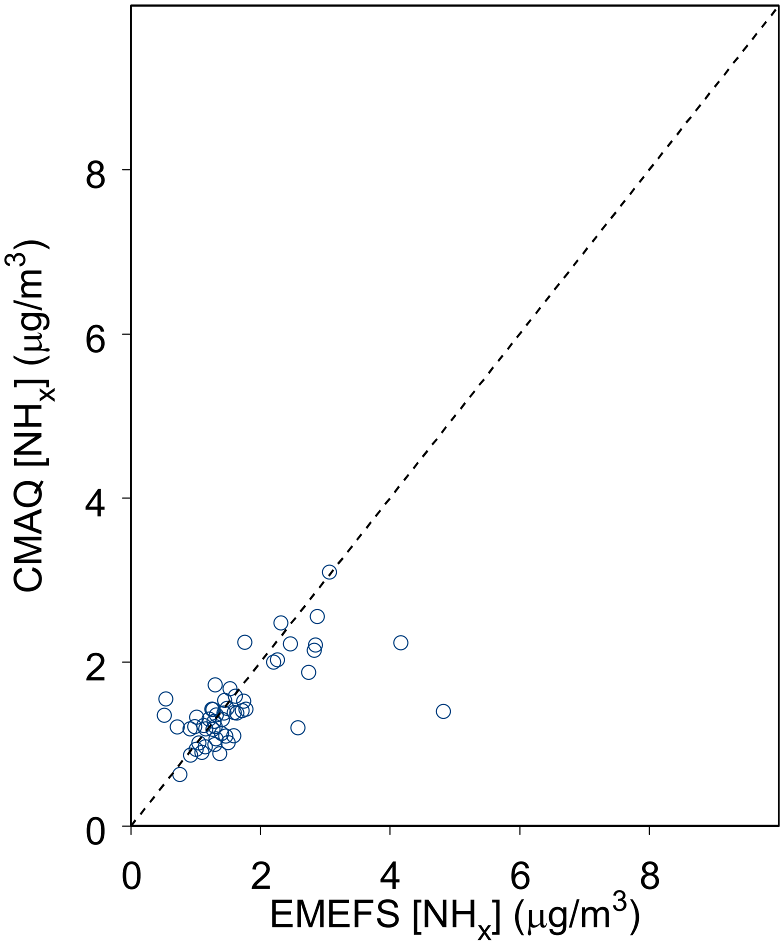
EMEFS [NH_x]: Before Emission Change

RMSE= 2.79 $\mu g/m^3$, R= 0.64

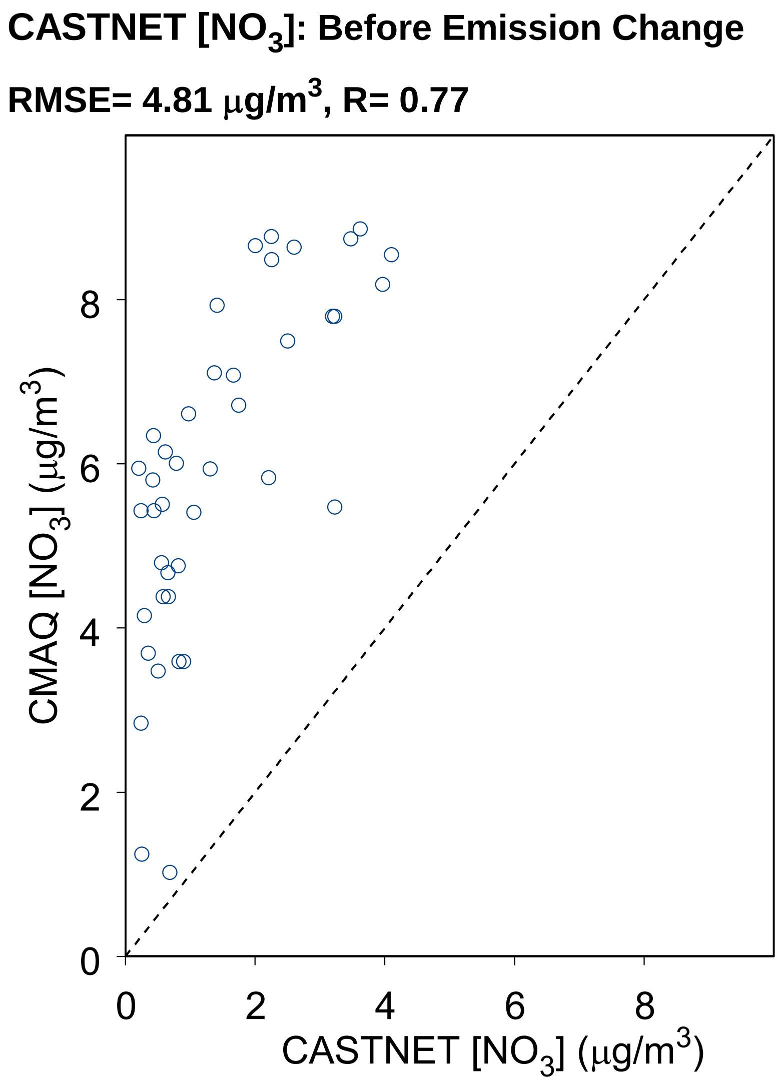


EMEFS [NH_x]: After 68% Emission Decrease

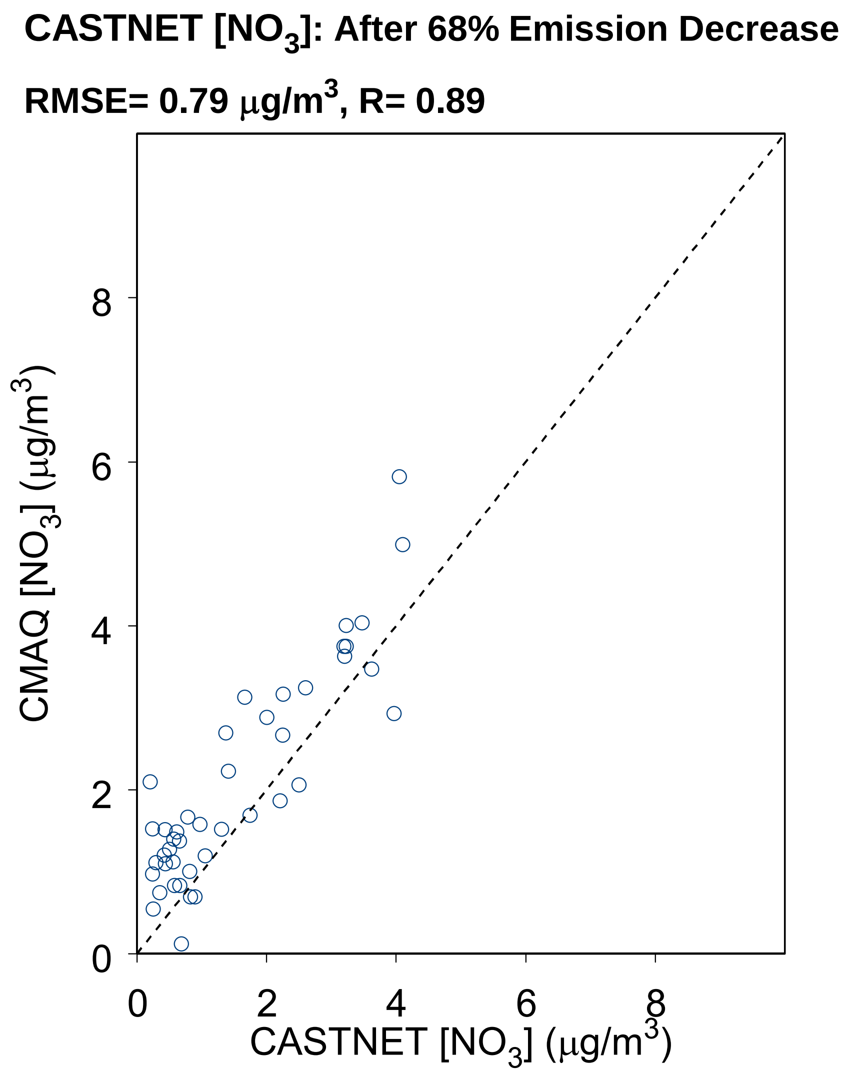
RMSE= 0.67 $\mu g/m^3$, R= 0.64



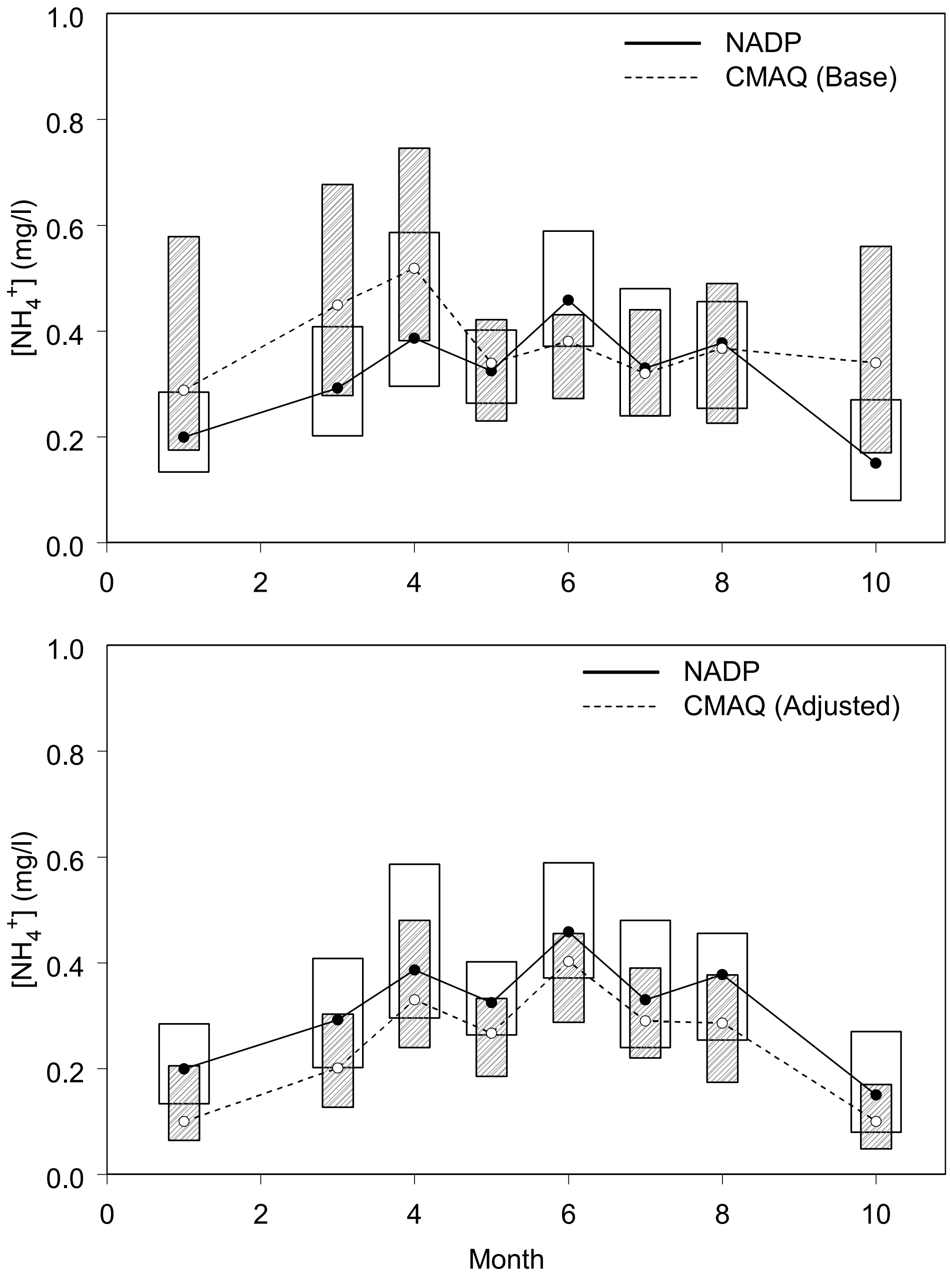
Jan 1990



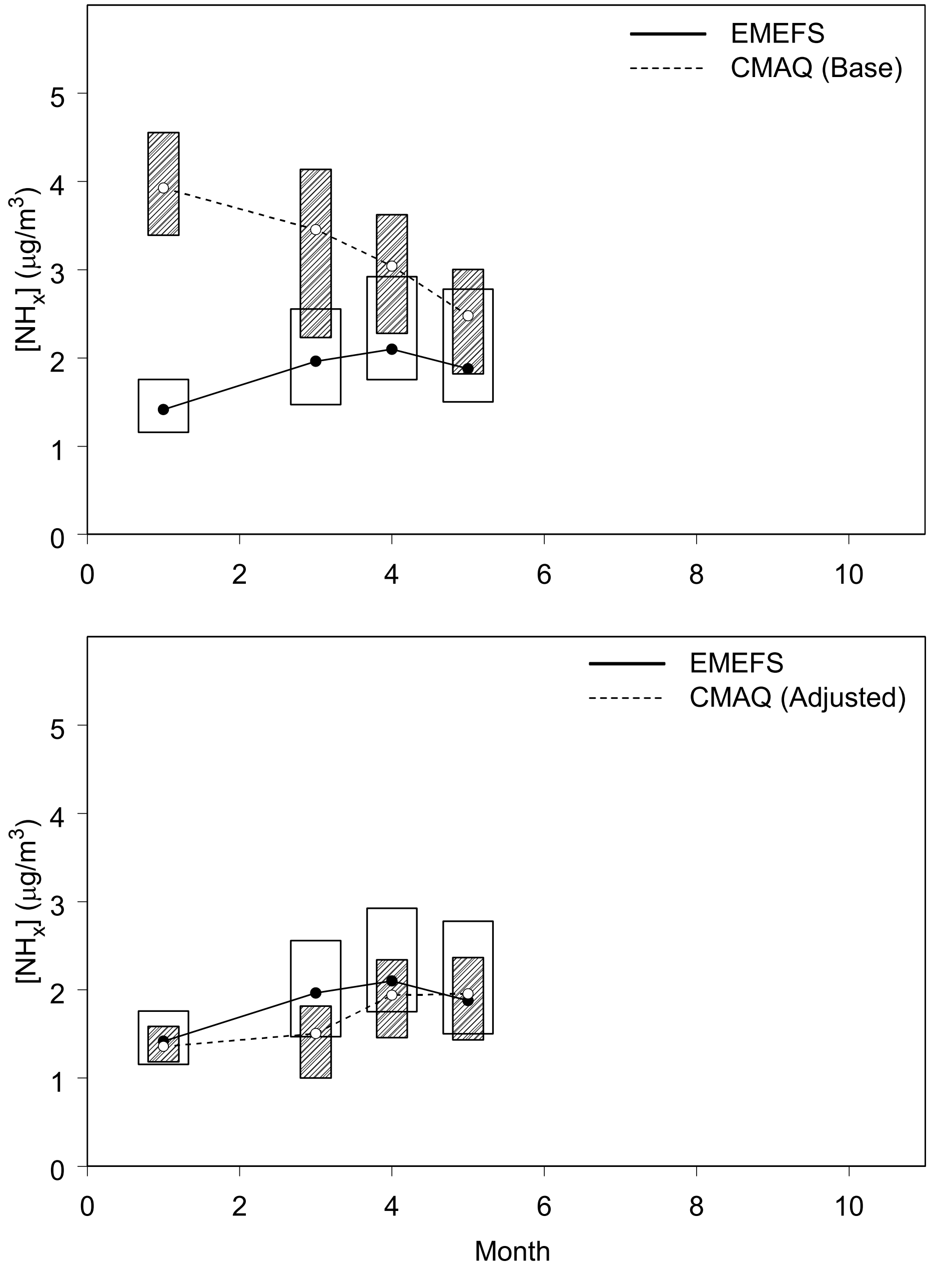
CASTNET [NO3]: Before Emission Change
RMSE= 4.81 μg/m3, R= 0.77
CMAQ [NO3] (μg/m3)
CASTNET [NO3] (μg/m3)
0
2
4
6
8



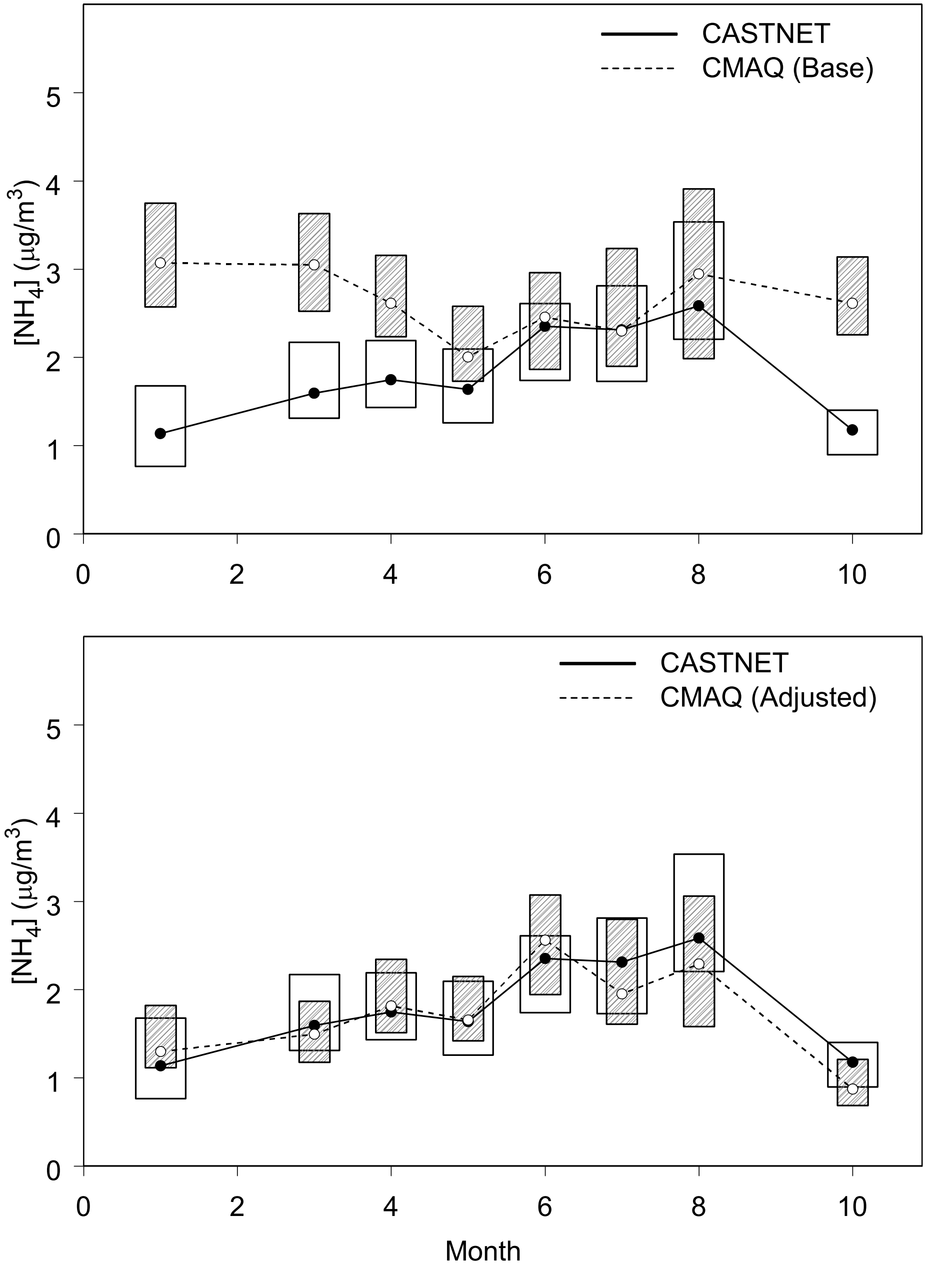
CASTNET [NO3]: After 68% Emission Decrease
RMSE= 0.79 μg/m3, R= 0.89
CMAQ [NO3] (μg/m3)
CASTNET [NO3] (μg/m3)
0
2
4
6
8



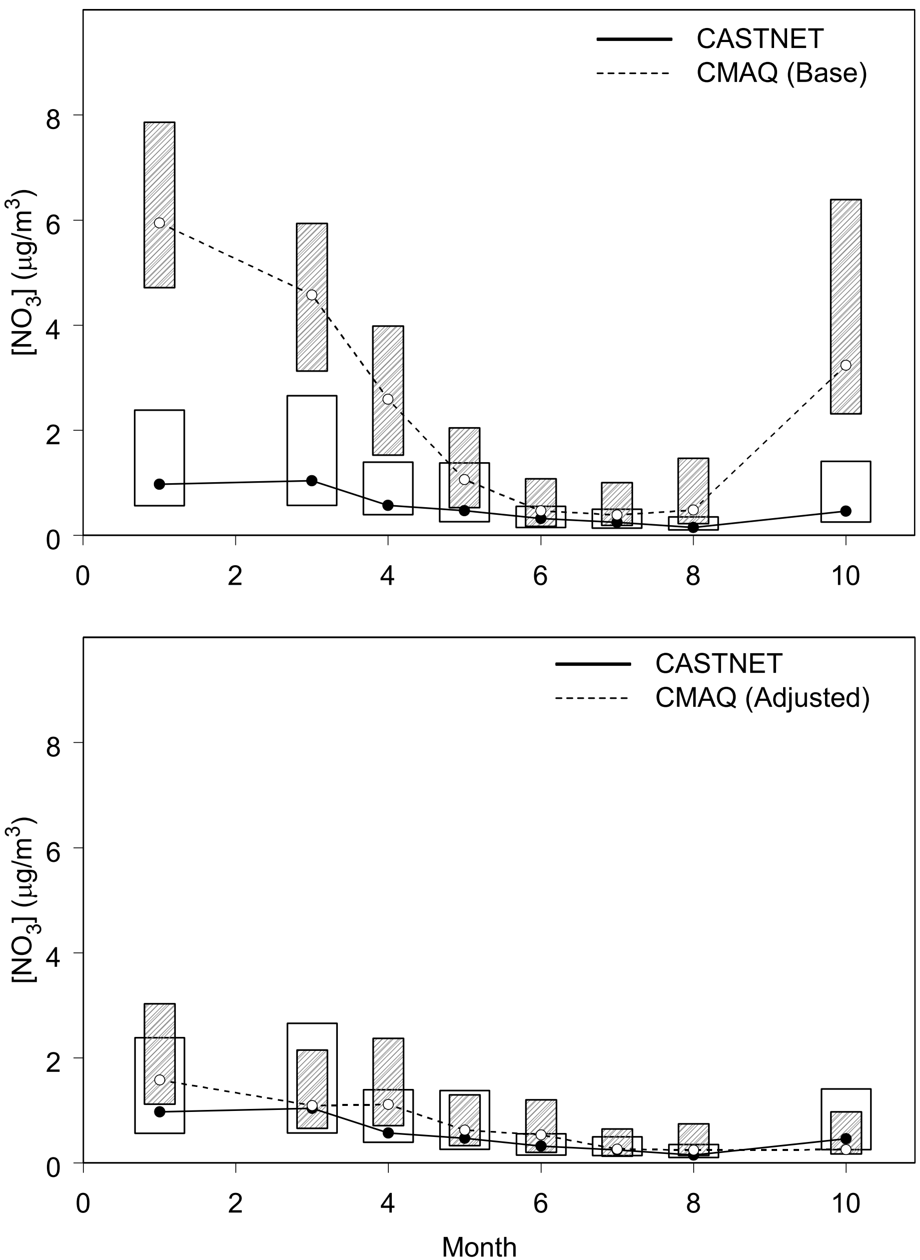
NADP
CMAQ (Base)
[NH4+] (mg/l)
1.0
0.8
0.6
0.4
0.2
0.0
0
2
4
6
8
10
NADP
CMAQ (Adjusted)
[NH4+] (mg/l)
1.0
0.8
0.6
0.4
0.2
0.0
0
2
4
6
8
10
Month



EMEFS
CMAQ (Base)
[NH$_x$] (μg/m^3)
0
1
2
3
4
5
0
2
4
6
8
10
EMEFS
CMAQ (Adjusted)
[NH$_x$] (μg/m^3)
0
1
2
3
4
5
0
2
4
6
8
10
Month



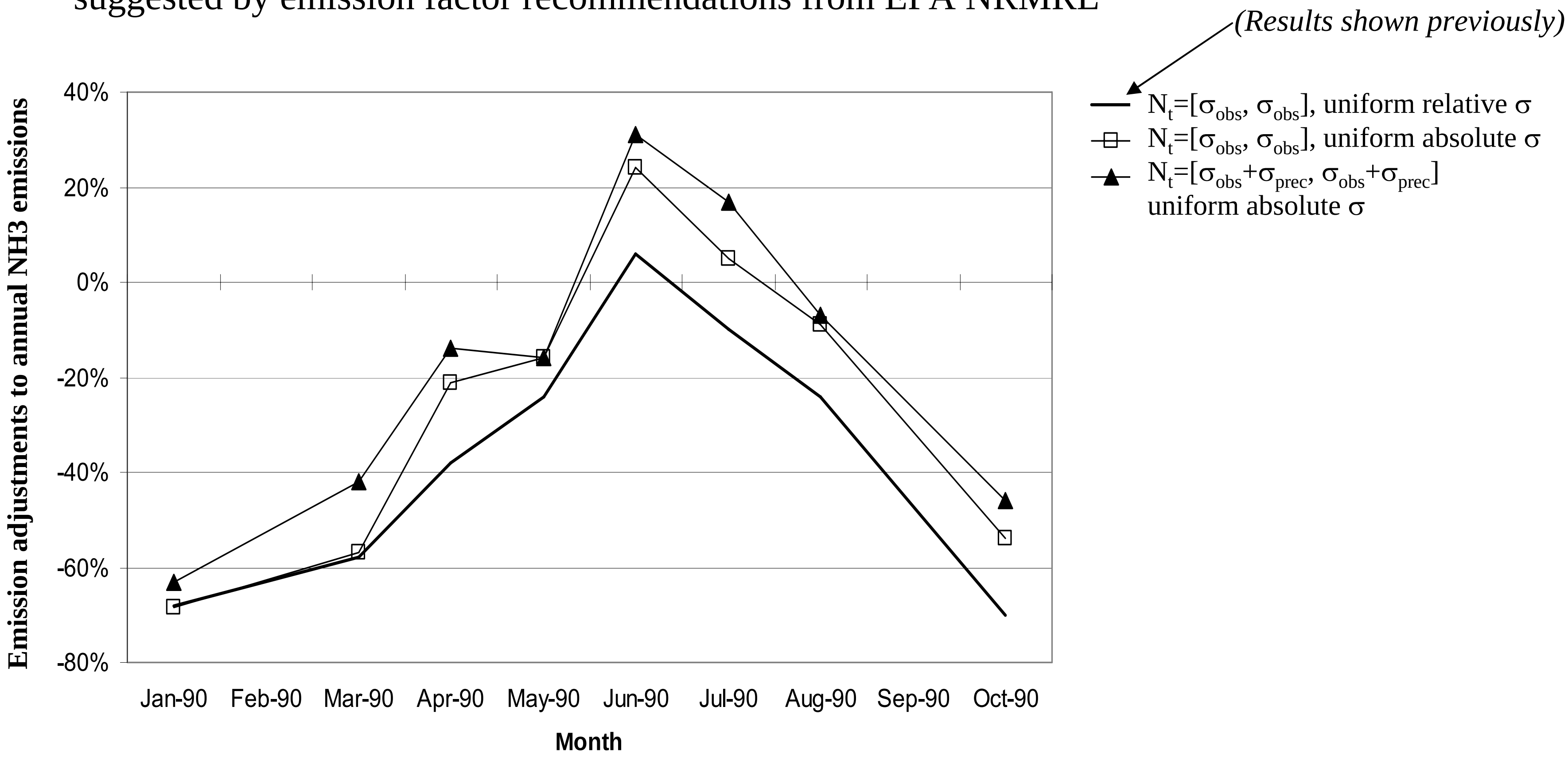
CASTNET
CMAQ (Base)
[NH4] (μg/m3)
0
1
2
3
4
5
0
2
4
6
8
10
CASTNET
CMAQ (Adjusted)
[NH4] (μg/m3)
Month



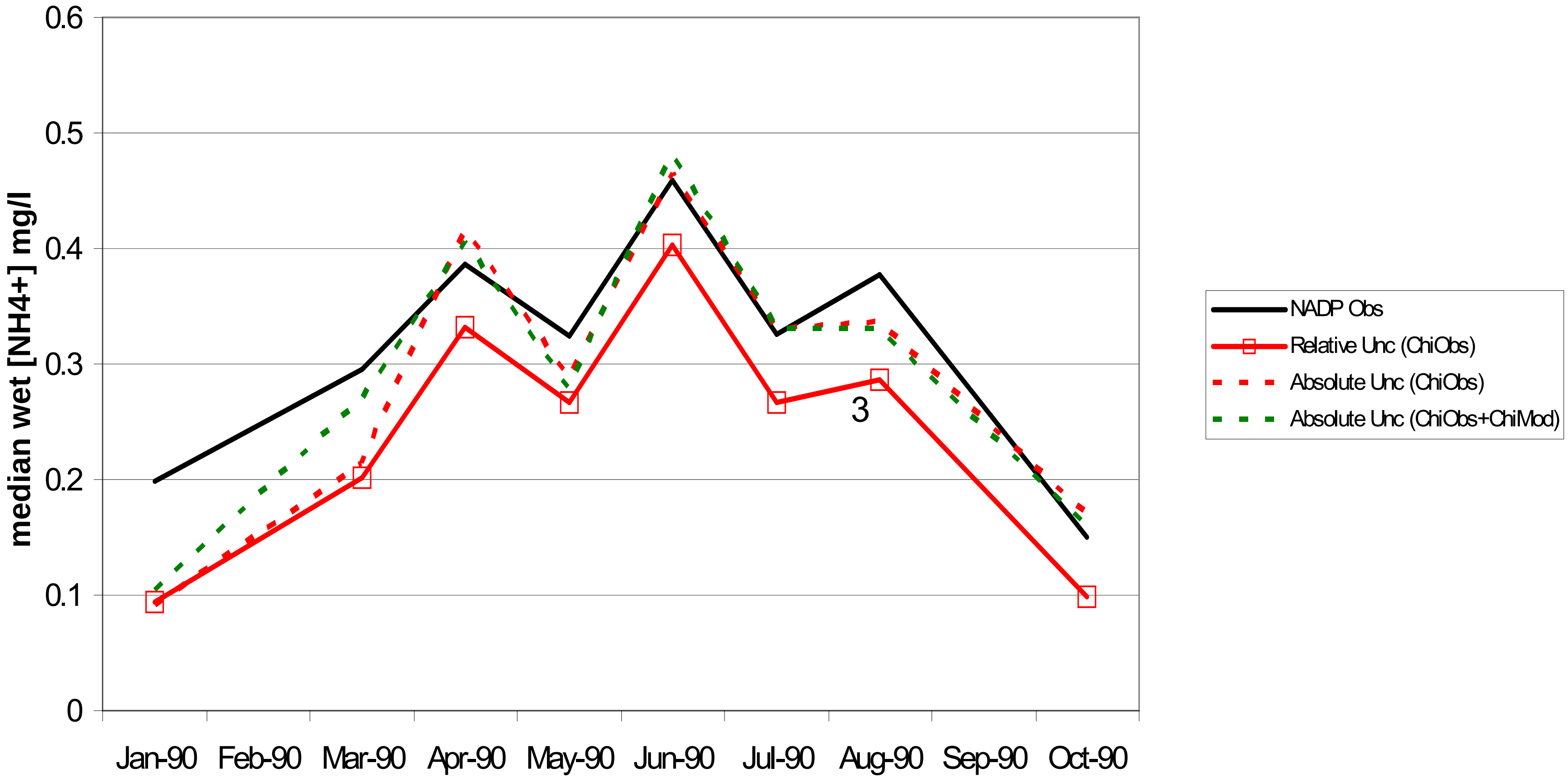
CASTNET
CMAQ (Base)
[NO3] (µg/m3)
CASTNET
CMAQ (Adjusted)
[NO3] (µg/m3)
Month
0
2
4
6
8
10

Sensitivity to $\mathbf{S}_\Sigma$ error representation:

- Additional adjustments calculated using uniform absolute uncertainty (square marker) and model precipitation uncertainty (solid triangle)
- Similar seasonality seen in all three results
- Biggest differences between using uniform absolute and relative uncertainties
- All results suggest total annual emissions are too high, which has also been suggested by emission factor recommendations from EPA NRMRL



Estimates of wet [NH_4^+]
based on this range of emission adjustments:



In Summary…

- Strong seasonal variability in NH_3 emissions suggested (as expected)
- Results also suggest annual NH_3 NEI inventory is high
- Using seasonally adjusted NH_3 emissions substantially improves model predictions of wet [NH_4^+], aerosol [NH_4^+], ambient [NH_x], and aerosol [NO_3^-] in winter and early spring

- Method very good for qualitative estimates (e.g., seasonal variations)
- Quantitative adjustment estimates are sensitive to representation of uncertainty
- Useful to have comparisons with other approaches

For more information on results presented here:

Gilliland, Dennis, Roselle, and Pierce, Seasonal NH3 emission estimates for the eastern United States based on ammonium wet concentrations and an inverse modeling method, JGR, 2003