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Canada

Trends of monitored nitrogen species at monitoring sites in North America

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Goals

First study:

- Analyze long-term geographical and temporal trends of NO_3^- and NH_4^+ in atmosphere and wet deposition in Canada
- Determine scavenging ratios of NO_3^- and HNO_3
- Estimate the relative contributions of particulate and gaseous nitrogen species to total nitrate and ammonium wet deposition.

Second Study

- Explore long-term trends of NH_3 and related causes at monitoring sites in Canada and U.S.
- Assess the uncertainties between different trend analysis tools

Methodology

First study:

- Daily air and wet deposition samples for major inorganic ions and trace gases. Valid air concentrations from 1983-2010 at 16 sites and precipitation measurements from 1984-2011 at 30 sites
- Temporal trends analysis using regression and the Mann-Kendall analysis (Gilbert, 1987)
- Monthly scavenging ratios - a pollutant's concentration in precipitation to that in air
- Relative contributions of gaseous and particulate species to nitrate and ammonium wet deposition using the scavenging ratio approach:

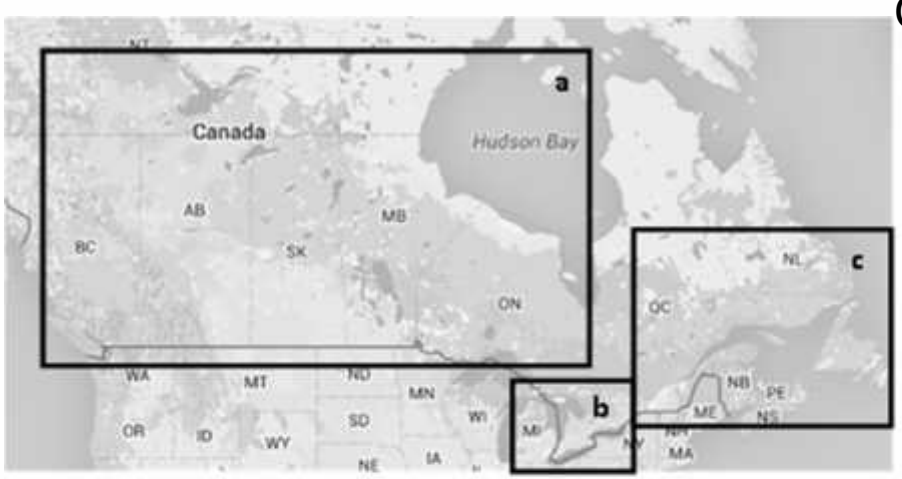
$$[pNO_3^-]_{prec} = W_{fPM}\,[pNO_3^-]_{air}\,P_f + W_{cPM}\,[pNO_3^-]_{air}\,(1-P_f),$$

$$[HNO_3]_{prec} = [\text{total } NO_3^-]_{prec} - [pNO_3^-]_{prec}$$

Second study:

- Ammonia trend analysis using two trend analysis tools: Mann-Kendall analysis (Gilbert, 1987) and the Ensemble Empirical Mode Decomposition (Wu et al., 2009)

Map of 30 CAPMoN sites



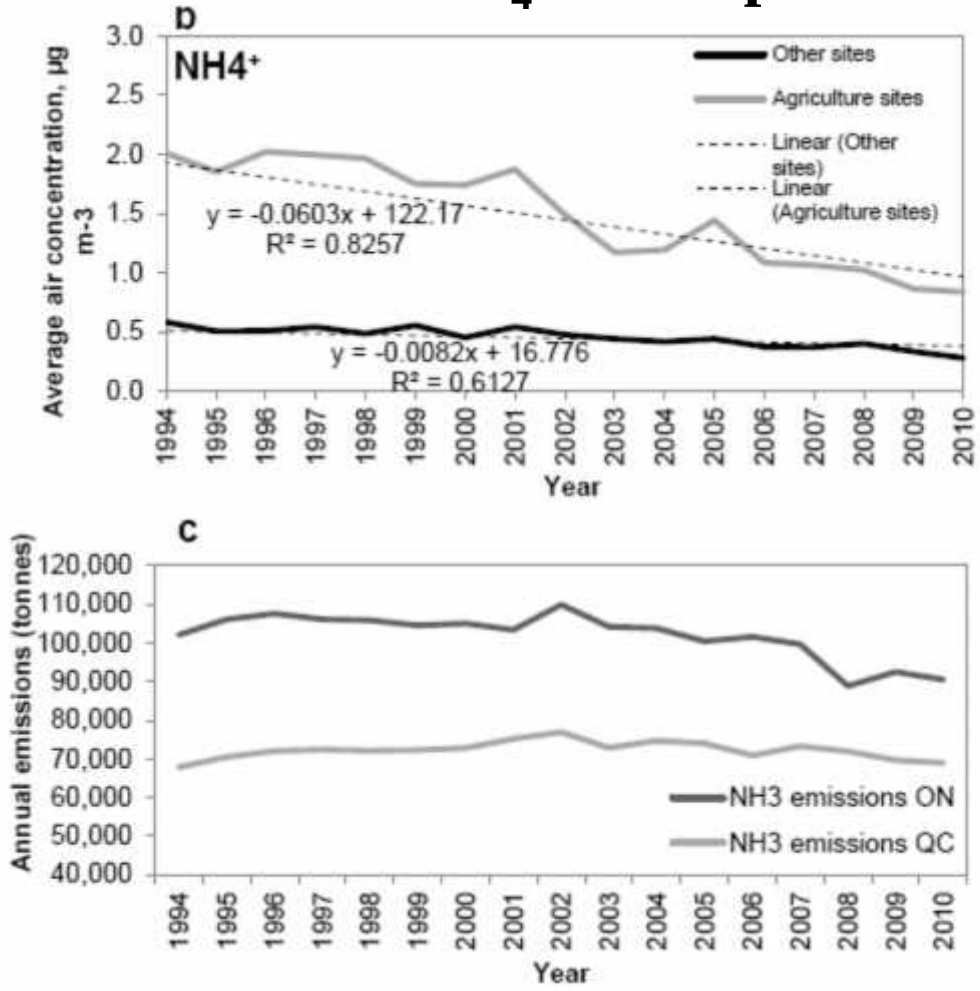
Quebec and eastern Canada (12 sites)

western/central Canada (10 sites)

southern Ontario (8 sites)

Results

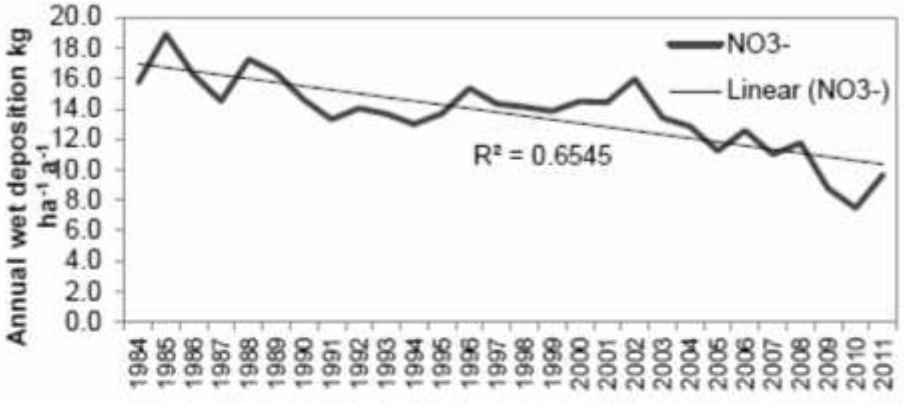
Widespread decline in atmospheric NH_4^+ in Canada, no significant trend in NH_4^+ wet deposition

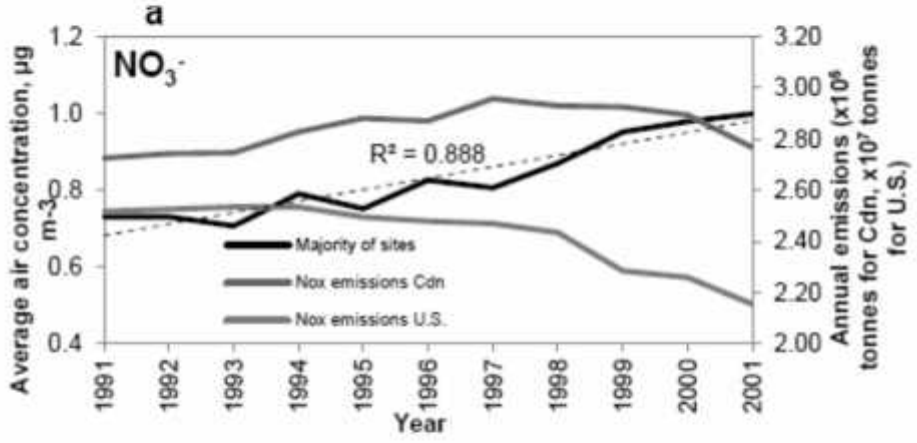


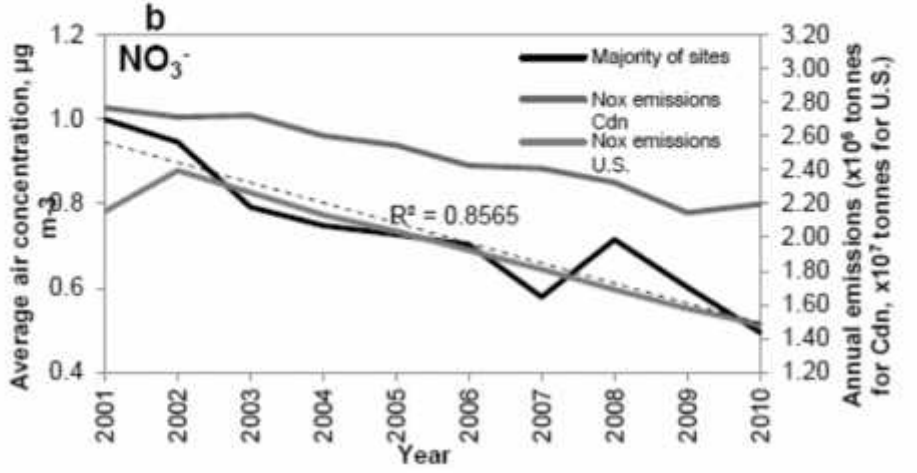
Results

- **Widespread decline in annual NO_3^- wet deposition, but different trends in air concentration before and after 2001**

- **The highest annual wet deposition rates for NH_4^+ and NO_3^- were found in southeastern Canada closest to industrial and urban areas**

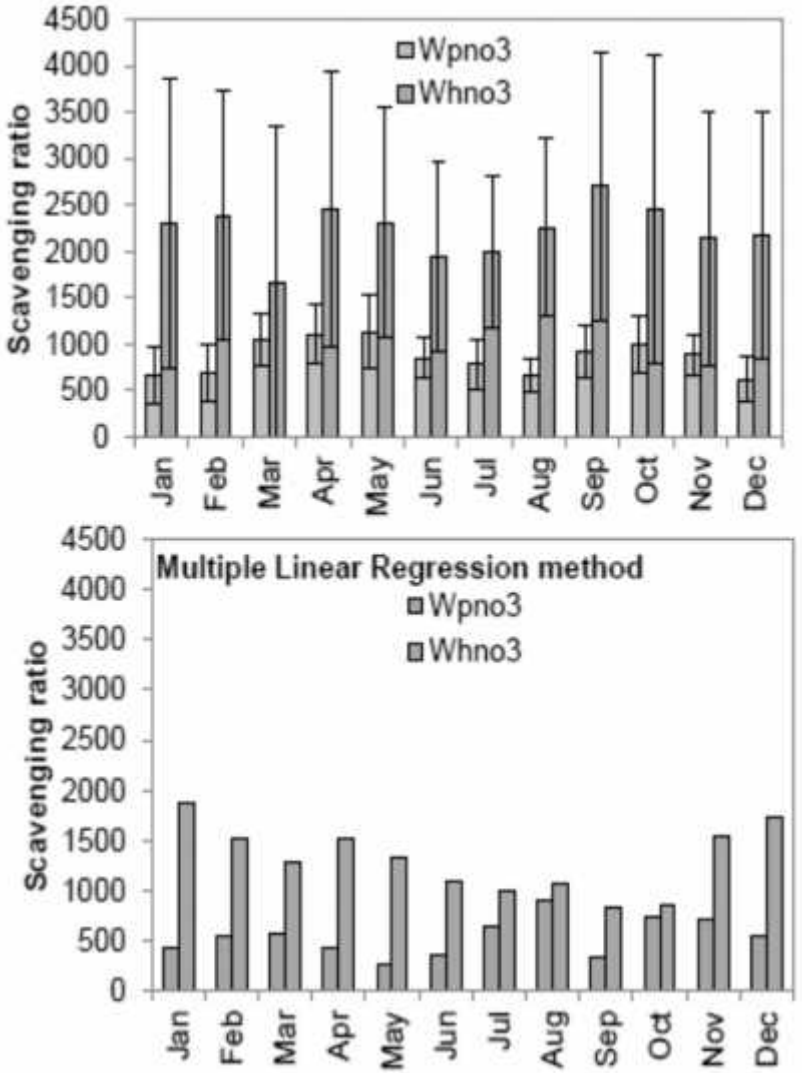






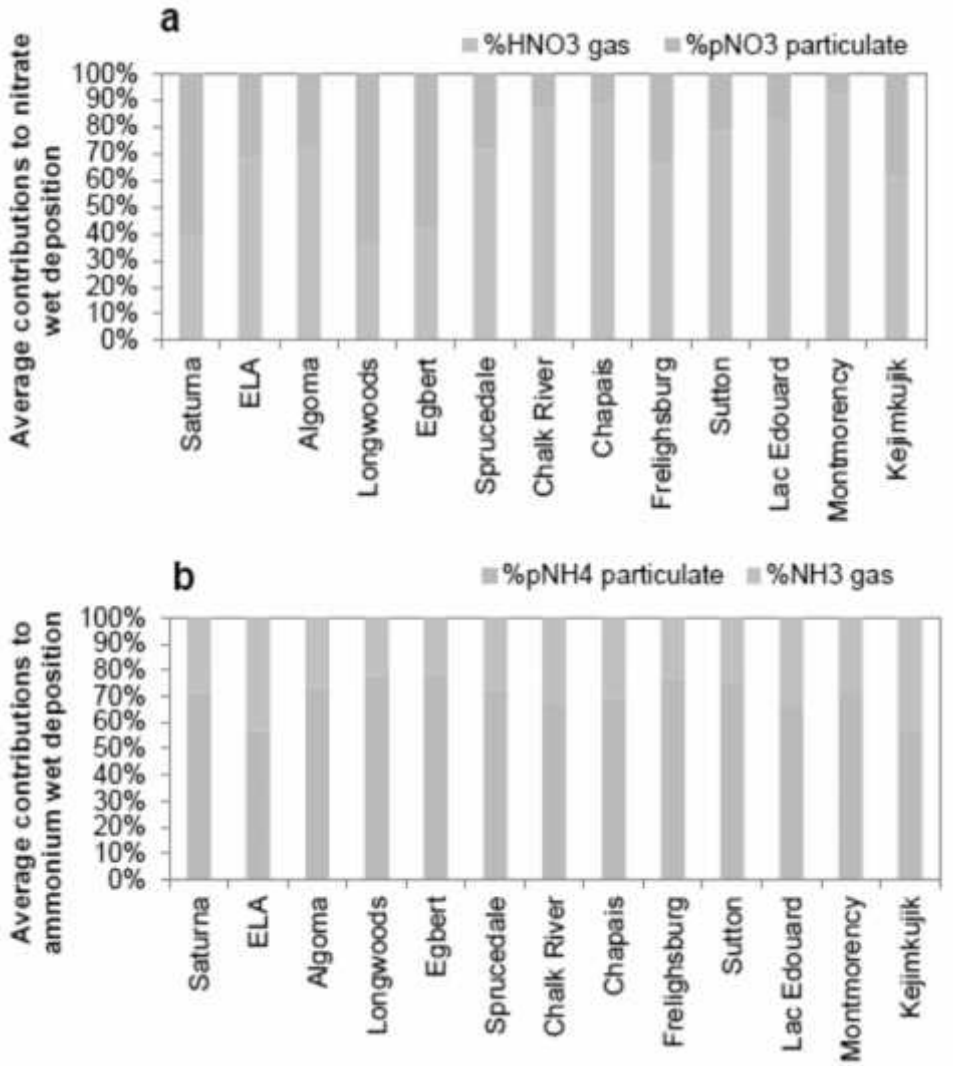
Results

- **Average scavenging ratio of HNO_3 was greater than pNO_3^-**
- **Most W_{pNO3} in literature are determined from total nitrate in precipitation and pNO_3^- in air, which overestimates W_{pNO3} (by a factor of 6 on average)**
- **When wet NH_3 scavenging is excluded, scavenging ratios of NH_4^+ can be overestimated by 4-48% (average: 22%).**



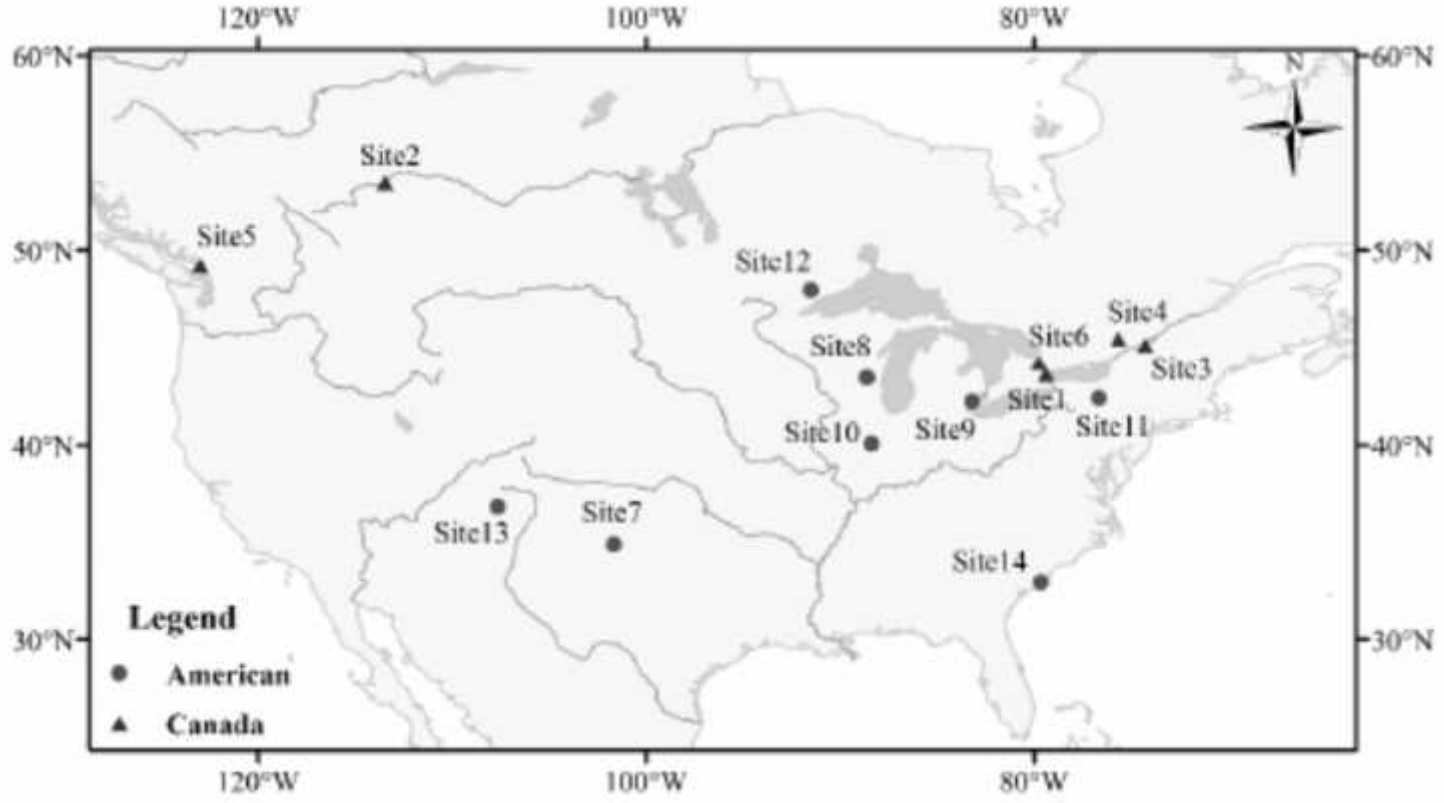
Results

- Average contribution to nitrate wet deposition: 28±23% from pNO_3^- and 72±23% from HNO_3
- Wet scavenging of pNO_3^- was higher at the sites closest to industrial and urban areas and at coastal sites
- Average contribution to ammonium wet deposition: 70±19% from pNH_4^+ and 30±19% from NH_3
- Particulate contributions were greater during cold months and lower during summer.



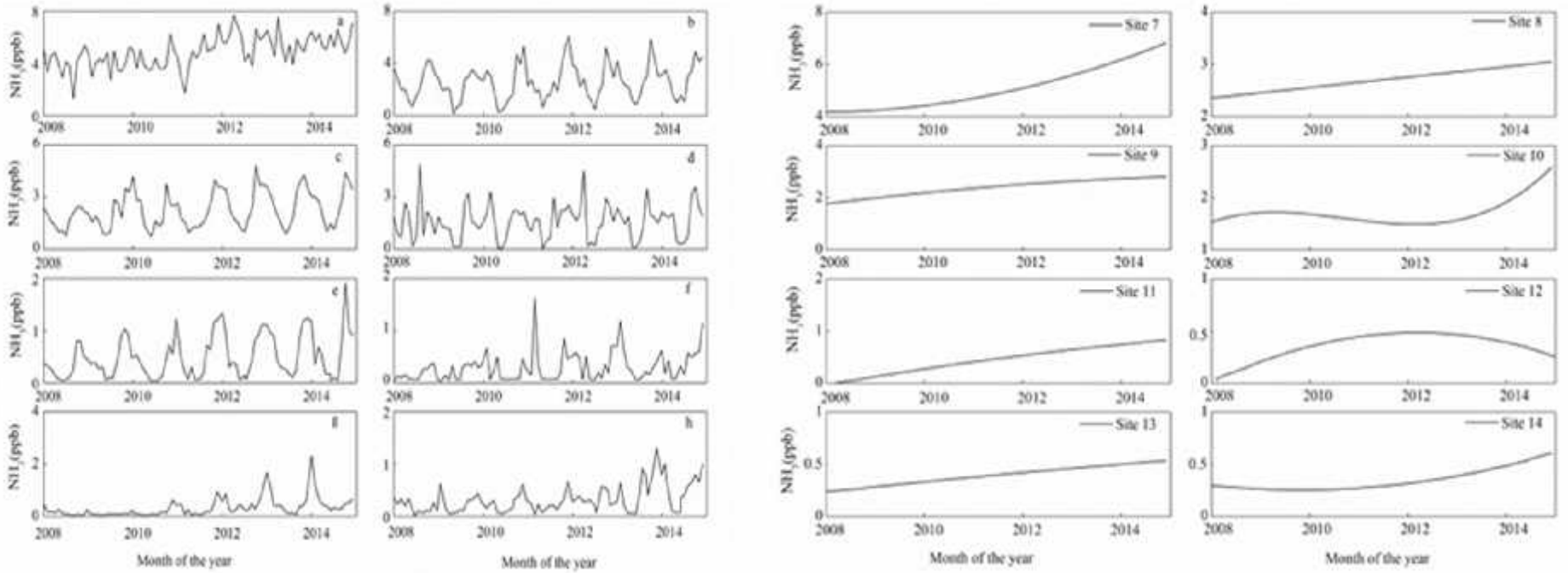
Map of sites with NH_3 data

Six Canadian sites and eight U.S. sites (> 7-year data)



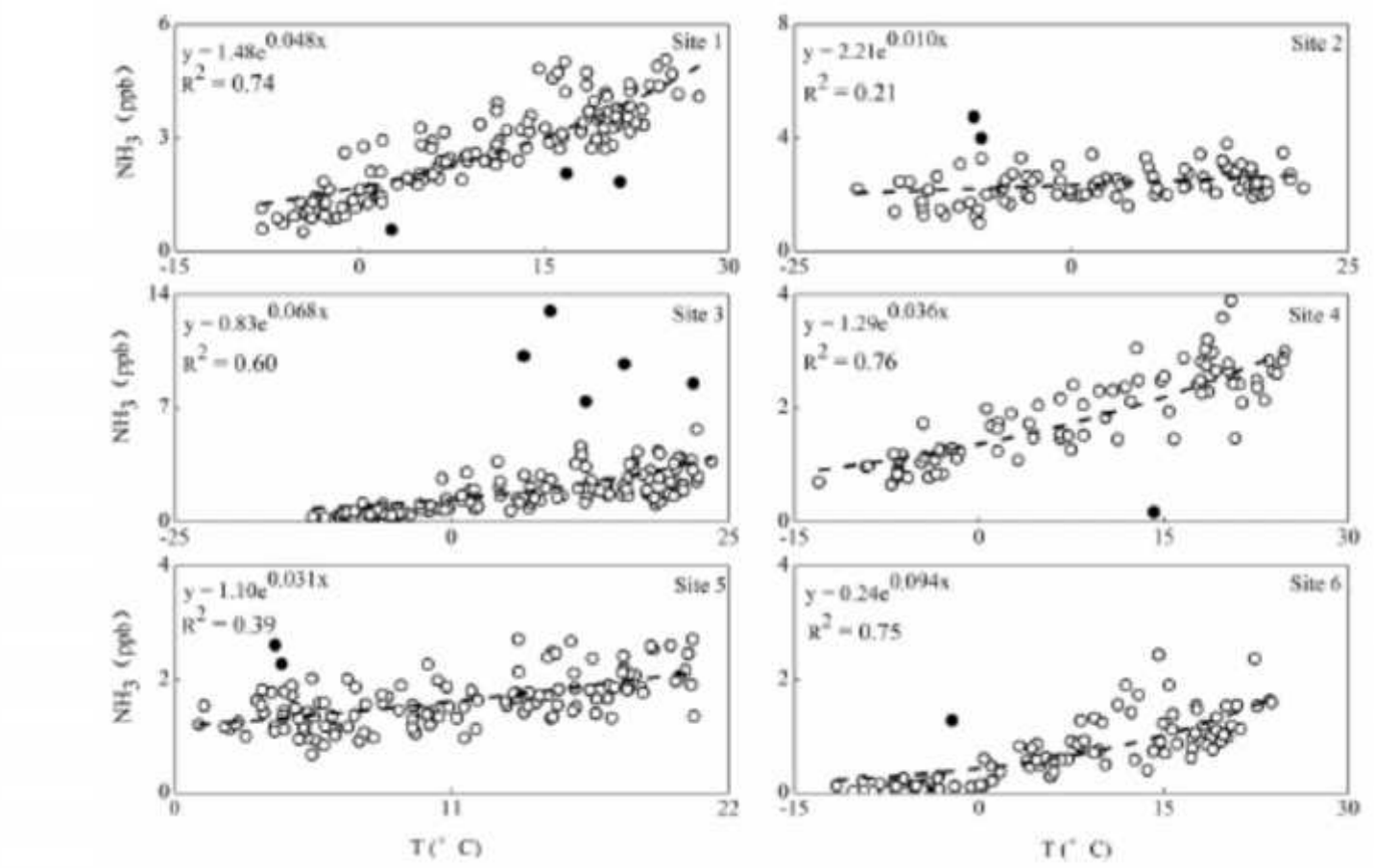
Results

Monthly average NH_3 and long-term trend extracted using Ensemble Empirical Mode Decomposition at the eight U.S. sites.



Results

Exponential correlations between atmospheric NH_3 and T at six Canadian sites



Results (second study)

- Moderate exponential correlations between atmospheric NH_3 and ambient T were found at nine sites - local biogenic emissions and/or NH_3/NH_4^+ partitioning were likely dominant factors at these sites.
- At the four Canadian sites, no decreasing trends in atmospheric NH_3 were found despite significant decreases in anthropogenic NH_3 emissions from main sectors in the last decade. The decreased NH_3 anthropogenic emission was compensated or overwhelmed by the increased biogenic emission and/or changes in NH_3/NH_4^+ partitioning. This was supported by pNH_4^+ data which exhibited a decreasing trend, likely caused by a combination of reduced SO_2 and NO_x emission and increased temperature.
- The M-K analysis showed an increasing trend in atmospheric NH_3 at seven out of the eight U.S. sites, which was also supported by the EEMD-extracted results.
- NH_3 increased by 20-50% from 2008 to 2015 at the three rural/agriculture sites and by 100%-200% at the four remote sites.