Environment and Climate Change Canada / Environnement et Changement climatique Canada

Application of NPRI data in source apportionment, decadal trend analysis and atmospheric deposition studies of monitored air pollutants

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1. Source apportionment of mercury

- Cheng I., Zhang L., Blanchard P., Graydon J.A., and St. Louis V.L., 2012. Source-receptor relationships for speciated atmospheric mercury at the remote Experimental Lakes Area, northwestern Ontario, Canada. Atmospheric Chemistry and Physics, 12, 1903-1922.
- Cheng I., Zhang L., Blanchard P., Dalziel J., Tordon R., Huang J., and Holsen T.M., 2013. Comparisons of mercury sources and atmospheric mercury processes between a coastal and inland site. Journal of Geophysical Research-Atmospheres, 118, 2434-2443.
- Cheng I., Zhang L., Blanchard P., Dalziel J., and Tordon R., 2013. Concentration-weighted trajectory approach to identifying potential sources of speciated atmospheric mercury at an urban coastal site in Nova Scotia, Canada. Atmospheric Chemistry and Physics, 13, 6031–6048.
- Cheng I., Zhang L., and Xu X., 2016. Impact of measurement uncertainties on receptor modeling of speciated atmospheric mercury. Scientific Reports, 6, 20676.
- Xu X., Liao Y., Cheng I., and Zhang L., 2017. Potential sources and processes affecting speciated atmospheric mercury at Kejimkujik National Park, Canada: comparison of receptor models and data treatment methods. Atmospheric Chemistry and Physics, 17, 1381-1400.
- Cheng I., Zhang L., Castro M., and Mao, H., 2017. Identifying changes in source regions impacting speciated atmospheric mercury at a rural site in the eastern United States. Journal of the Atmospheric Sciences, 74, 2937–2947.

Receptor-based source apportionment analysis approaches

Uses ambient air concentrations to identify and apportion sources; sources are unknown

- Multivariate methods:
  - Positive Matrix Factorization (PMF) model
  - Principal Components Analysis (PCA)
  - Cluster analysis
- Hybrid concentration-trajectory methods:
  - Potential Source Contribution Function (PSCF)
  - Concentration-Weighted Trajectory (CWT)
  - Other variations: gridded frequency distribution, residence time weighted concentration, simplified quantitative transport bias analysis

See a detailed review in Cheng et al. (ACP 2015)

NPRI data used in Hg source apportionment studies

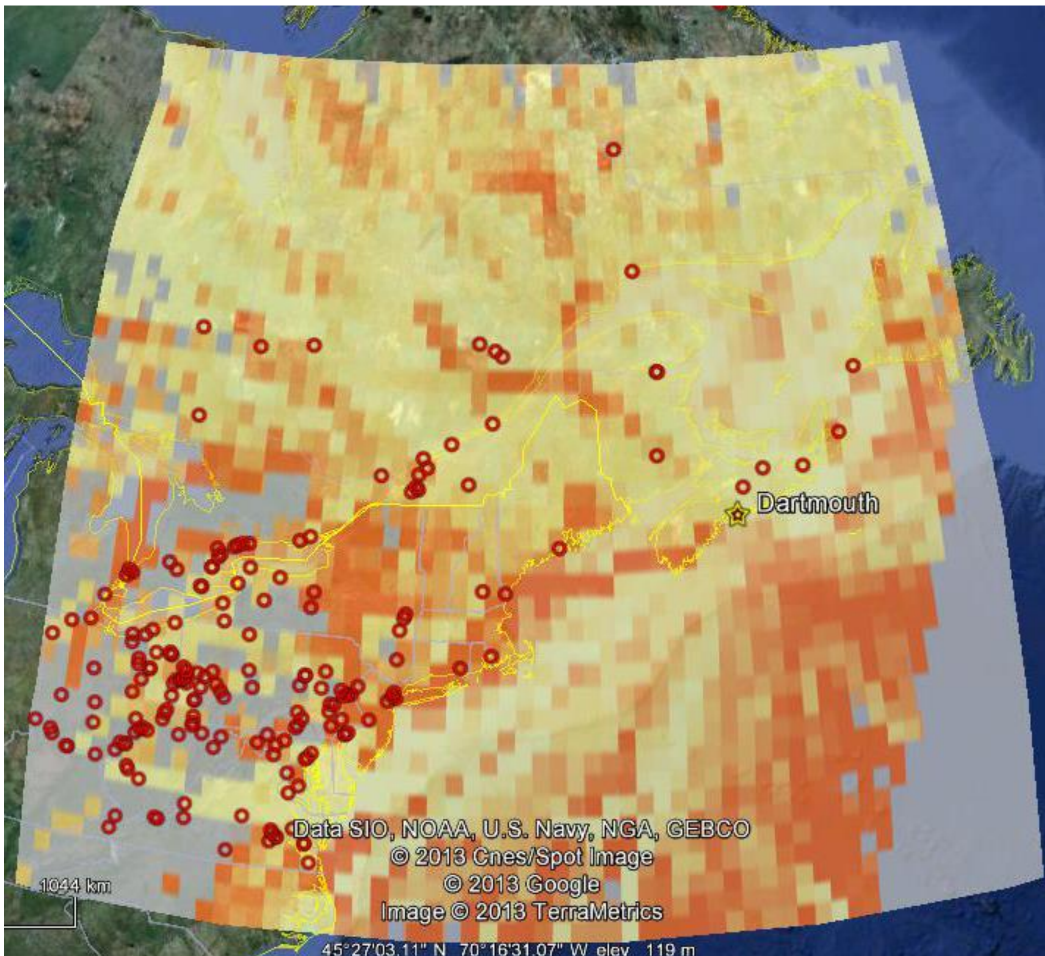
Mercury emissions from facilities (point sources) from specific provinces (Manitoba, Ontario, Quebec and Atlantic provinces) in 2005-2014, downloaded from the NPRI website.

Post processing:

- filtered point sources that emitted > 5 kg/year
- mercury emissions from point sources within each 0.5x0.5 degree grid cell were summed (in 2 studies), plotted the source locations and the receptor sites on a Google Earth map

Modelled Hg source regions using Concentration-Weighted Trajectory

weak regions (10th percentile CWT) **major source regions (90th percentile CWT)**



Source regions impacting atmospheric Hg at Dartmouth, NS

•CWT predicted source areas where there are Hg point sources (NPRI data)

• CWT predicted source areas from the Atlantic Ocean (emission of Hg)

Lessons learned

- Anthropogenic sources (NPRI data) only capture a fraction of the total atmospheric mercury emissions
- Emissions of mercury from the Atlantic Ocean is a significant source of mercury at the coastal sites
- Emissions from non-point sources not captured by NPRI, such as wildfires, biomass combustion, soil, vegetation and water surfaces, are also important sources of atmospheric mercury

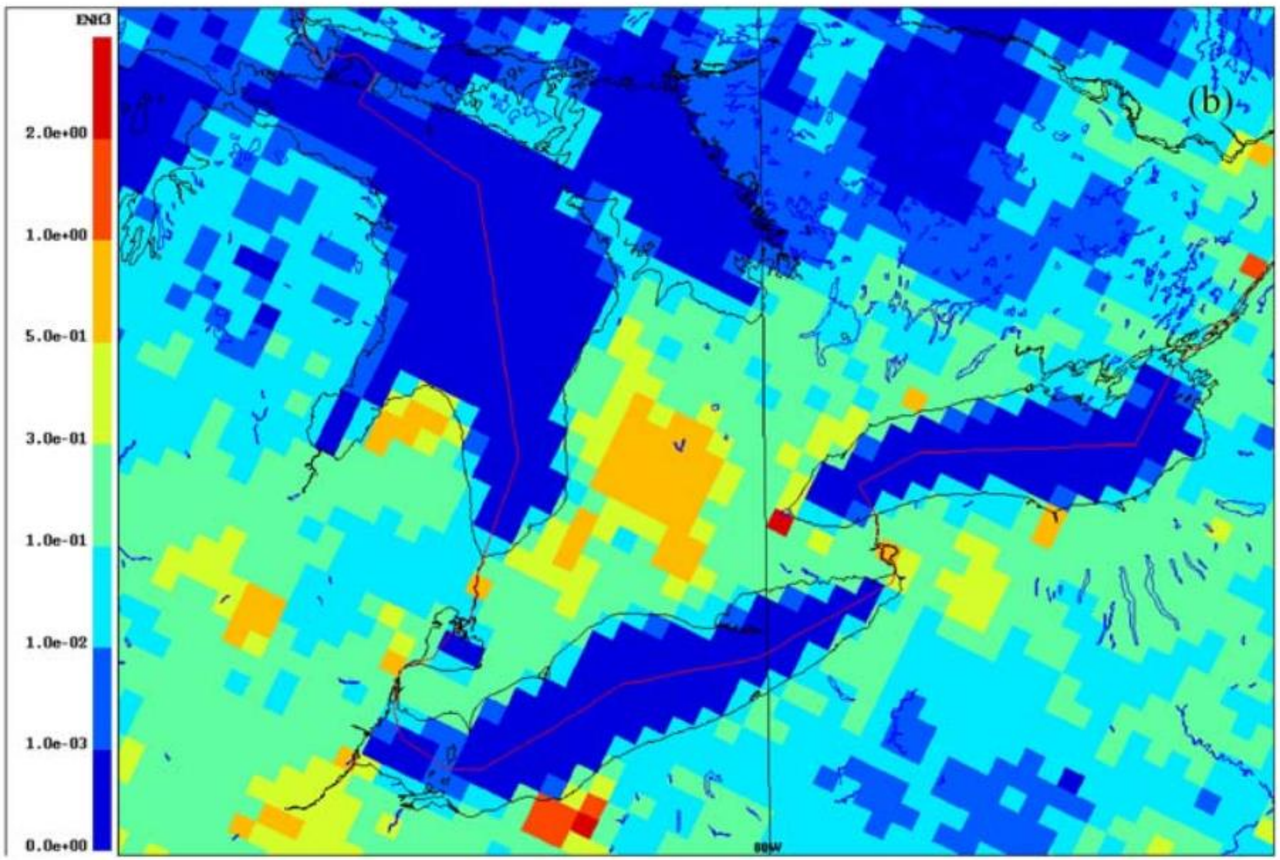
2. Acidifying pollutants – source apportionment

Receptor-based source apportionment of atmospheric ammonia

- **Yao X. and Zhang L., 2013. Analysis of passive-sampler monitored atmospheric ammonia at 74 sites across southern Ontario, Canada. Biogeosciences, 10, 7913-7925.**
- **Yao X., Hu Q., Zhang L., Evans G.J., Godri K.J., and Ng A.C., 2013. Is vehicular emission a significant contributor to ammonia in the urban atmosphere? Atmospheric Environment, 80, 499-506.**

Data from National Agri-Environmental Standards Initiative (NAESI): Gridded ammonia emission inventory at 15km x 15km (Ontario) or 1km x 1km (Toronto) resolution

NAESI NH3 emission inventory in the unit of ktonnes/year/grid element (15kmx15km)



2. Acidifying pollutants - Decadal trends analysis

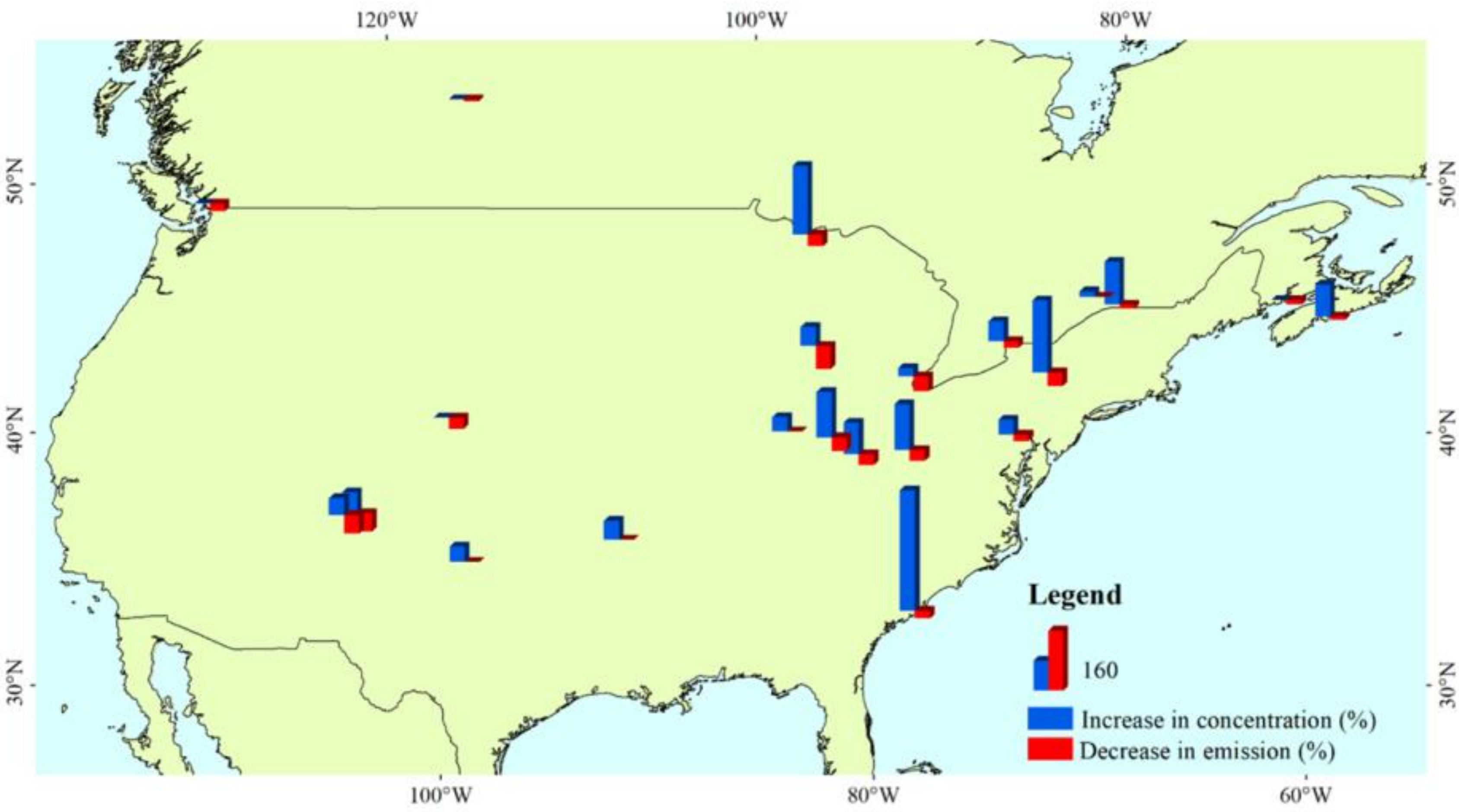
Trend analysis of air concentration and wet deposition of various pollutants

- Cheng I. and Zhang L., 2017. Long-term air concentrations, wet deposition, and scavenging ratios of inorganic ions, HNO3 and SO2 and assessment of aerosol and precipitation acidity at Canadian rural locations. Atmospheric Chemistry and Physics, 17, 4711-4730.
- Yao X. and Zhang L., 2019. Causes of large increases in atmospheric ammonia in the last decade across North America. ACS Omega, 4, 22133-22142.
- Yao X. and Zhang L., 2020. Decoding long-term trends in wet deposition of sulfate, nitrate, and ammonium after reducing perturbation from climate anomaly. Atmospheric Chemistry and Physics, 20, 721-733.
- Ongoing studies (PM2.5 speciation, OC/EC, dry and total deposition ......)

Emission data used in decadal trends analysis

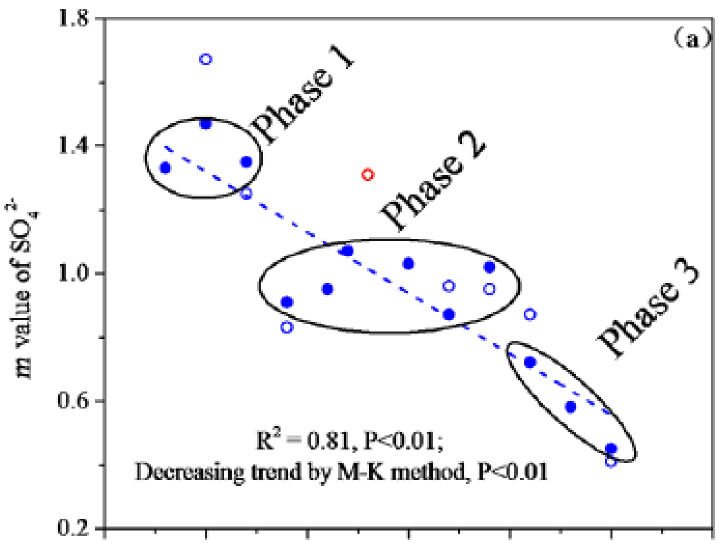
Air Pollutants Emissions Inventory (APEI) data: provincial level NOx, SO_2 and NH_3 emissions including point sources (NPRI data) as well as mobile sources, open sources and miscellaneous sources from 1991-2018

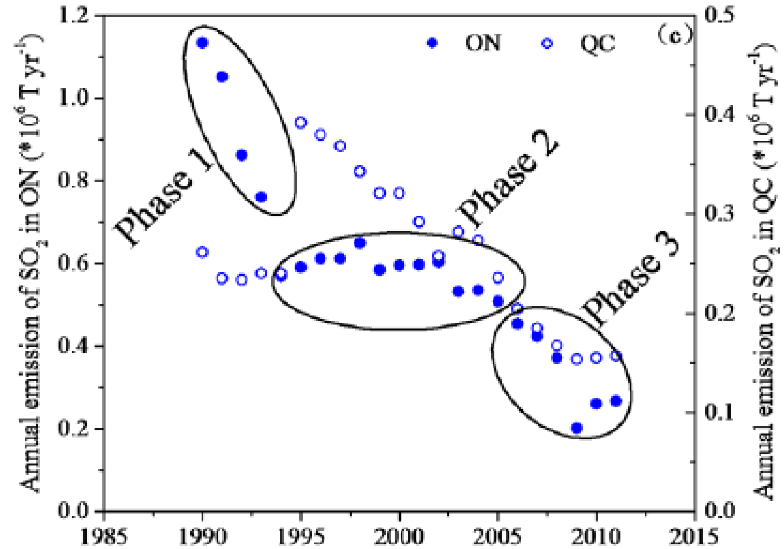
Percentage increase in annual NH3 concentration at each sampling site and percentage decrease in annual NH3 emission in corresponding state or province

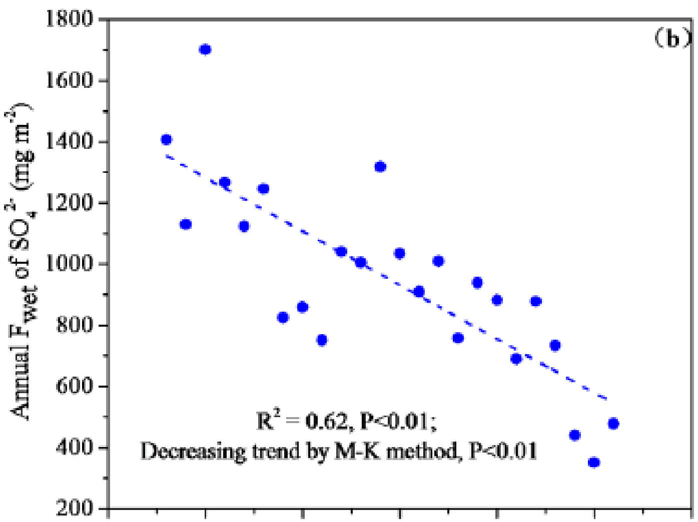


Analysis approaches in trend analysis

- Mann-Kendall
- Ensemble Empirical Mode Decomposition
- Linear Regression
- Piecewise Linear Regression
- New method developed for data preprocessing for excluding climate anomalies

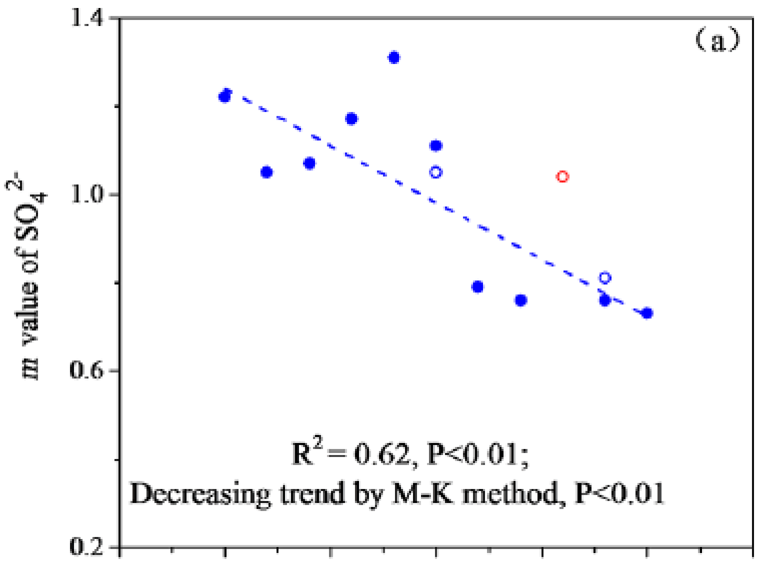


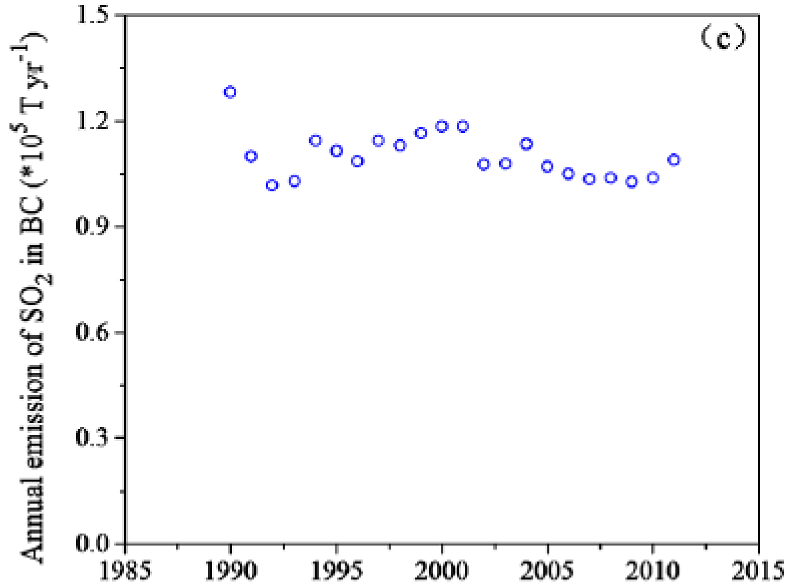


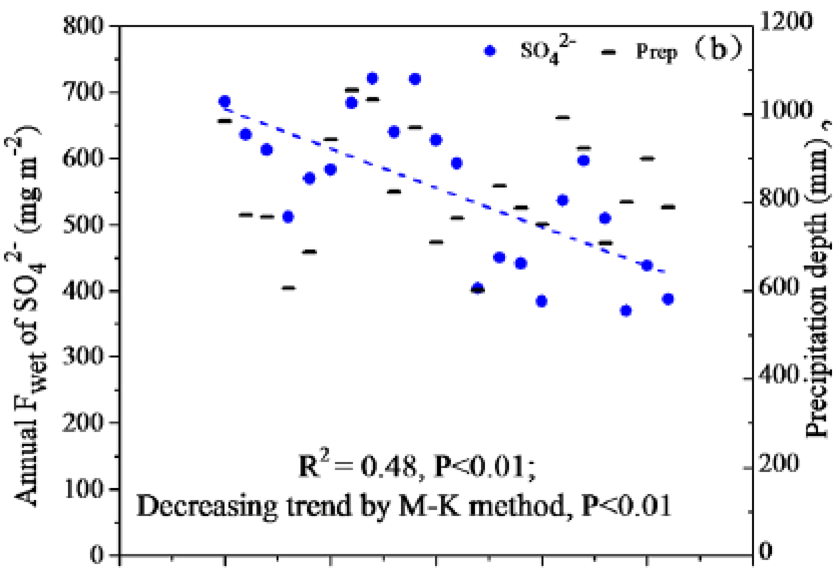


- The new approach allows for statistically identifying inflection points on decreasing trends in the wet deposition fluxes of SO_4^{2-} and NO_3^- in northern Ontario and Québec
- The inflection points match well with the three-phase mitigation of SO_2 emissions and two-phase mitigation of NOx emissions in Ontario
- Better results if combining sites in a region

Site: Chapais, QC







Decadal climate anomalies dominated the decreasing trends in the wet deposition fluxes of SO_4^{2-} and NO_3^- at a western coastal site.

Site: Saturna, BC

Lessons learned

In some studies APEI data was more suitable for the analysis of air quality trends because it considered all types of emissions including facility emissions (NPRI data) as well as emissions from mobile sources, open sources and miscellaneous sources

3. PACs and trace elements

- Goal – Mapping atmospheric deposition in the Canadian oil sands region
- Approach – Adjusting modelled gridded concentrations with limited measurements
- Model input data needed: emission inventory in the region

- **Qiu X., Cheng I., Yang F., Horb E., Zhang L., and Harner T., 2018. Emissions databases for polycyclic aromatic compounds in the Canadian Athabasca oil sands region – development using current knowledge and evaluation with passive sampling and air dispersion modelling data. Atmospheric Chemistry and Physics, 18, 3457-3467.**
- **Qiu et al, 2020. Emission databases of trace elements in the Canadian Athabasca oil sands region. In preparation.**

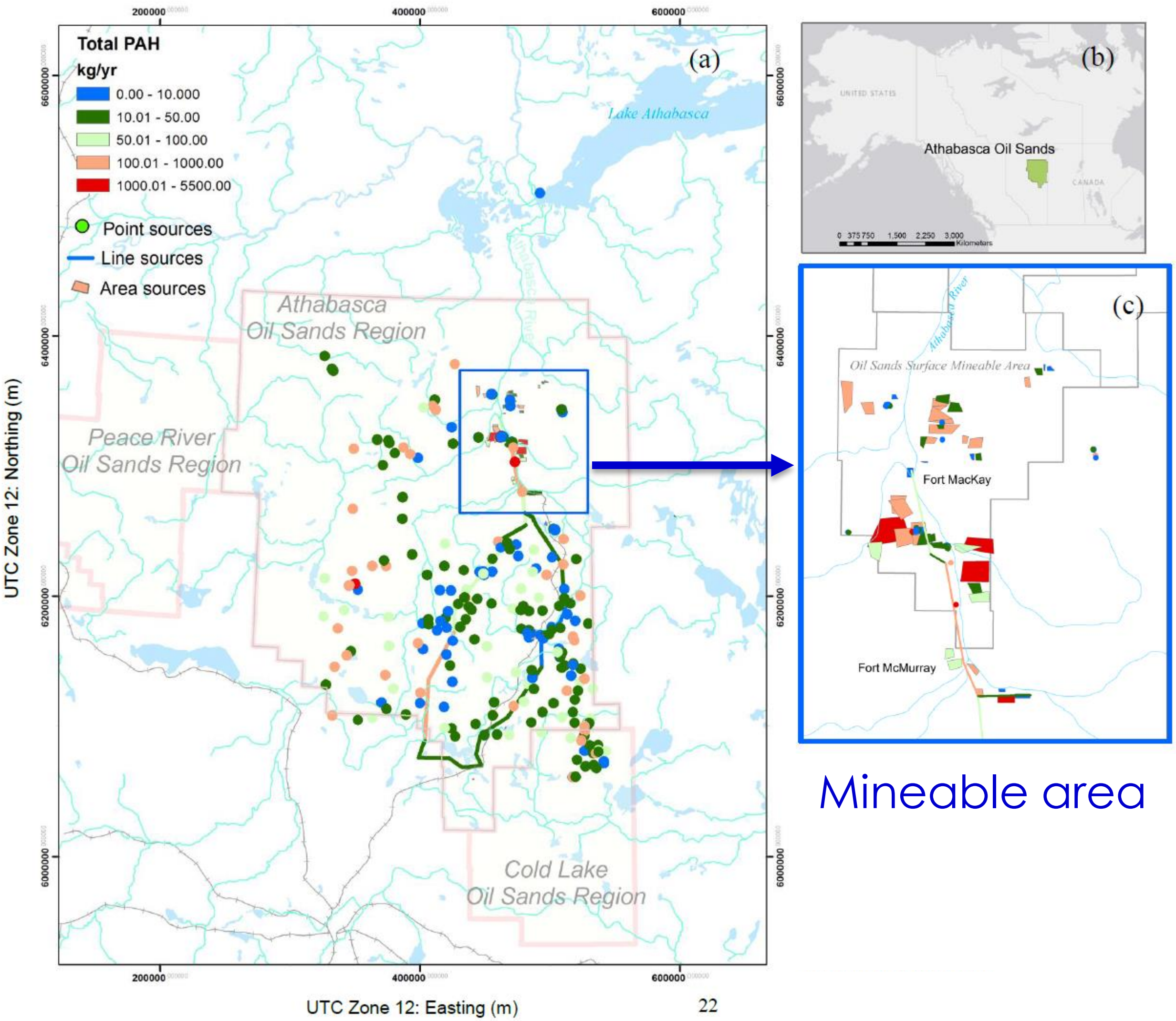
NPRI data used in atmospheric deposition studies

- NPRI facility based emissions have been used to validate and correct APEI point source emissions inventories: facility IDs, names, facility total emissions, major point sources emissions, locations, year of in operations and decommissions, etc.
- NPRI data is used to provide some VOC species and metals emissions which were not provided in APEI emissions inventory.

Approach of generating PACs emission inventory

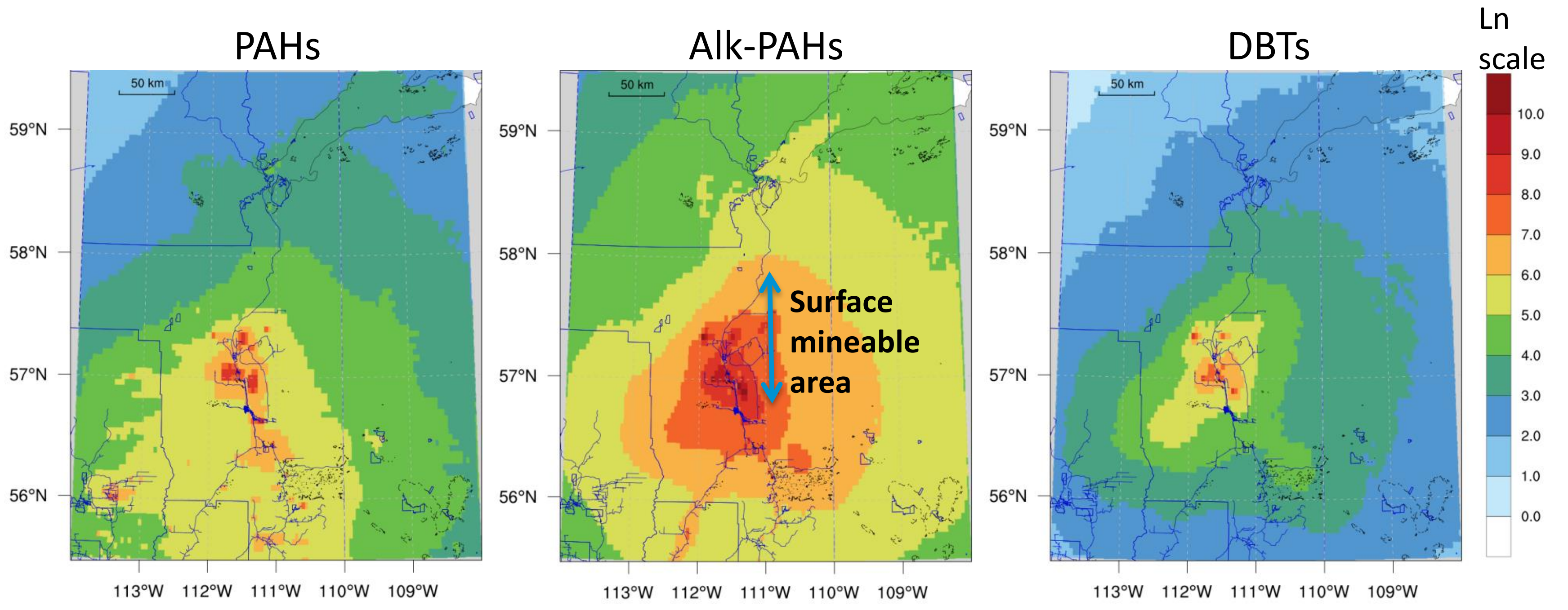
- Develop PAC emissions speciation profiles for the oil sands region by using
  - NPRI facility VOCs emissions
  - CEMA facility source information
  - U.S. EPA Speciate 4.0 program for PACs emission factors
  - SMOKE speciation profiles with SCC cross-reference
- Generate emission inventory by source types to estimate PACs emissions in the oilsands area
- Cross-check with existing "Environmental Impact Assessment" reported data and literature

PACs emission inventory



Mineable area

Results - Annual dry deposition



2.7-7,900 µg m^{-2} yr^{-1}

17-56,000 µg m^{-2} yr^{-1}

1.5-7,800 µg m^{-2} yr^{-1}

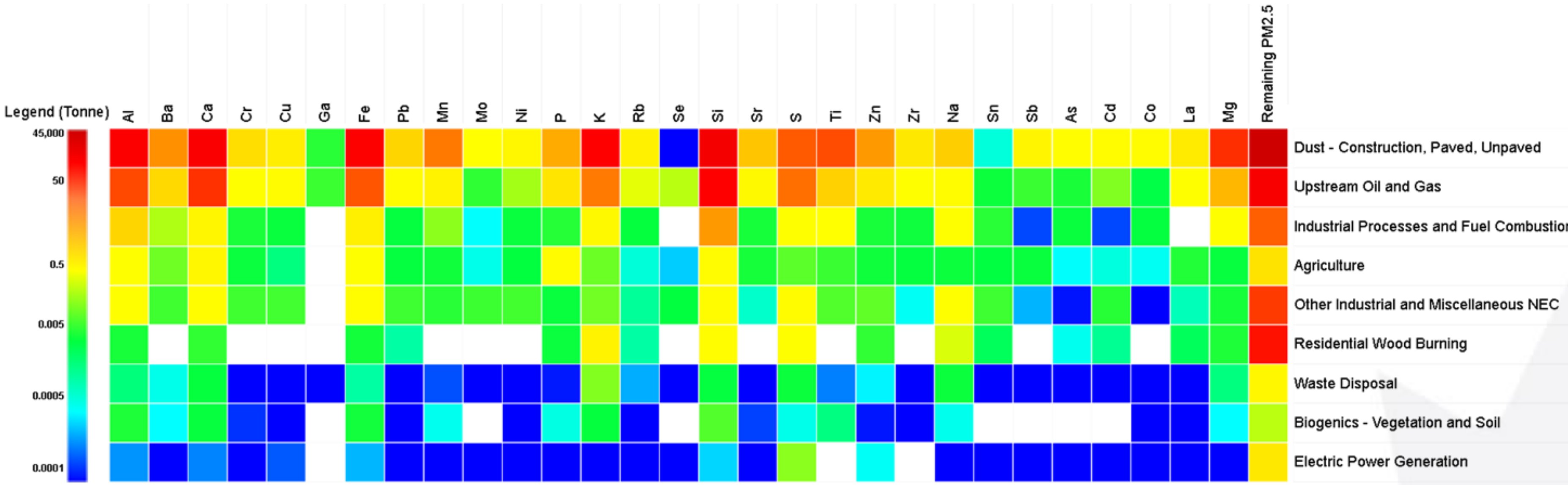
- **Alk-PAHs, PAHs, and DBTs contributed 74%, 19% and 7% to average annual dry dep in 2011**
- **Dry dep hotspot located over the surface mineable area**
- **Higher fluxes observed over a larger area for alk-PAHs, extending beyond the surface mineable area**

Cheng et al. (EST 2018)

Approach of generating trace elements emission inventory

- Develop trace metal emissions speciation profiles for the oilsands region by using
  - A combination of JOSM and NPRI facility $PM_{2.5}$ and PM_{10} emissions
  - U.S. EPA Speciate 4.0 program for trace metal emission factors
  - SMOKE speciation profiles with SCC cross-reference
- Generate emission inventory by source types to estimate trace metal emissions in the oils ands area
- Cross-check with existing NPRI reported data and literature

Trace elements emissions in $PM_{2.5}$ breakdown by sectors



For trace metal emissions derived from $PM_{2.5}$ the top three target trace metal emission categories are **Dust – Construction, Paved, Unpaved Roads ($5.43x10^3$ tonne/year), Upper Oil and Gas ($1.74x10^3$ tonne/year) and Industrial Processes and Fuel Combustion ($7.48x10^1$ tonne/year).**

Lessons learned

- NPRI data provides reliable, facility annually reported CAC and toxic emissions.
- Other resources such as APEI data or provincial/municipal/organization's emissions data can be used to complement the NPRI data.

Thank you!