Identification of the Origins of Elevated Atmospheric Mercury Episodes Using a Lagrangian Modelling System

D. Wen, J. Lin, F. Meng and J.J. Sloan

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University of Waterloo

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Approach

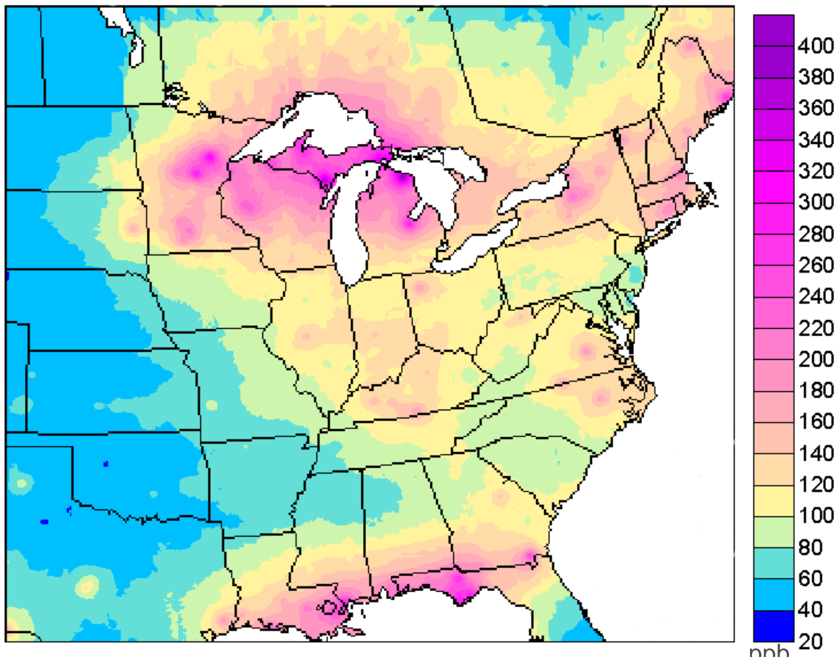
- Previously developed a model for the analysis of atmospheric mercury transport in North Eastern North America[1]:
  - ➢ Nested Eulerian (Bullock CMAQ-Hg) model. Domains:
    - ✓ North America
    - ✓ Great Lakes
    - ✓ Southern Ontario.
- Model application[2] gives "natural" Hg emission from soil, water and vegetation; adds this to anthropogenic

[1] Gbor *et al.*, " Improved Model for Mercury Emission, Transport and Deposition", Atmospheric Environment, **40**, 973-983 (2005).

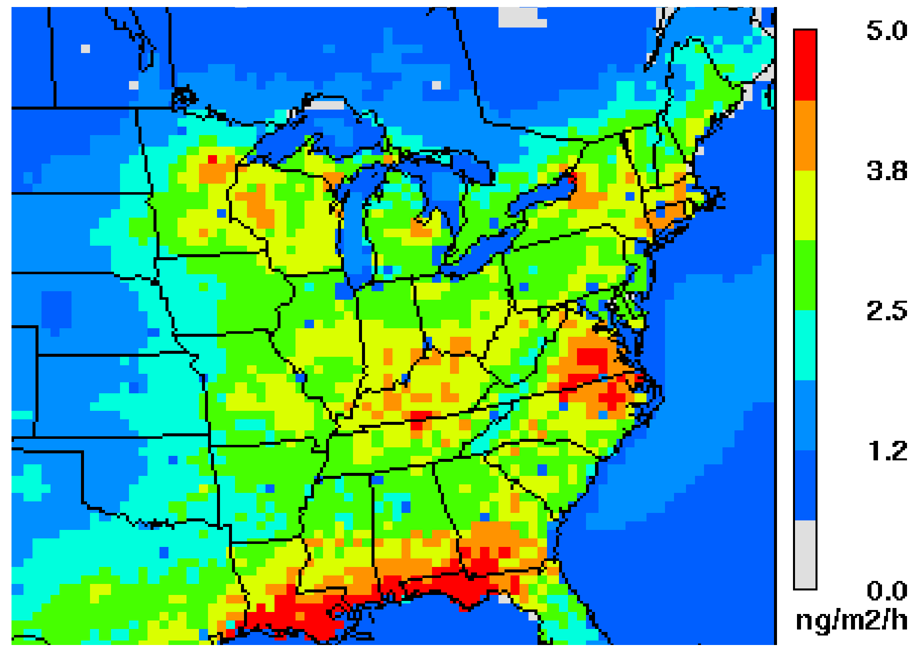
[2] Gbor *et* al. "Modeling of mercury emission, transport and deposition in North America", *Atmospheric Environment* **41** 1135–1149 (2007);

Natural Mercury Emissions

- Natural Hg includes mineral and historical anthropogenic deposition.
    - ➢ Natural emission are based on measured soil and water mercury levels



Soil mercury concentration (ppb)



Natural mercury emission (ng/m^2/h)

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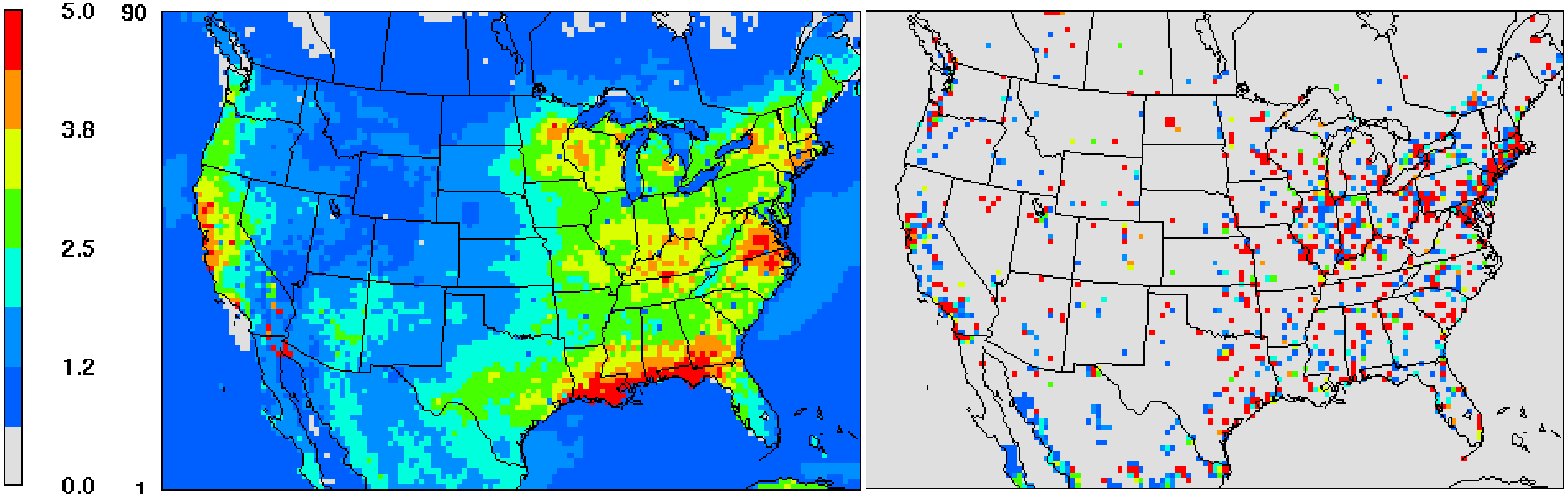
Natural and Anthropogenic Emissions

- Average mercury emission fluxes (ng/m²/h)

1 Jan. to 30 Dec., 2002



Natural Anthropogenic

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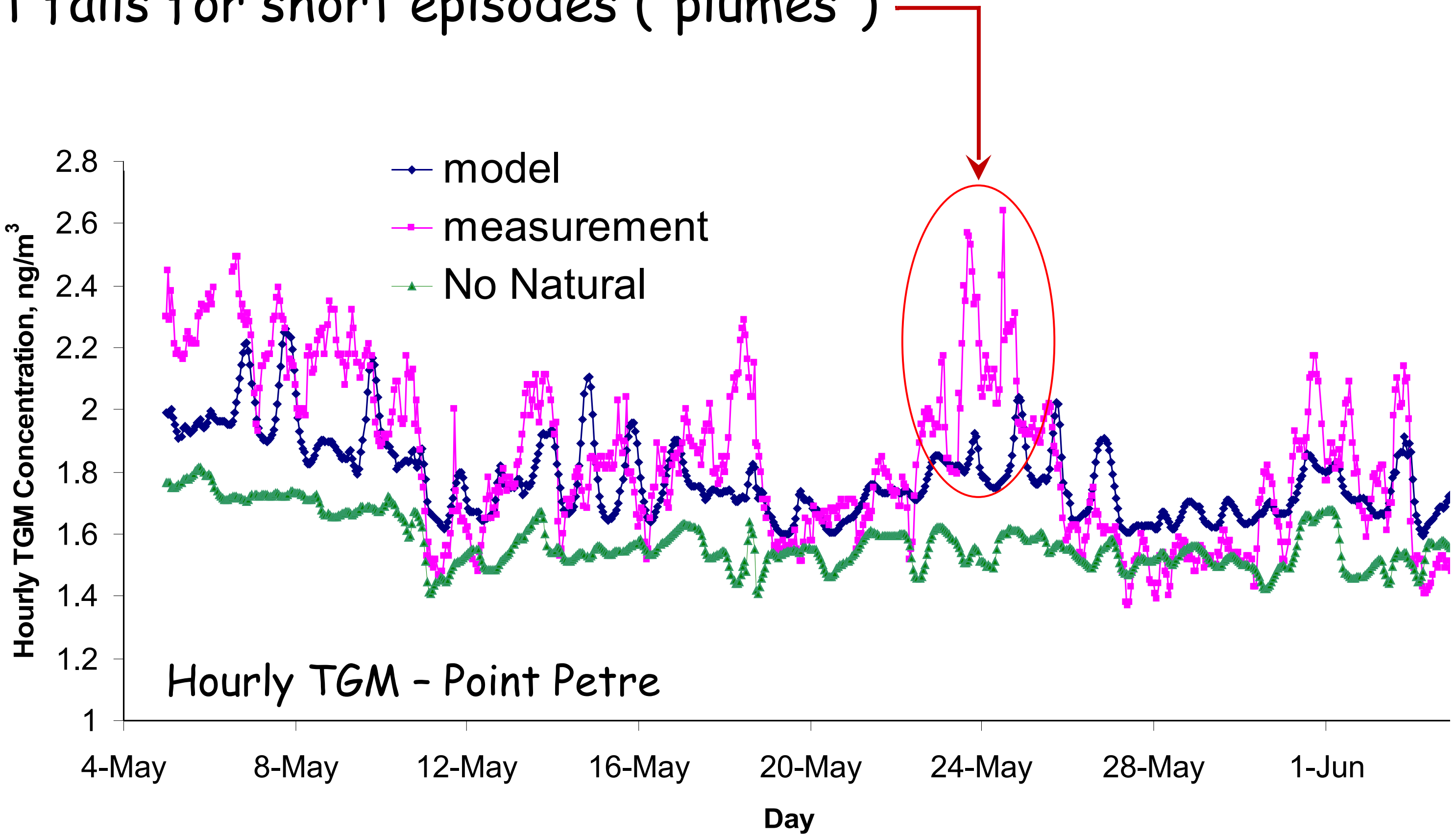
CMAQ-Hg CTM Comparison with Measurement

- Eulerian CTM (including natural emission) does well in most cases, but fails for short episodes ("plumes")



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Analysis of Model – Measurement Differences

oal: Identify sources of episodic differences

pproach:

1) Systematically compare the time series of CMAC-Hg CTM predictions with measurements to identify episodes that are not well described by the CTM
2) Examine these episodes using Lagrangian model

- Same meteorology and same emissions are used with both models. This saves computational time and effort.

dvantage: Eulerian CTM can be run at low resolution; Lagrangian used to analyse short-term differences

Lagrangian Modelling to Identify Plumes

- Why does Eulerian CTM differ from measurement?
  - Differences with short term measurements due to spatial averaging at (low) 36 km resolution.

- Examine differences with: *Stochastic Time-Inverted Lagrangian Transport* (STILT) Model*
  - simulates upstream influences on a receptor by following the evolution of a particle ensemble backward in time
  - Interpolates wind fields to the location of each particle
  - Simulates turbulent motions in PBL by a Markov chain process based on observed meteorological parameters.

*Lin, J.C., *et al.*, *J.G.R.* **108**, 4493 (2003)

Hg Transport with STILT

- Tracer emitted at any (surface) location is divided <u>equally</u> among particles originating there at altitudes below the turbulent mixing height.
  - Particle density at a specified receptor directly yields the tracer concentration at the receptor location.
  - Backward transport of particles from a receptor thus maps out locations and <u>strengths</u> of sources contributing to that receptor.
- Source strength: given by surface flux, particle density and residence time.
- Wet and dry deposition of the tracer are included

Source-Receptor Connection: the Footprint

- Source footprint: the concentration change at the receptor for a unit surface flux at the footprint location that persists for a specified time interval:

$$f(x_r, t_r \mid x_i, y_j, t_m) = \frac{m_{air}}{h\bar{\rho}(x_i, y_j, t_m)} \frac{1}{N_{tot}} \sum_{p=1}^{N_{tot}} \Delta t_{p,i,j,k}$$

  - $\bar{\rho}(x_i, y_j, t_m)$: local density of particles at the source (x_i, y_j, t_m)
  - $\Delta C_{m,i,j}(x_r, t_r) = f(x_r, t_r \mid x_i, y_j, t_m) F(x_i, y_j, t_m)$: Change in receptor concentration due to ensemble of air parcels remaining at source having emission flux: $F(x_i, y_j, t_m)$ for a time $\Delta t_{p,i,j,k}$

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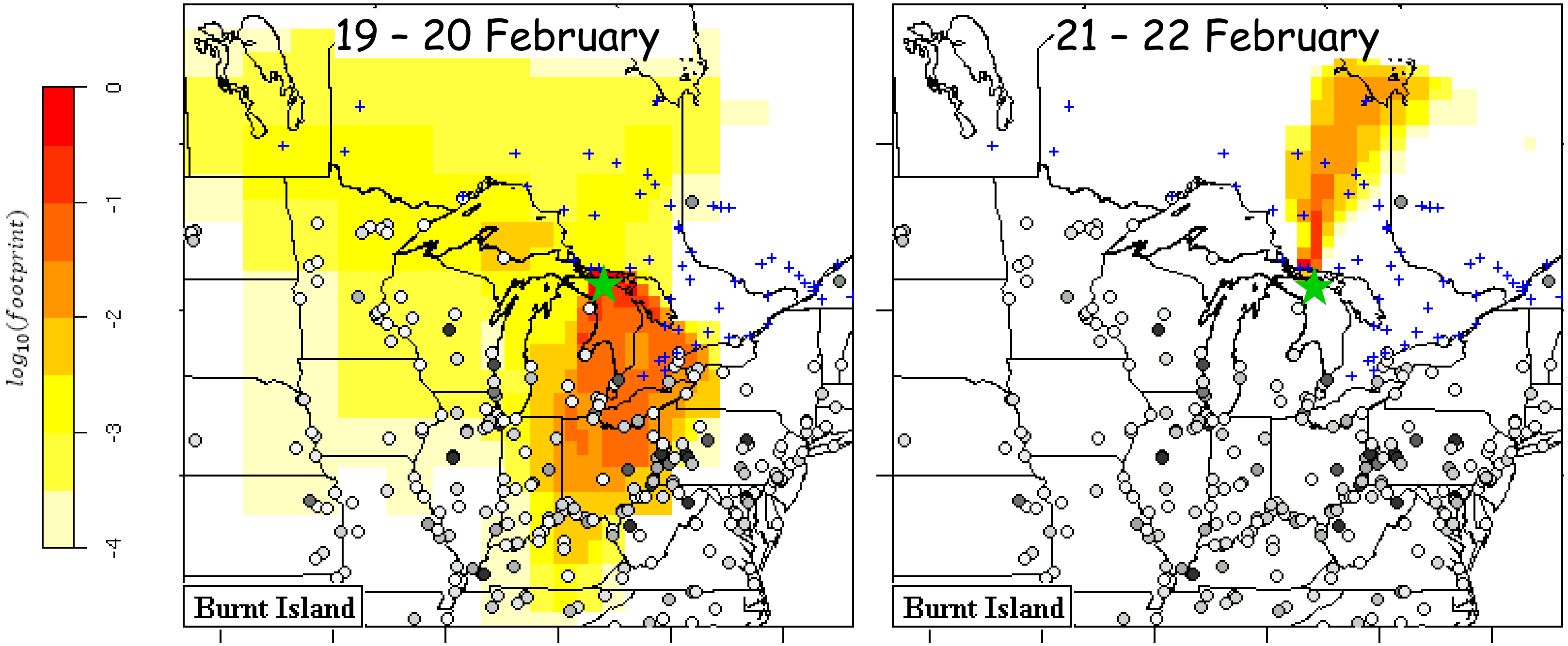
Example: Source Footprints for Hg at Burnt Island Receptor (February 2002)



- Points: Locations of Hg point sources
- Colour: footprint ($\log_{10}$ [ppm/μmole/m^2/s])

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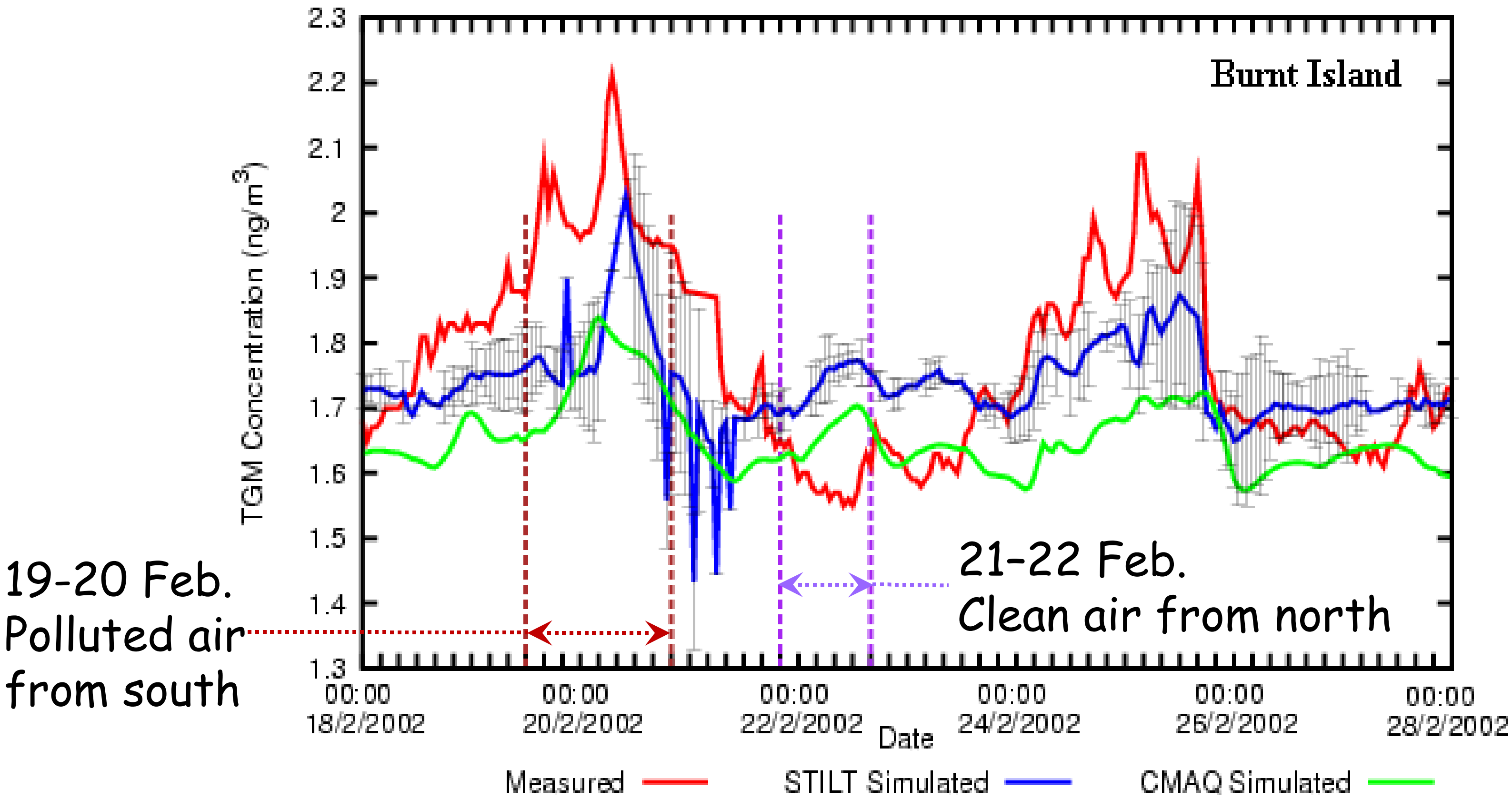
Hg Concentrations at Burnt Island (February 2002)



- STILT reproduces episodes better than (low resolution) regional model

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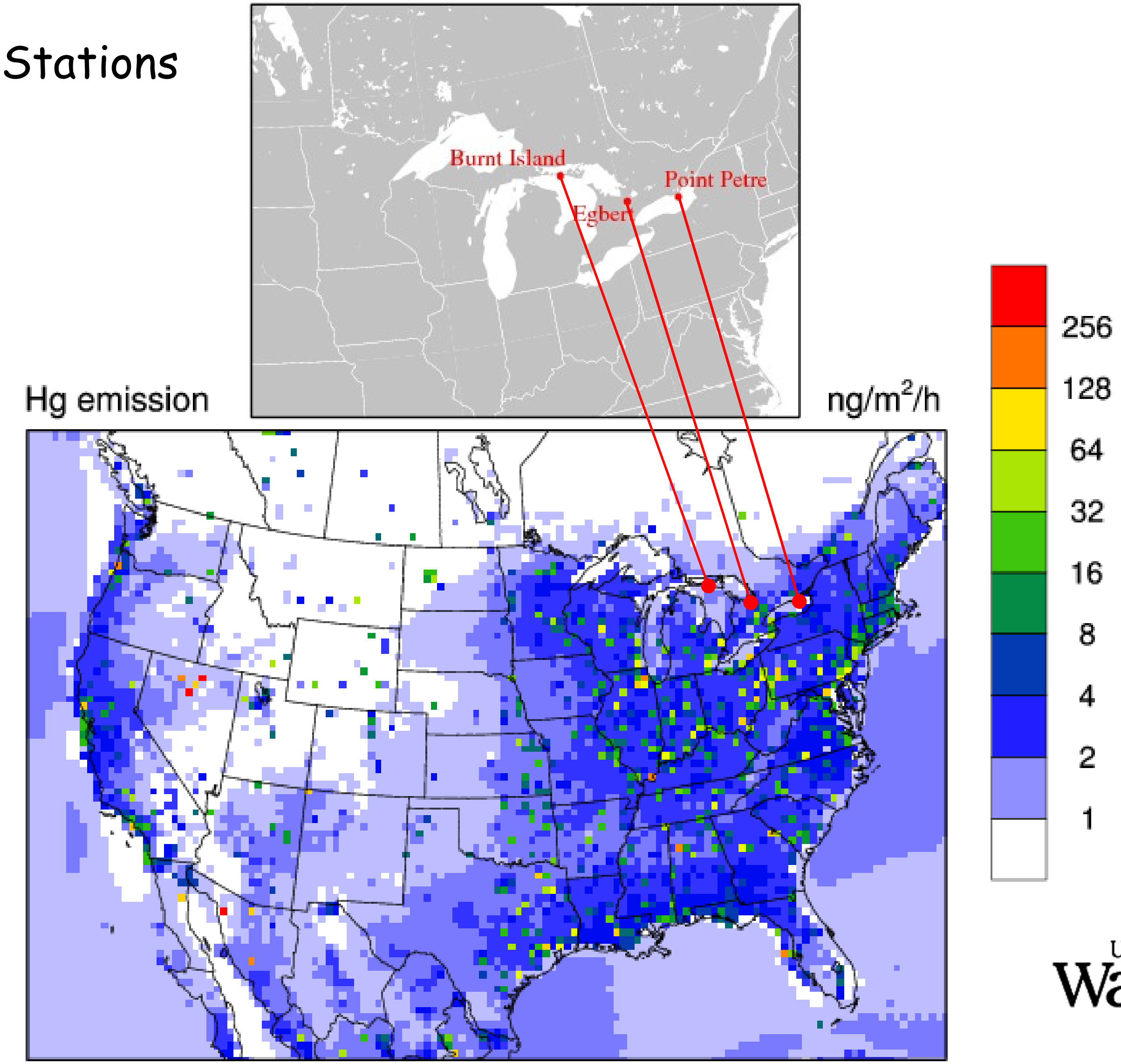
This Study: Total Hg Emission and Measurements

Measurement Stations

- Burnt Island
- Egbert
- Point Petre



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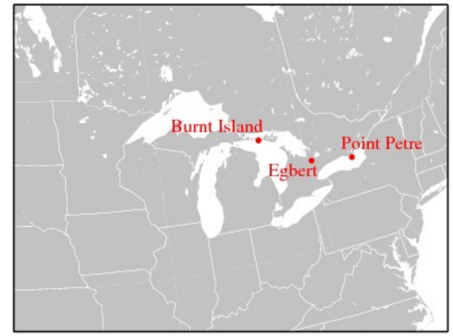
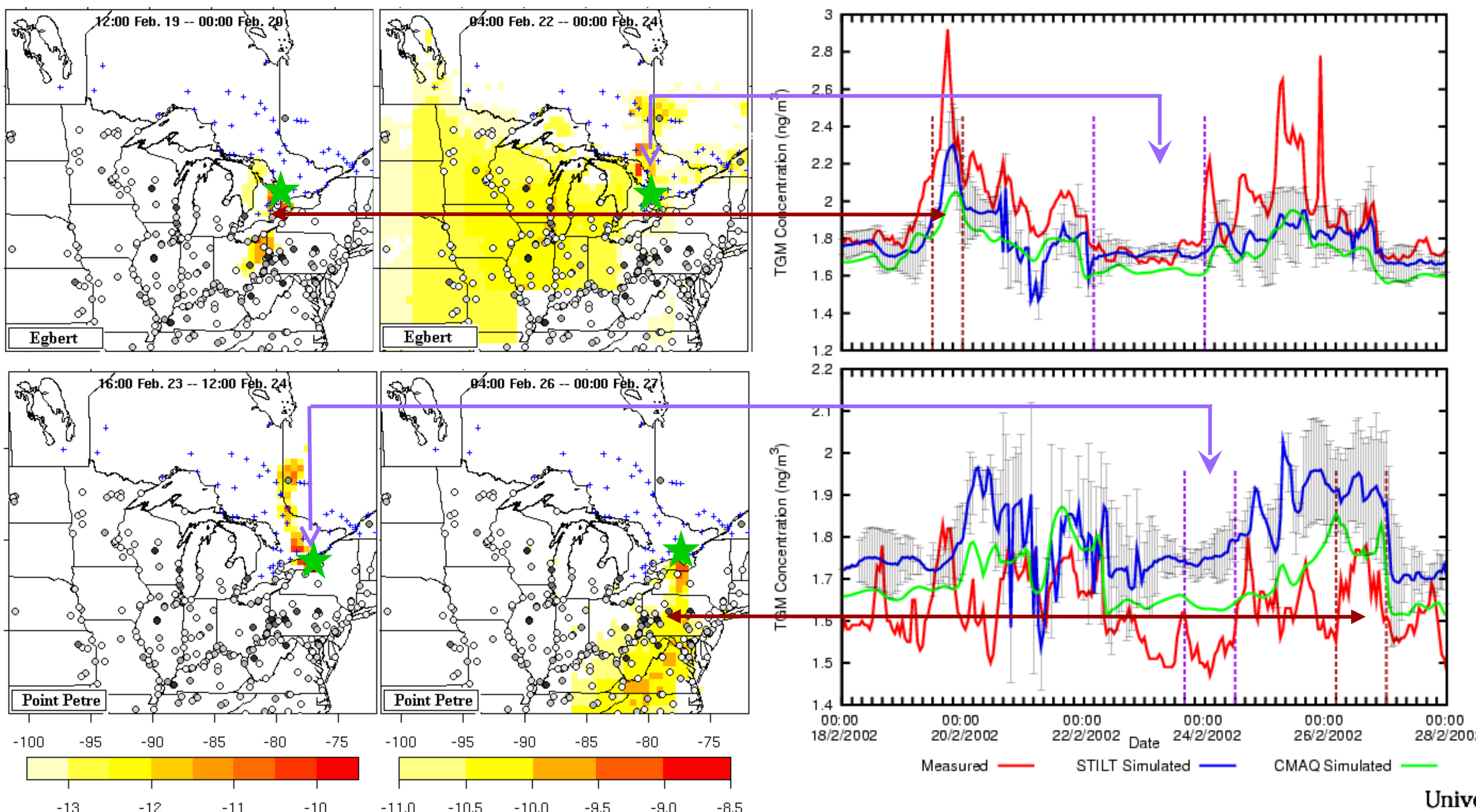
Monitoring Sites Egbert and Point Petre (February 2002)





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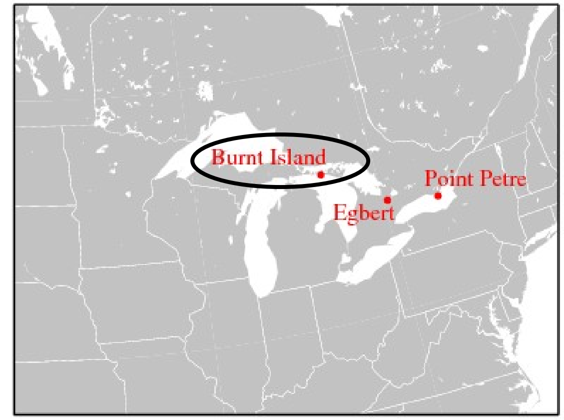
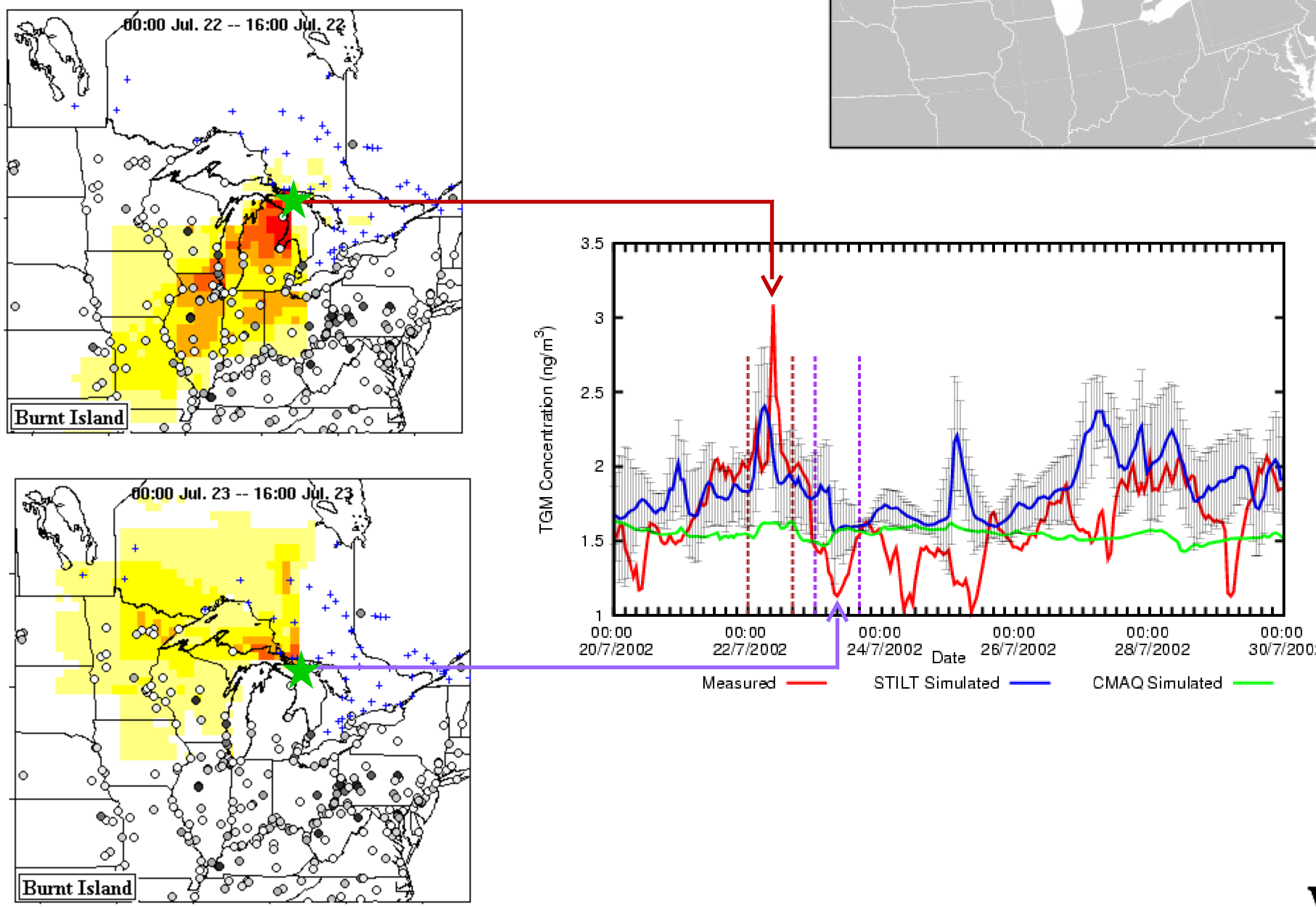
Burnt Island, July 2002





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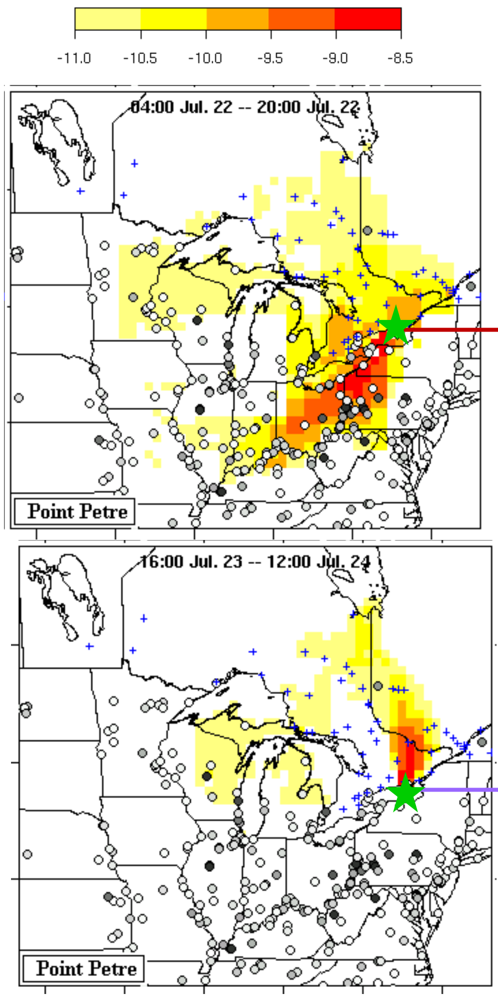
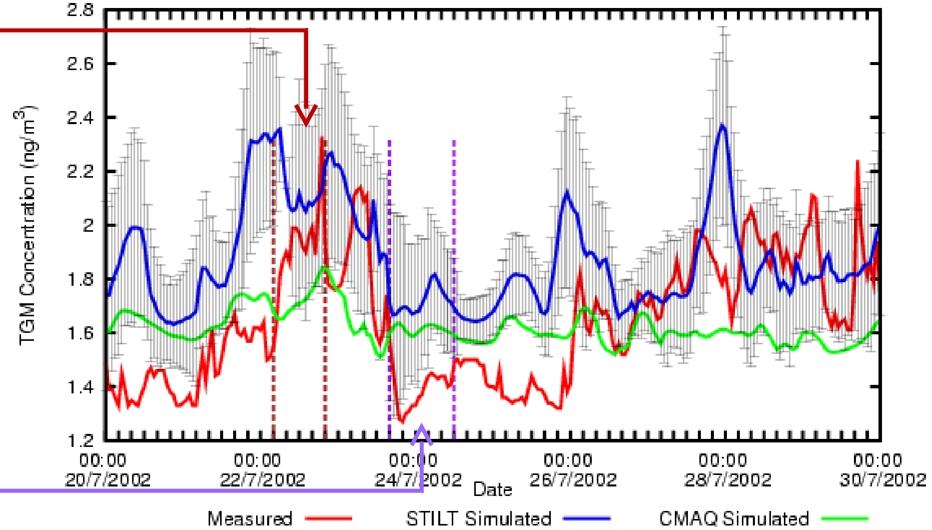
Point Petre, July 2002





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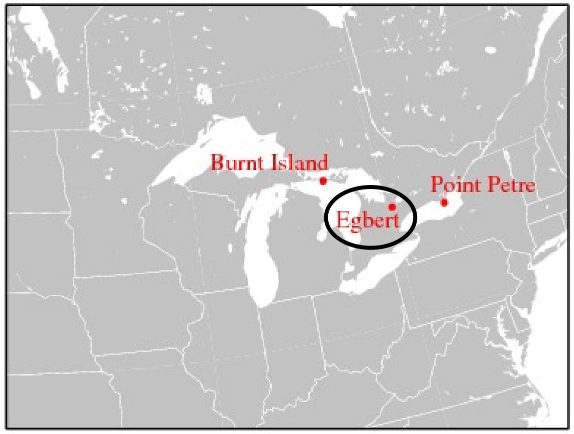
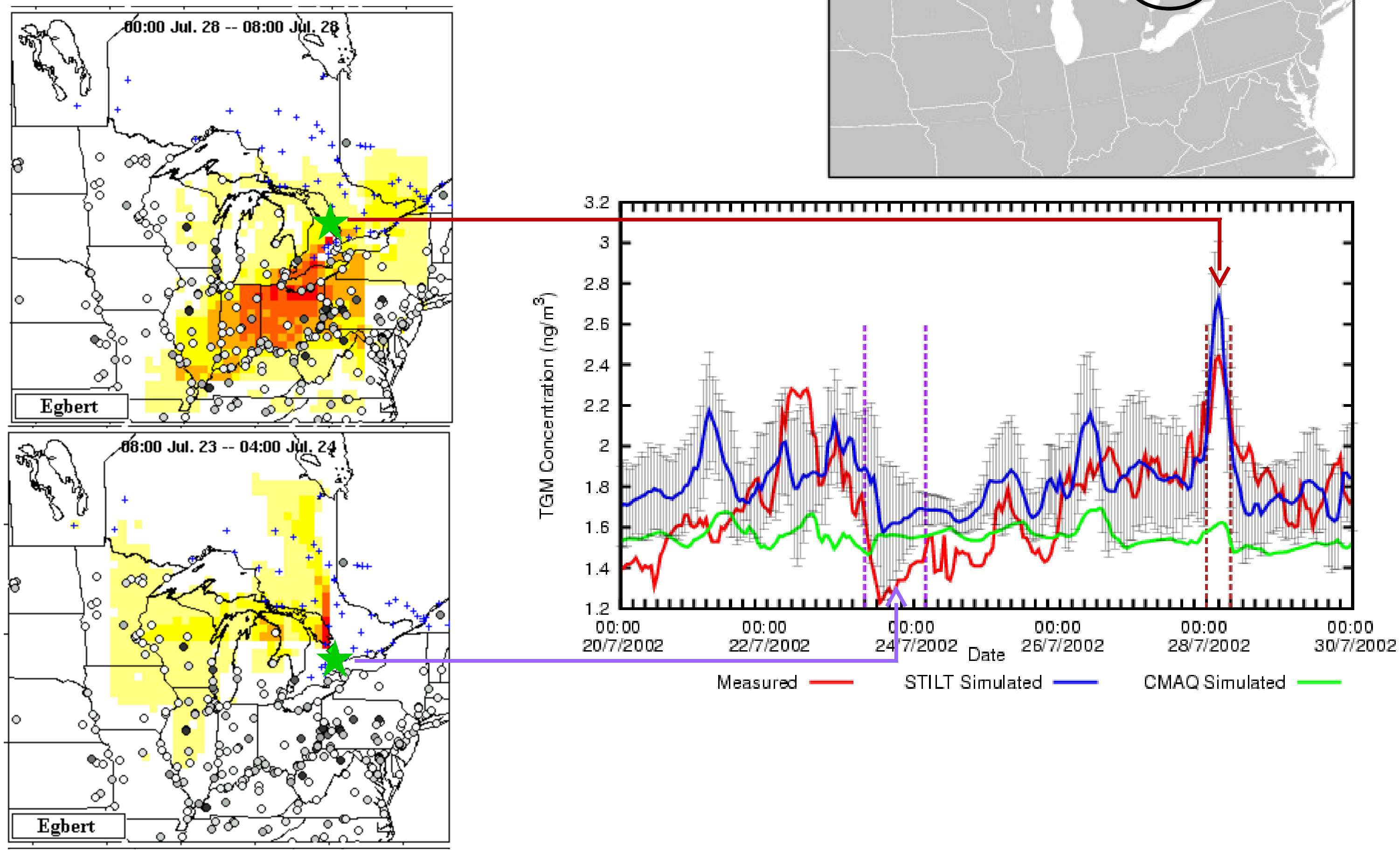
Egbert, July 2002





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Conclusions

- Lagrangian model can identify and quantify sources causing short term plumes that are not well characterised by Eulerian CTM
- Same meteorology and emissions are used in both cases leading to a small increase in computational effort
- Lagrangian particle model examines only that part of the space that is relevant to the measurement
- Use of large numbers of Lagrangian particles (hundreds-thousands) ensures accuracy of source identification