High Resolution Simulations of Particle Sulfate Formation in Lake Breeze Fronts: Process Tracking and Implications for Forecasting.

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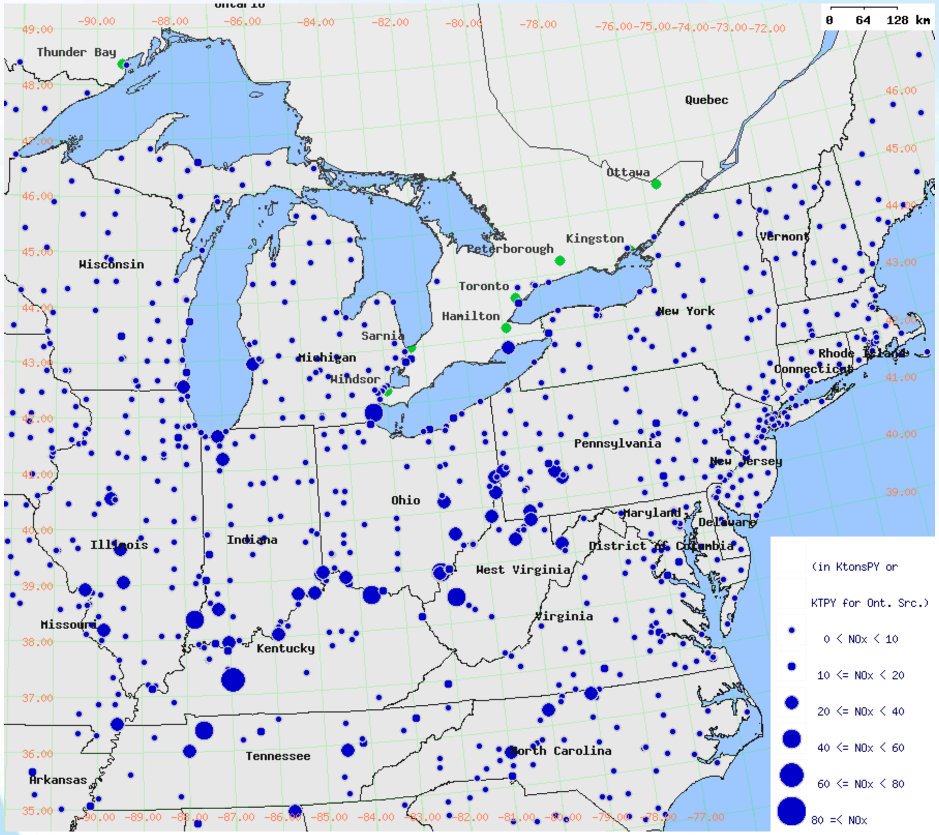
Air Quality Forecasting Workshop, Boulder, Co, Dec 2009

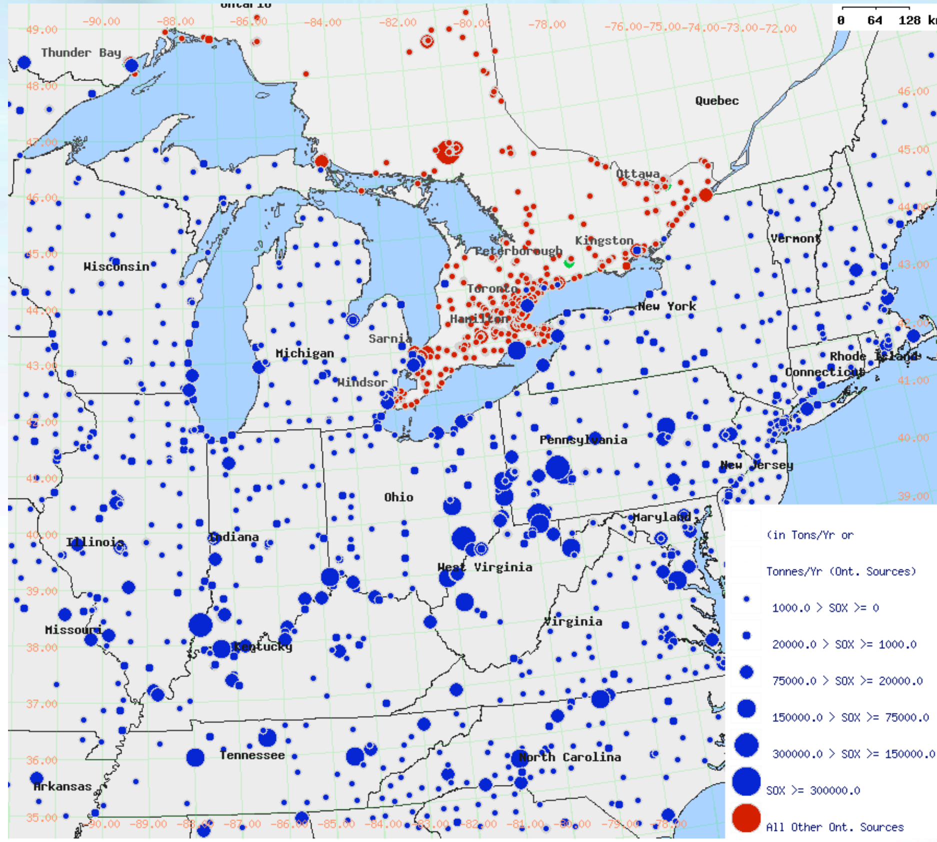
BAQS-Met: Border Air-Quality Study-Meteorology

Point Source Emissions

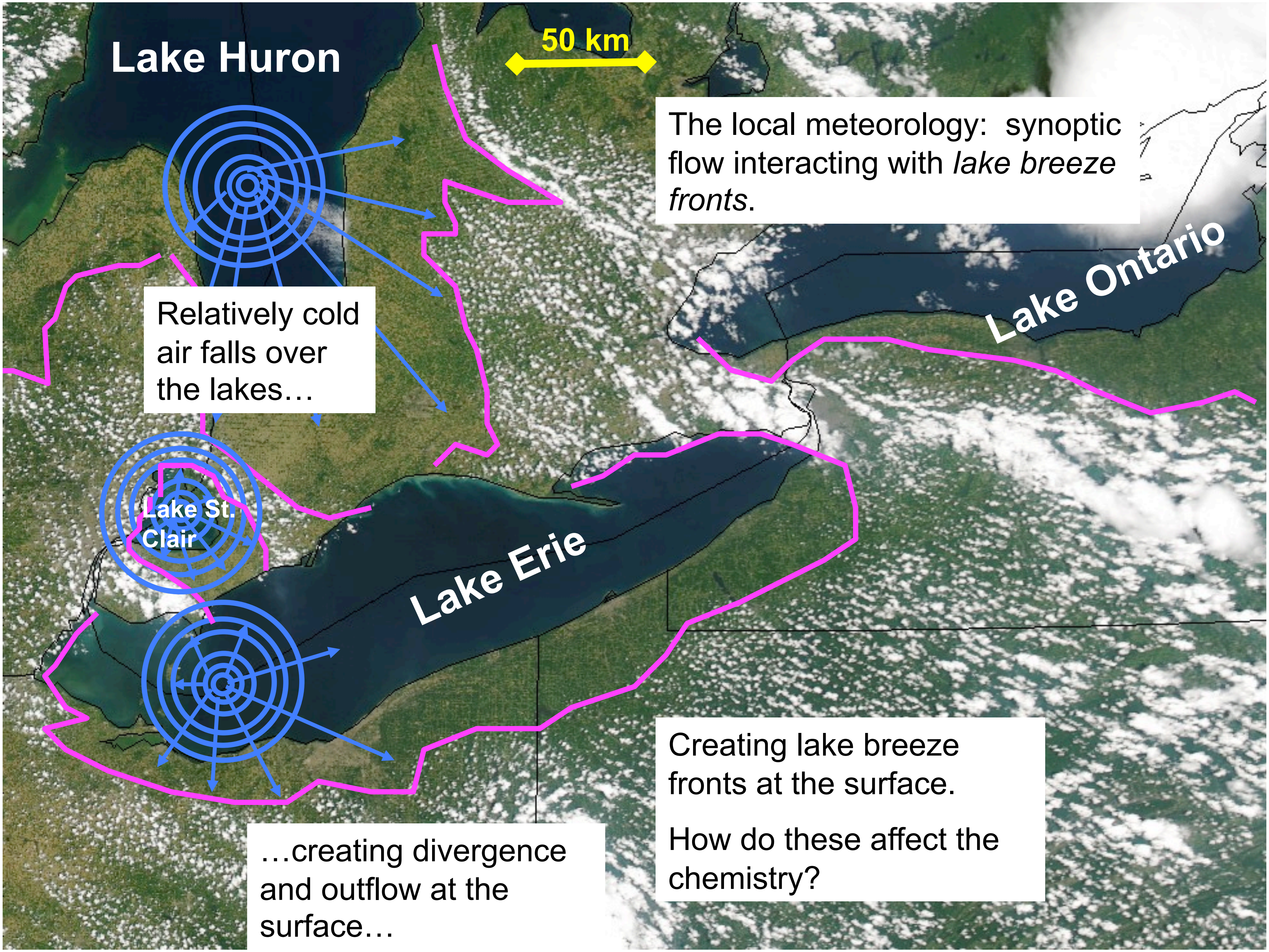
nominally for the year 2000



NO$_x$ ~ 500K t/y



SO_2 ~ 600K t/y


Lake Huron
50 km
The local meteorology: synoptic flow interacting with *lake breeze fronts*.
Lake Ontario
Relatively cold air falls over the lakes…
Lake St. Clair
Lake Erie
Creating lake breeze fronts at the surface.
How do these affect the chemistry?
…creating divergence and outflow at the surface…

What is the nature of the lake influence?

- What is the impact of the local circulation and emissions on local air-quality (versus long-range transport)?
- How do trace gases and particles evolve downwind of a large, midlatitude urban and industrial centre (Detroit)?
- Some analysis with AURAMS…

A Unified Regional Air-Quality Modelling System (AURAMS)

National Emission Inventories (Cdn, U.S.)

Population data
Econometric data
Land-use data

Geophysical data

Meteorological observations (OA)

SMOKE
point mobile area biogenics

GEM/GEM-LAM
(prognostic meteorological model)

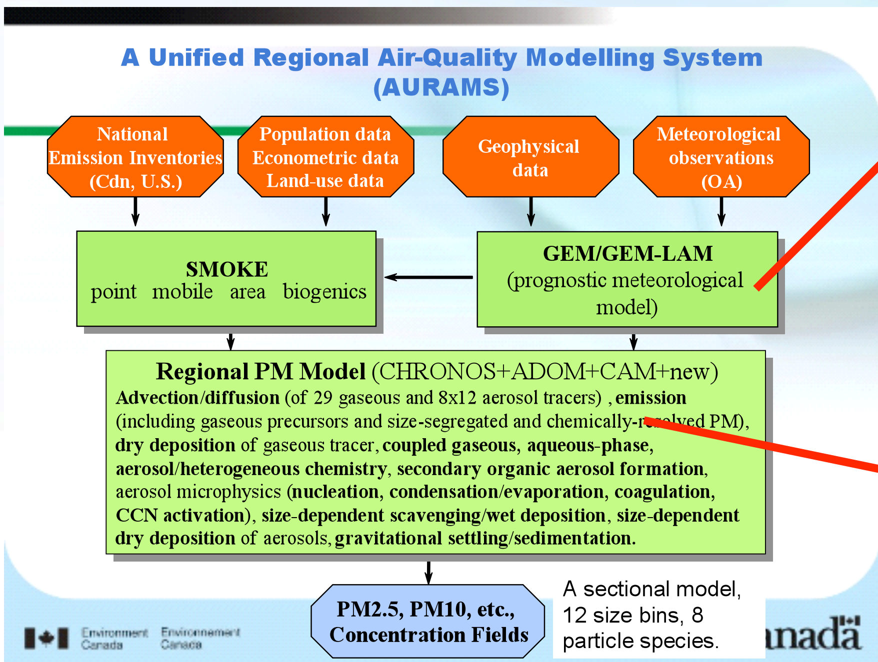
Regional PM Model (CHRONOS+ADOM+CAM+new)

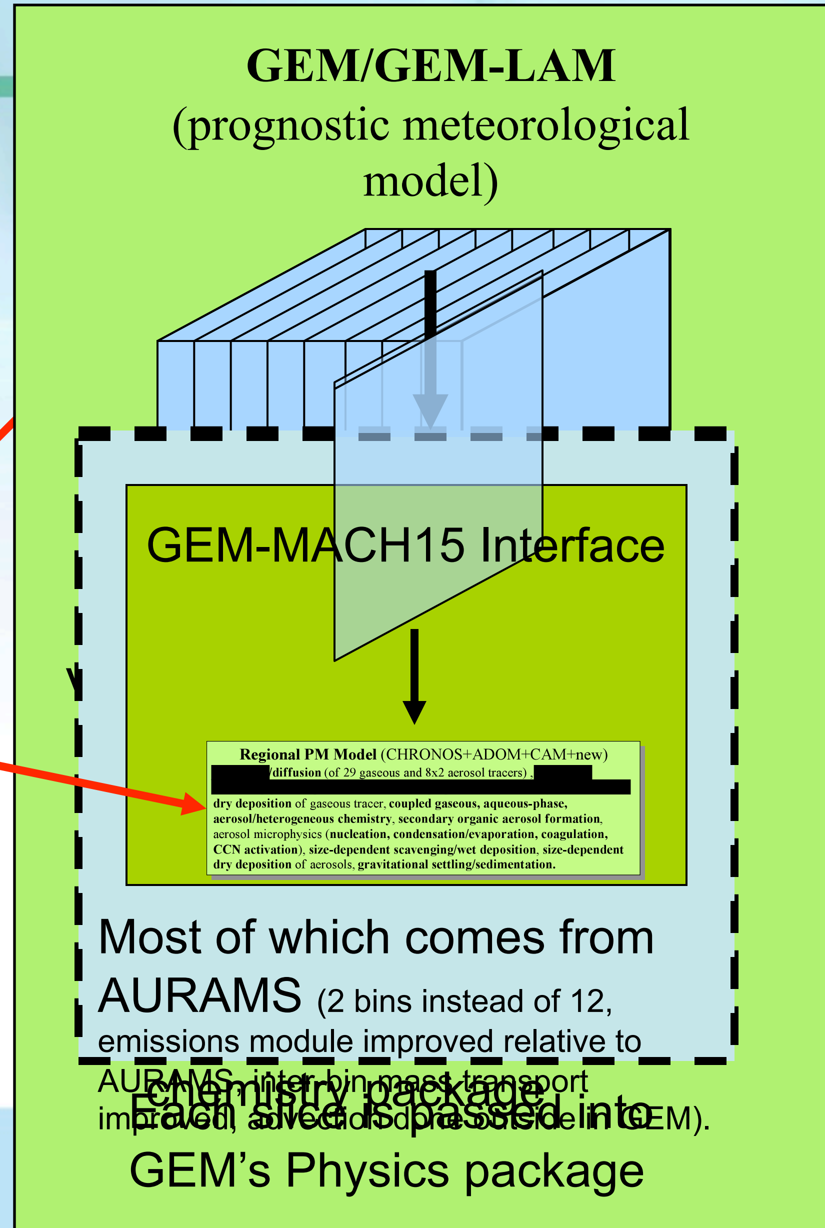
Advection/diffusion (of 29 gaseous and 8x12 aerosol tracers) , **emission** (including gaseous precursors and size-segregated and chemically-resolved PM), **dry deposition** of gaseous tracer, **coupled gaseous, aqueous-phase, aerosol/ heterogeneous chemistry**, **secondary organic aerosol formation**, aerosol microphysics (**nucleation, condensation/evaporation, coagulation, CCN activation**), **size-dependent scavenging/wet deposition**, **size-dependent dry deposition** of aerosols, **gravitational settling/sedimentation.**

PM2.5, PM10, etc., Concentration Fields

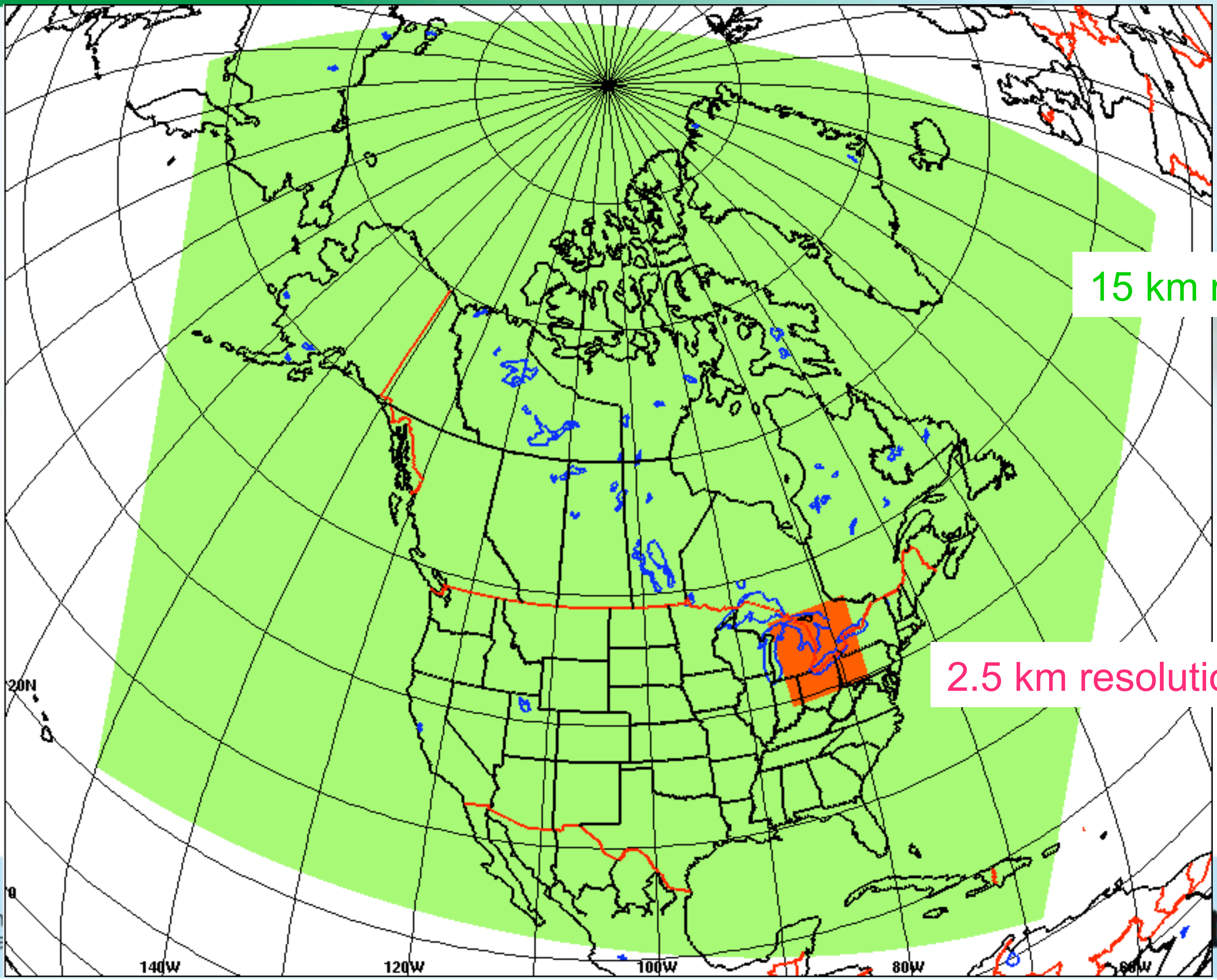
A sectional model, 12 size bins, 8 particle species.

Relationship between AURAMS and GEM-MACH15

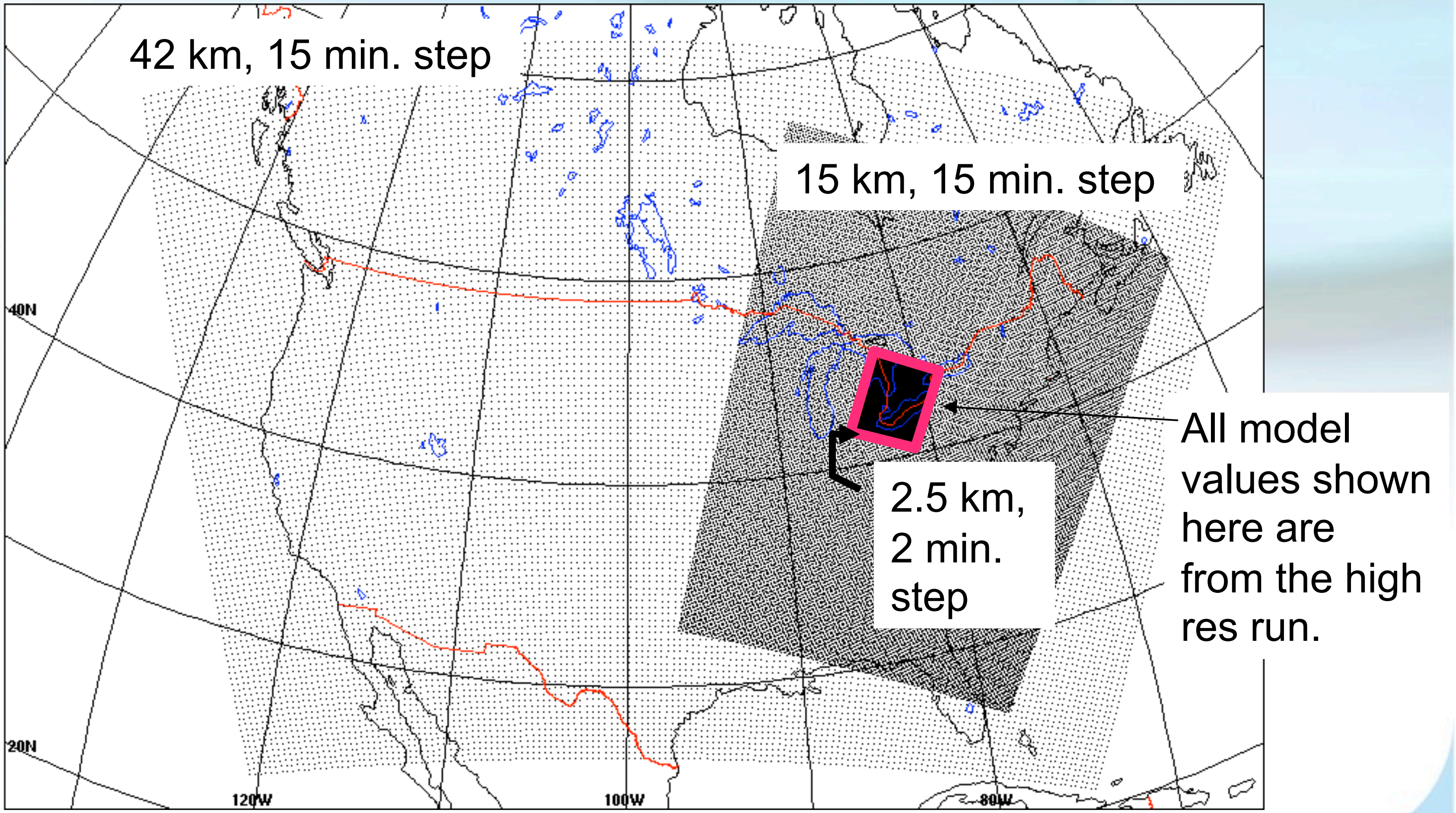




GEM nested grids for AURAMS input



AURAMS Nested Modelling Domains



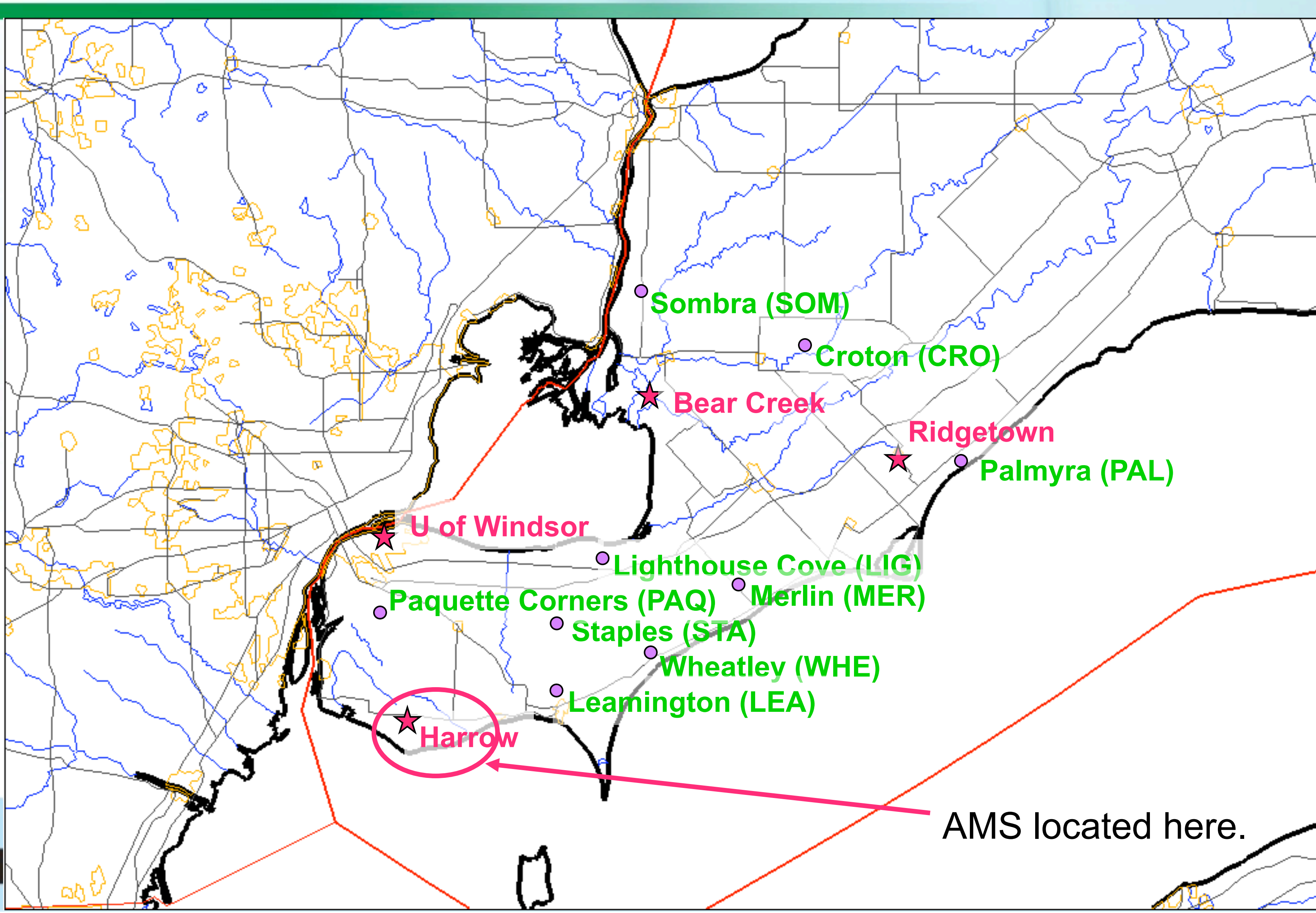
28 vertical levels (14 below 2km agl).

All of the above processes can change particle mass

- PM may be brought in from elsewhere (advection, diffusion).
- Particles may be created (nucleation)
- Particles can grow (condensation, coagulation)
- Mass may change (+/-) due to heterogeneous chemistry.
- Below clouds, particles may be scavenged.
- Particles may settle and reach the ground.

→ ***Which processes dominate, and why?***

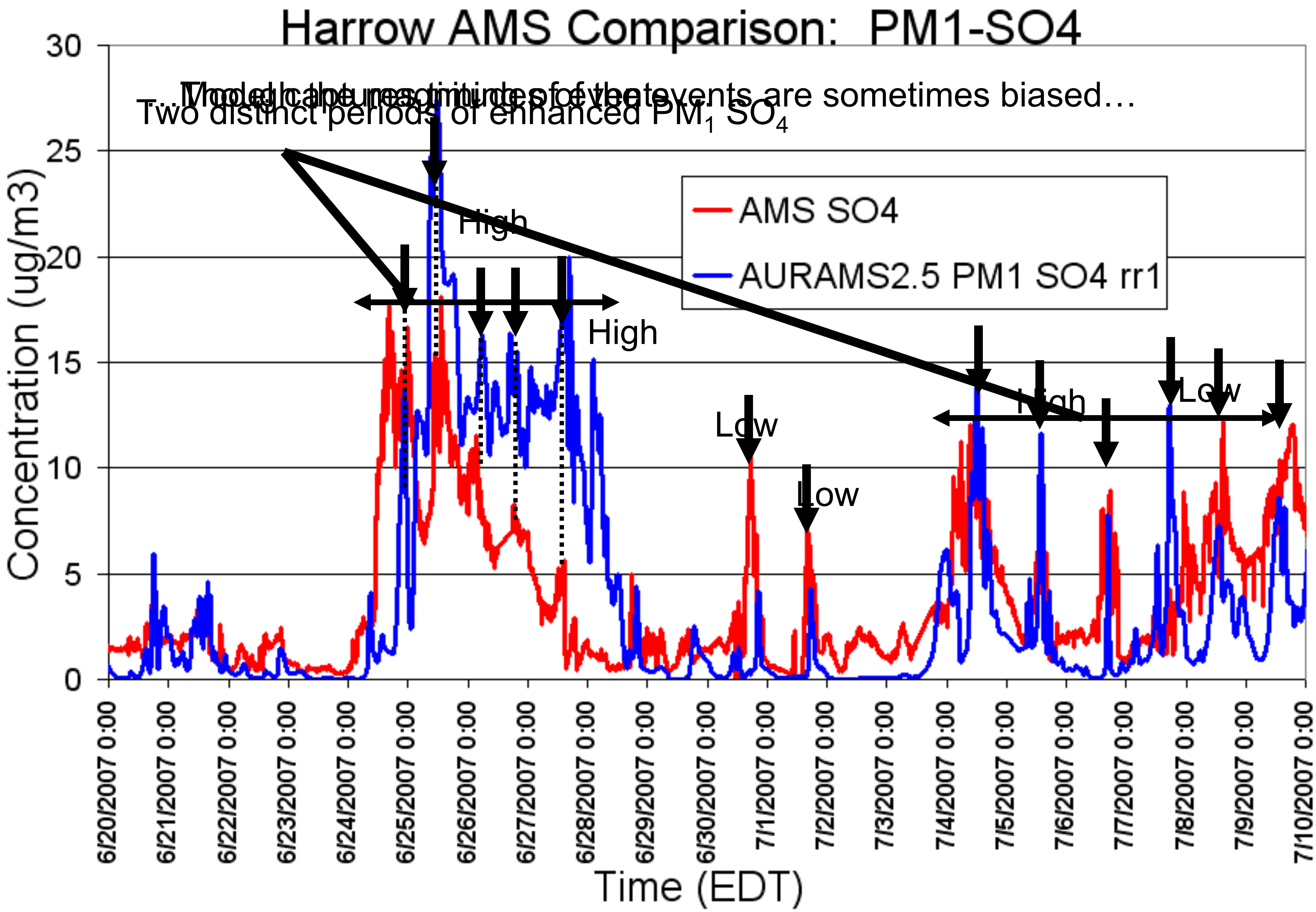
Supersites and EC ozone and total PM stations



Comparison to surface observations: Harrow supersite.

- AURAMS' PM_1-SO_4: model values are aggregation of first 6 particle bins + 0.042 of the 7th bin: equivalent to AMS size range.

Comparison to surface obs: Harrow.



Stats (10 minute averages matched)

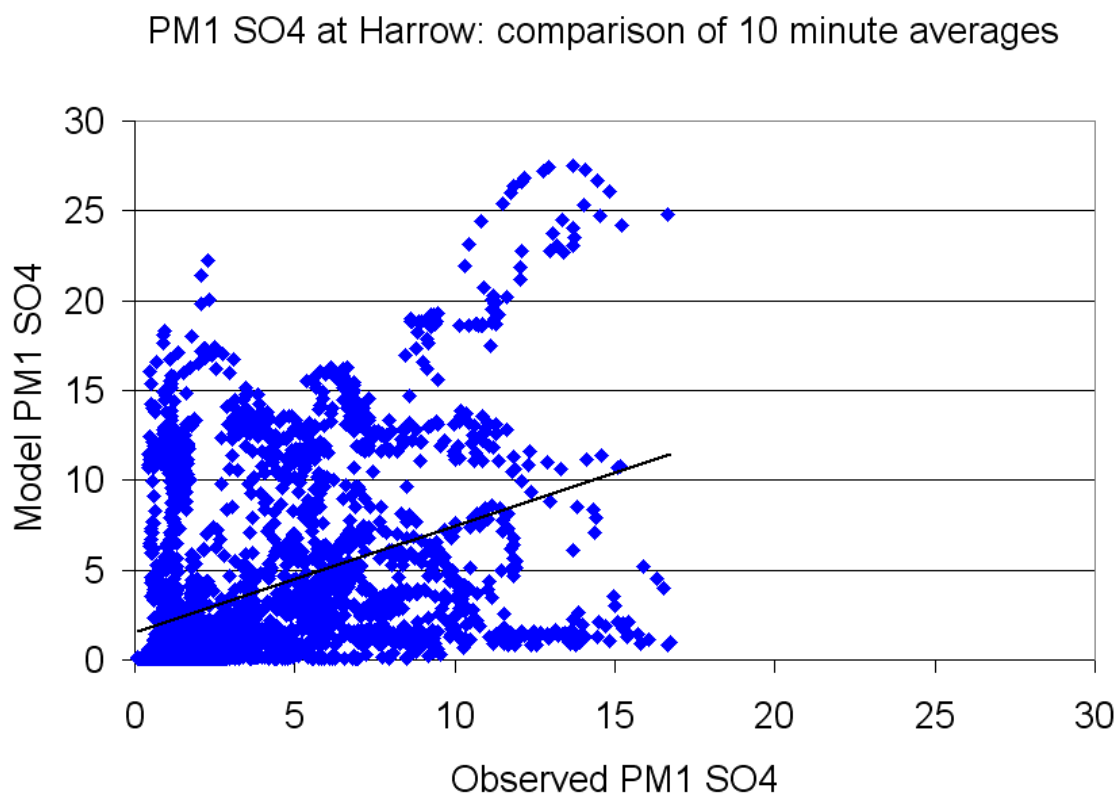
- R: 0.3964
- Best fit:

model = 0.5933 obs + 1.556

- Mean bias:

-3.243E-03 ug/m^3

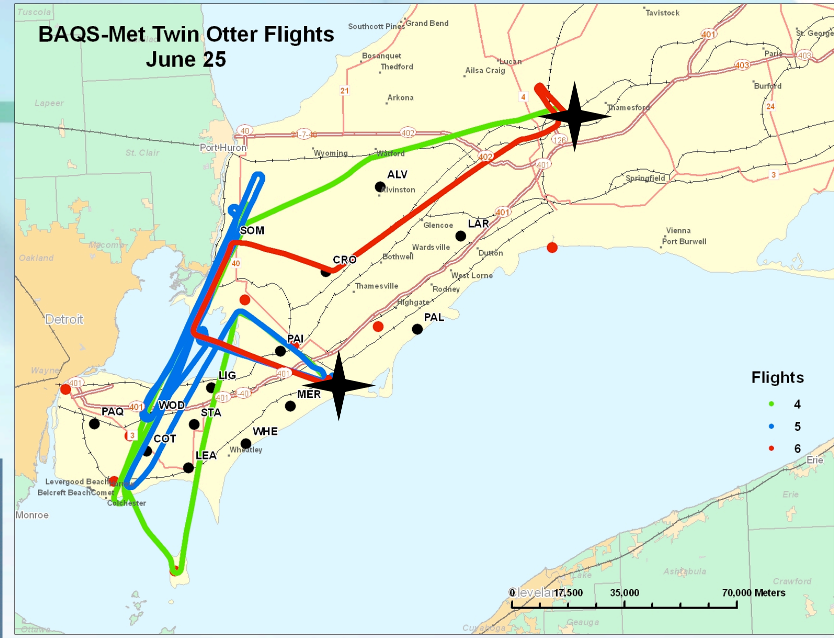
- Mean error: 3.376 ug/m^3

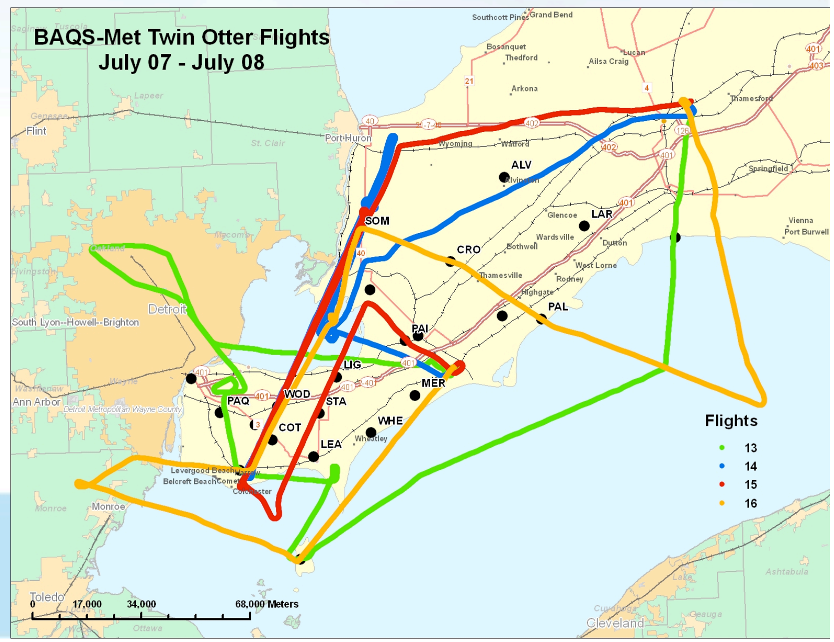


Overpredictions in the 1st half of the period are being offset by underpredictions in the second half.

Aircraft: Twin Otter with AMS on board

- National Research Council Twin Otter
  - 16 flights for ~30 hrs





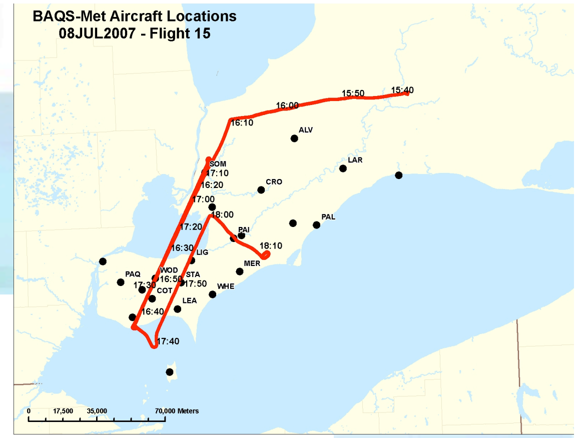
Comparison to Aircraft AMS measurements

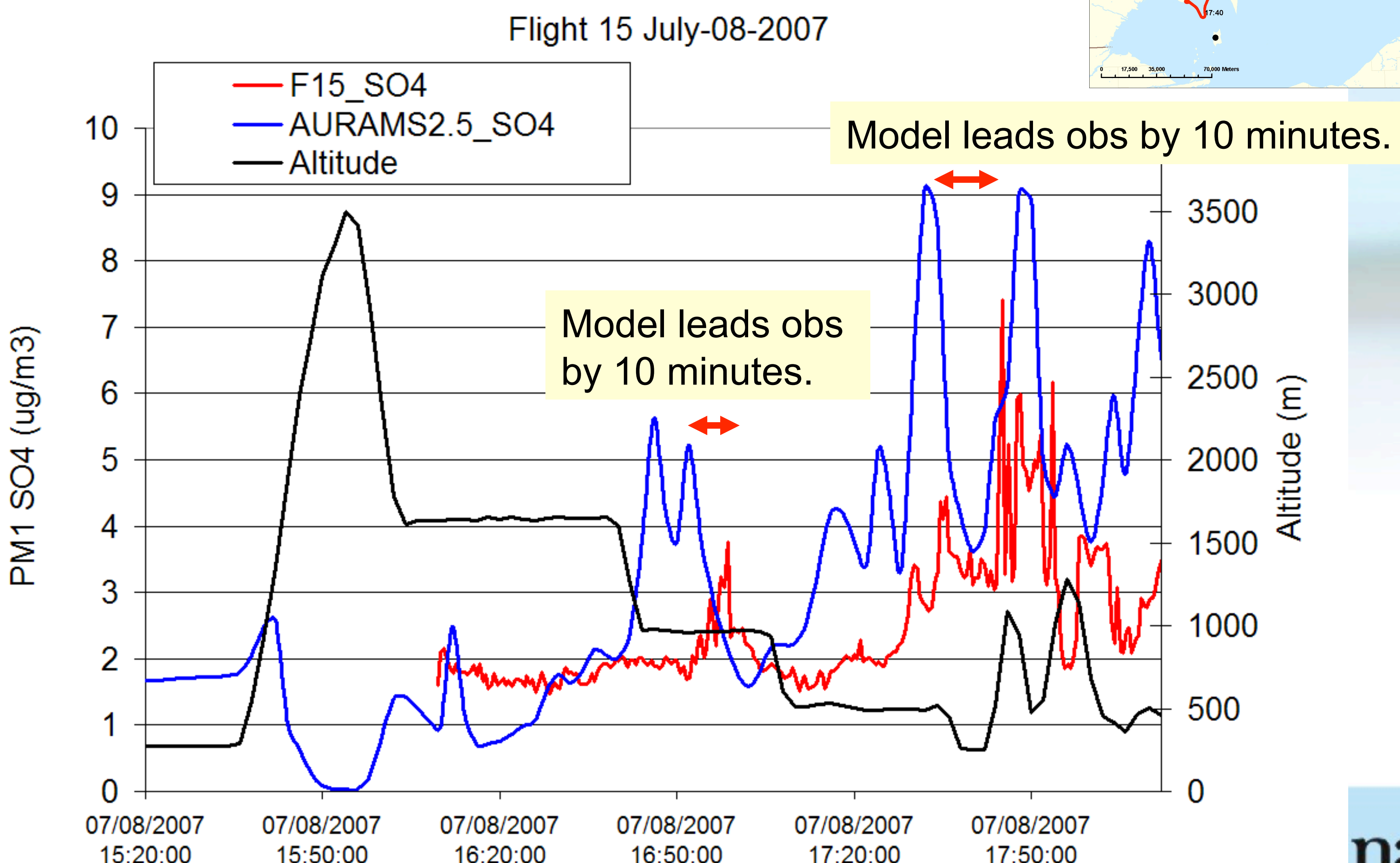
- R: 0.5541
- Model = 1.029 Obs + 3.663
- Mean Bias: 3.759
- Mean Error: 4.592

… total for all flights (967 two-minute averages).

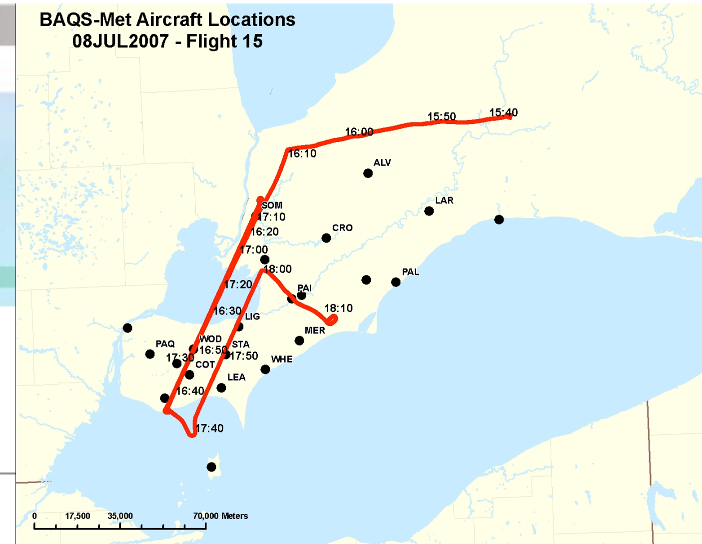
Individual flights were better or worse than this (e.g. Flight 15, R = 0.67, Flight 9 R = - 0.019)

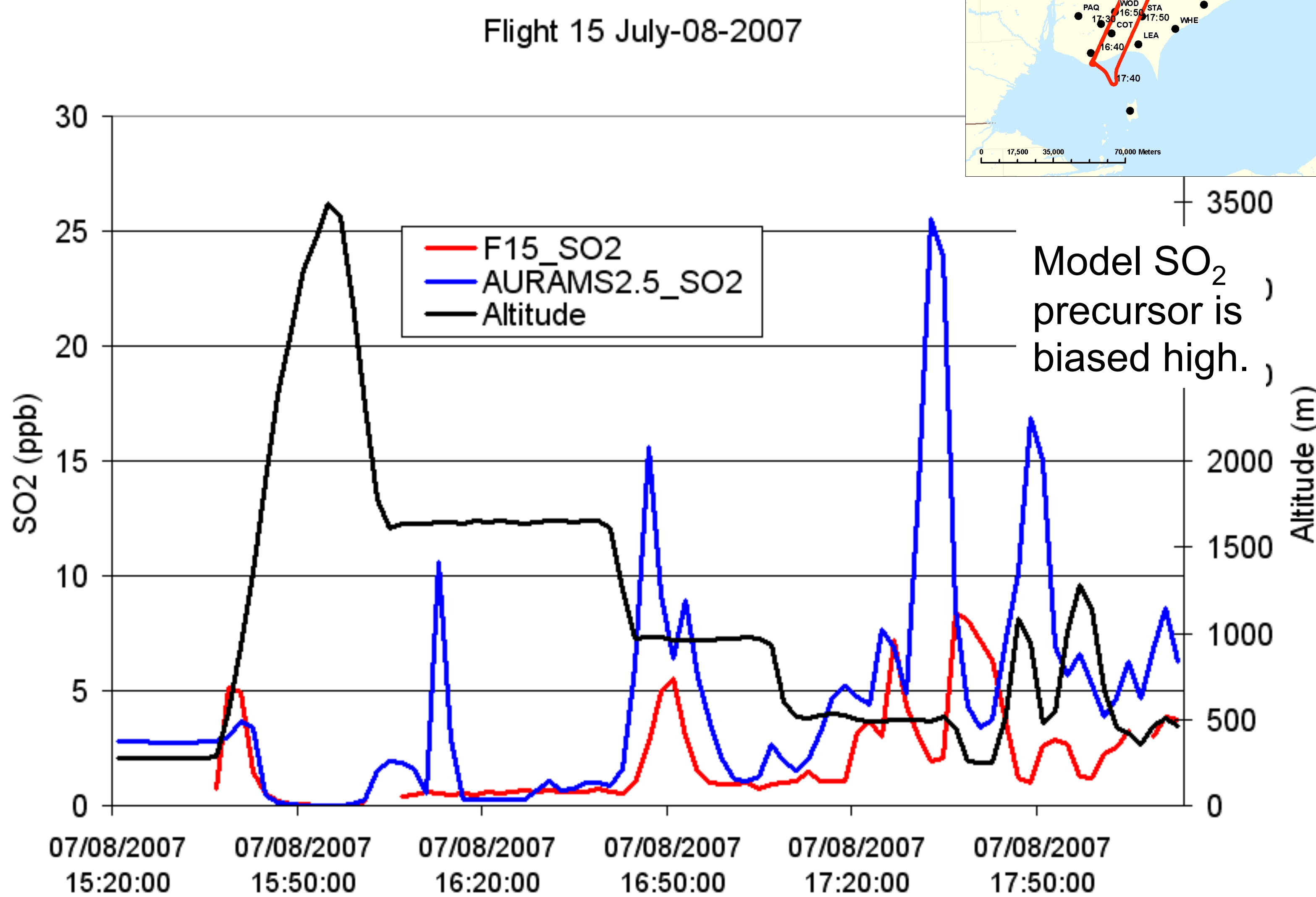
Example: Flight 15 (R=0.67)





Example: Flight 15 (R=0.67)

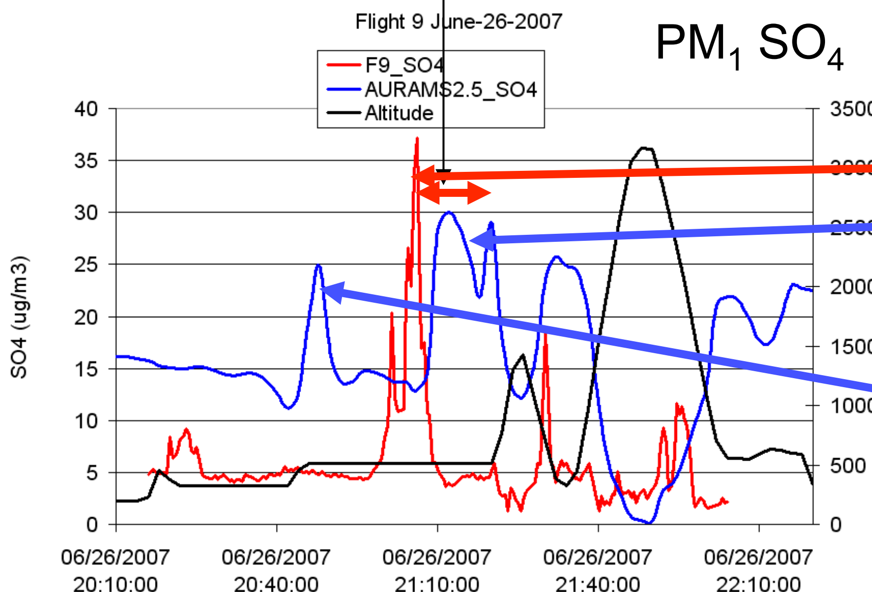


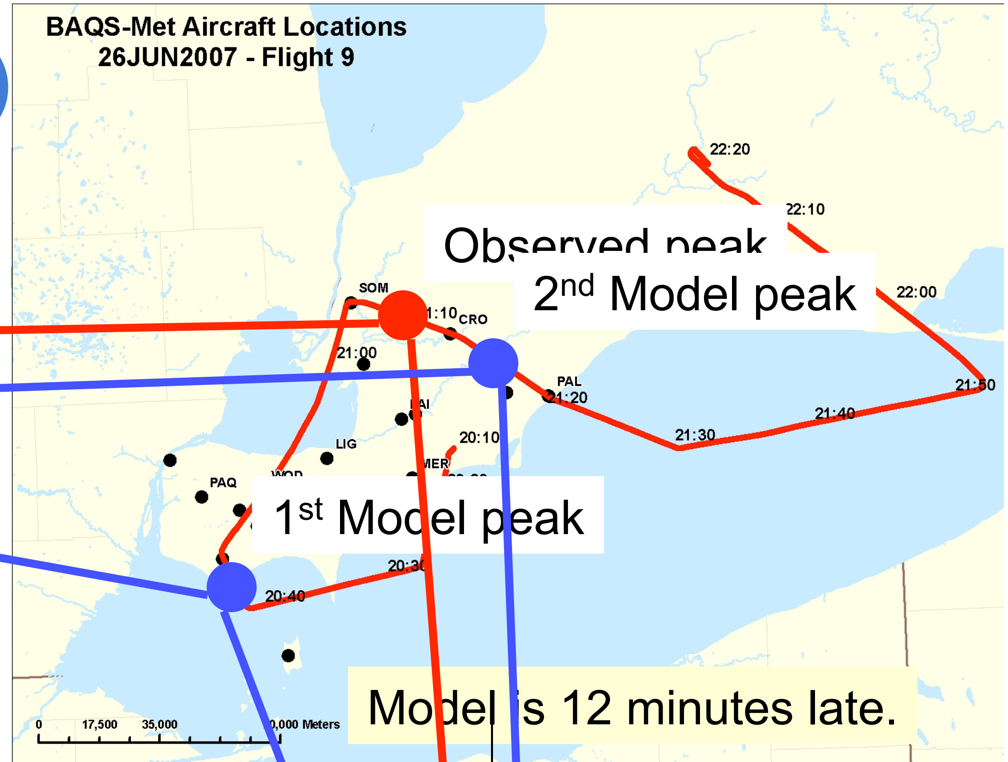


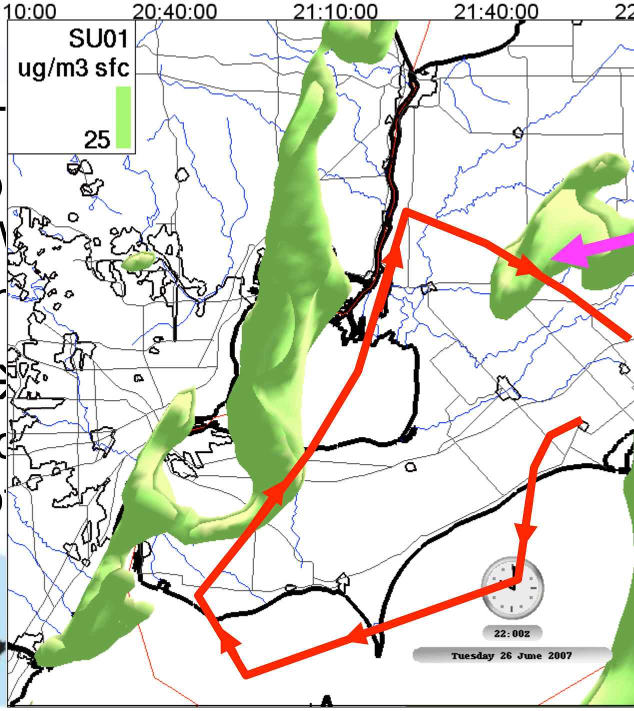
Model SO_2 precursor is biased high.

Flight ( )

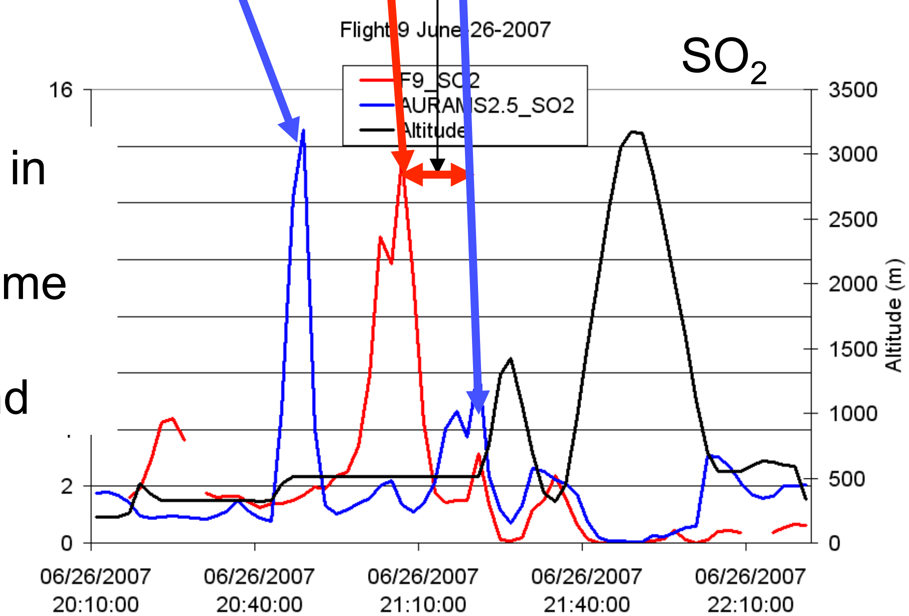
Model is 12 minutes late.







2nd peak in model is aged plume from Cleveland



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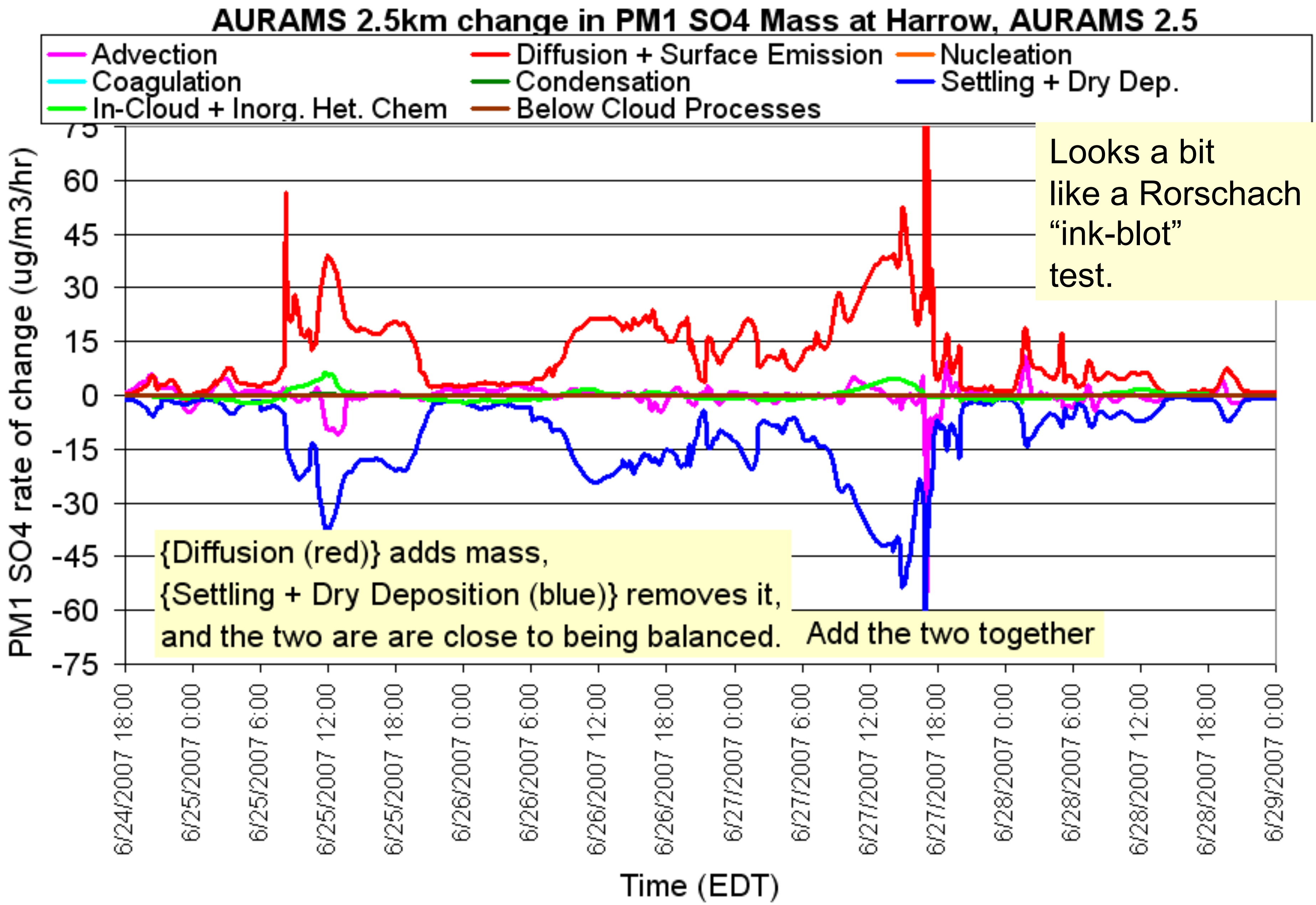
What created the (model) PM_1 SO_4?

- The above suggests that:
  - Timing is everything → small errors in timing can have a big impact, when you're comparing aircraft observations to model at this scale.
  - Some of the emissions from major point sources may be too high.

What other information can be gleaned from the model?

- Analysis using mass trackers across the particle processes in AURAMS.
- First episode (24th 18:00 EDT – 29th 0:00 EDT)
- At Harrow, extract out time series of the mass trackers (change in mass across each process; operator splitting).

Mass tracking of Particle Sulfate at Harrow.



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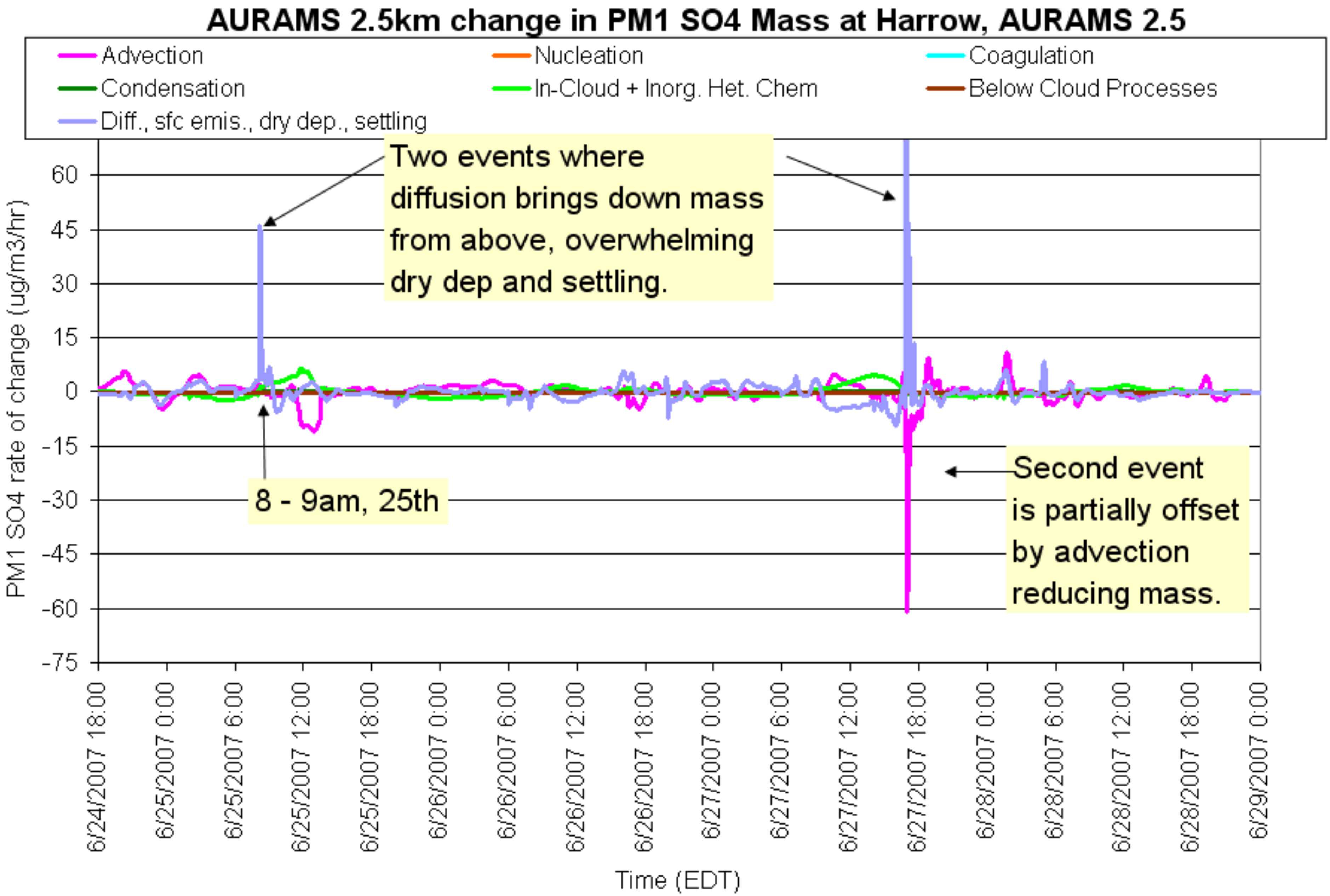

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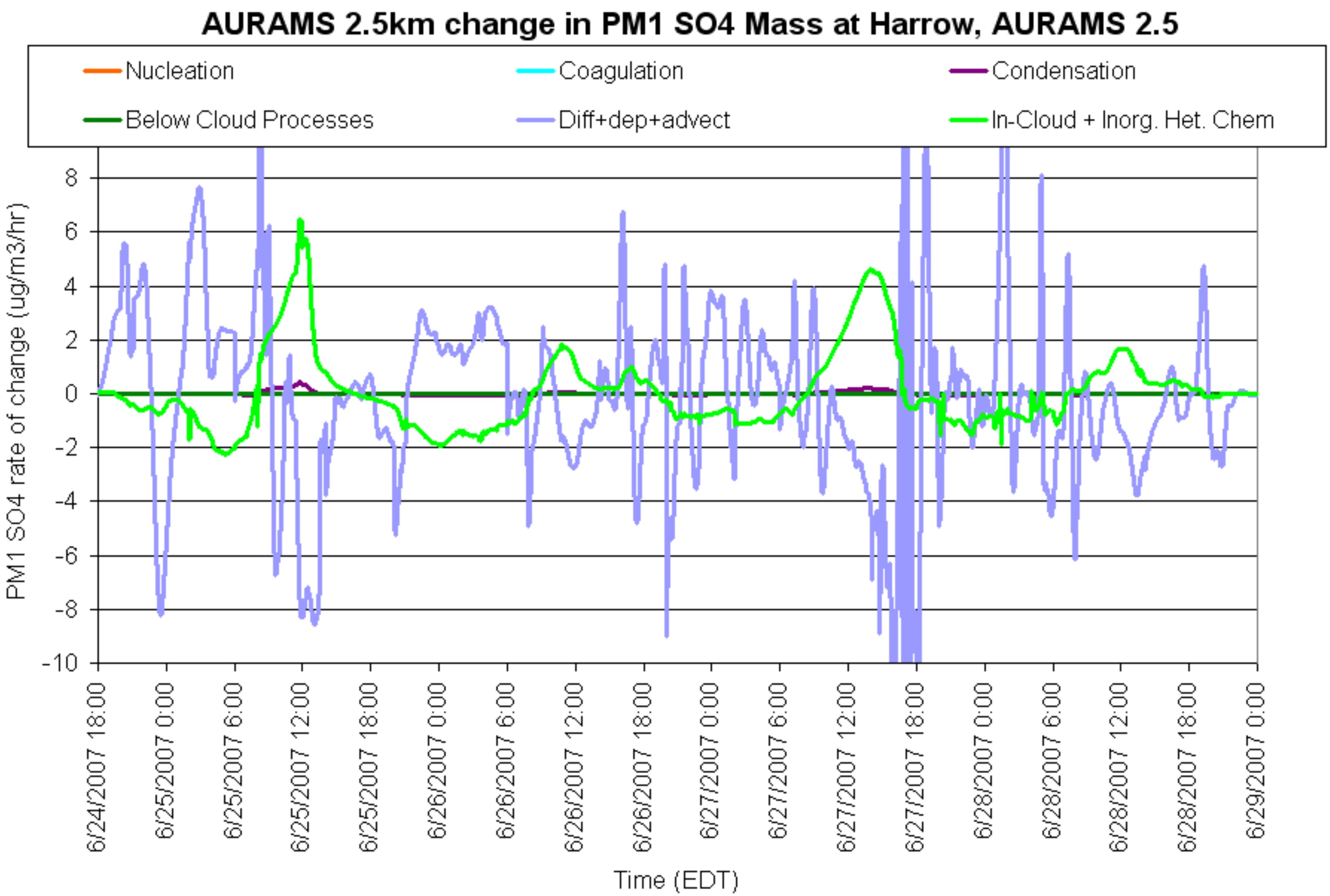

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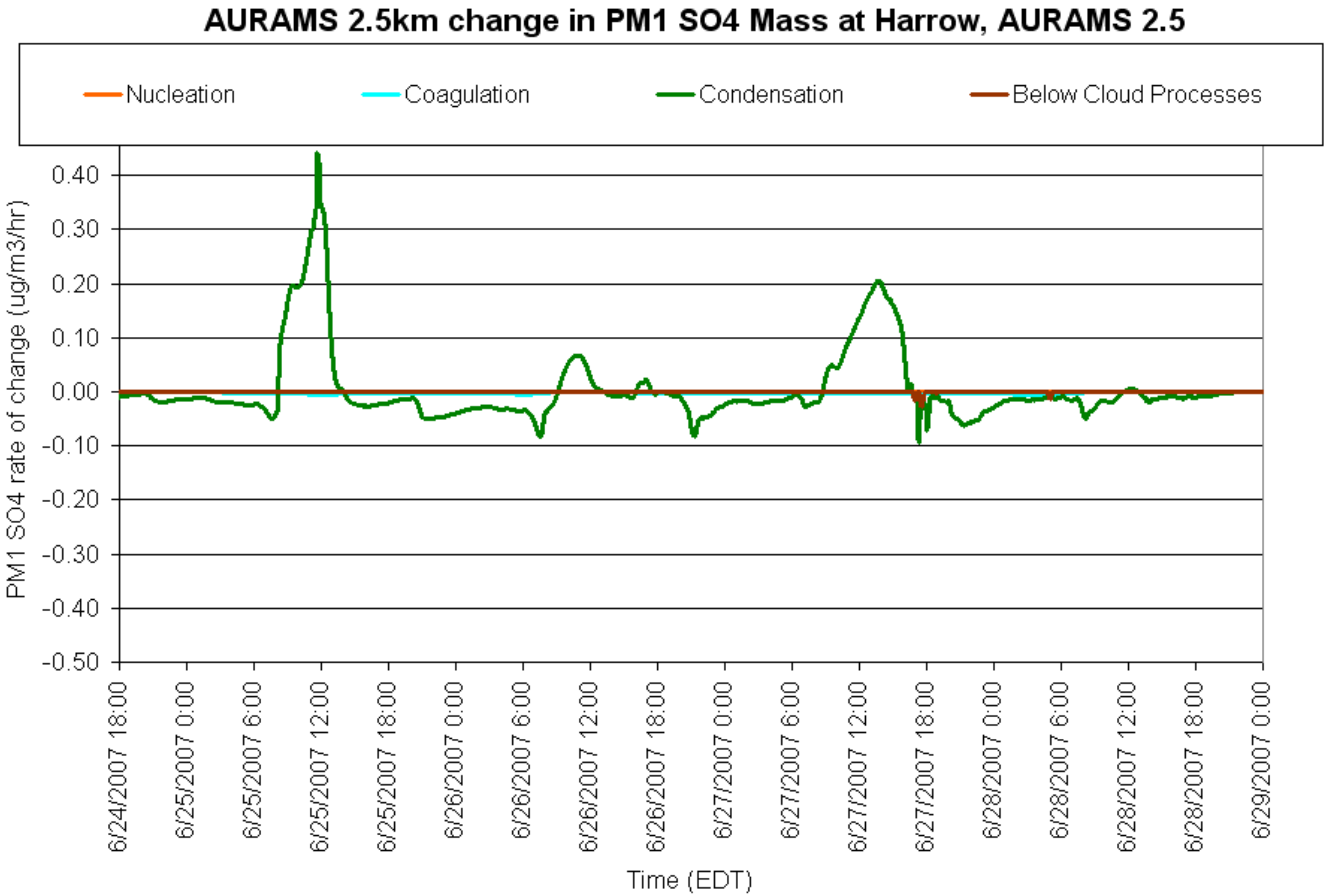

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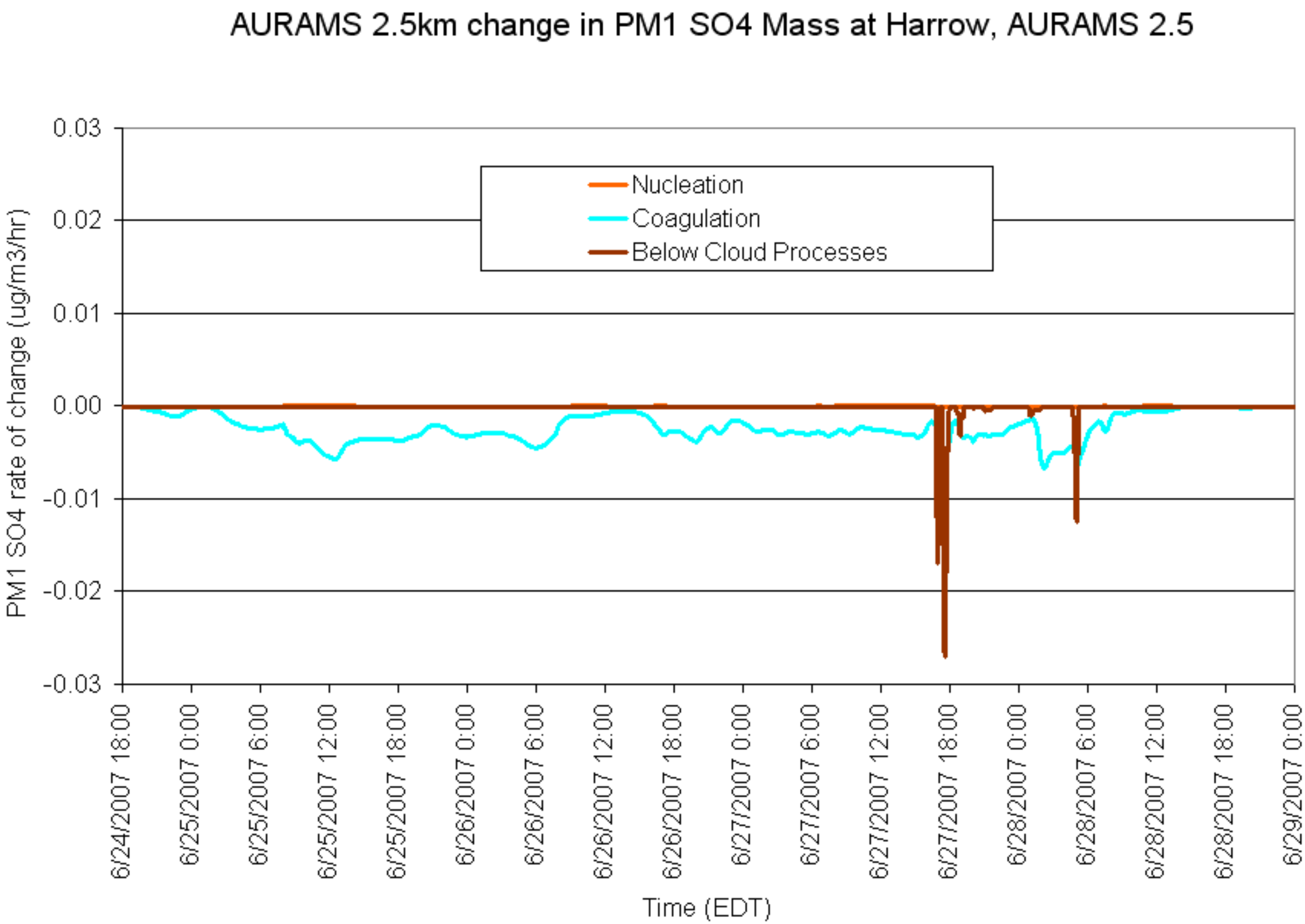

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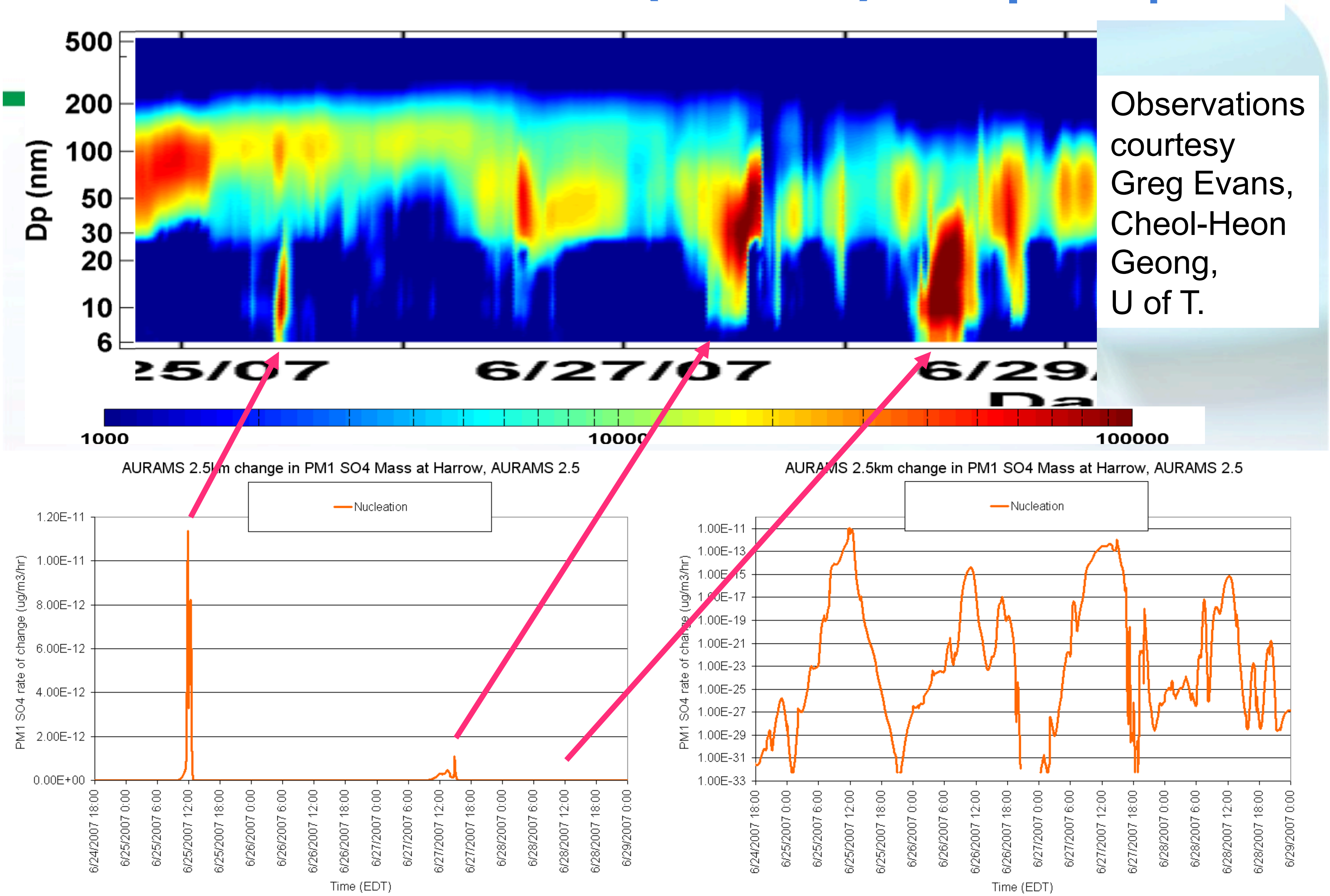
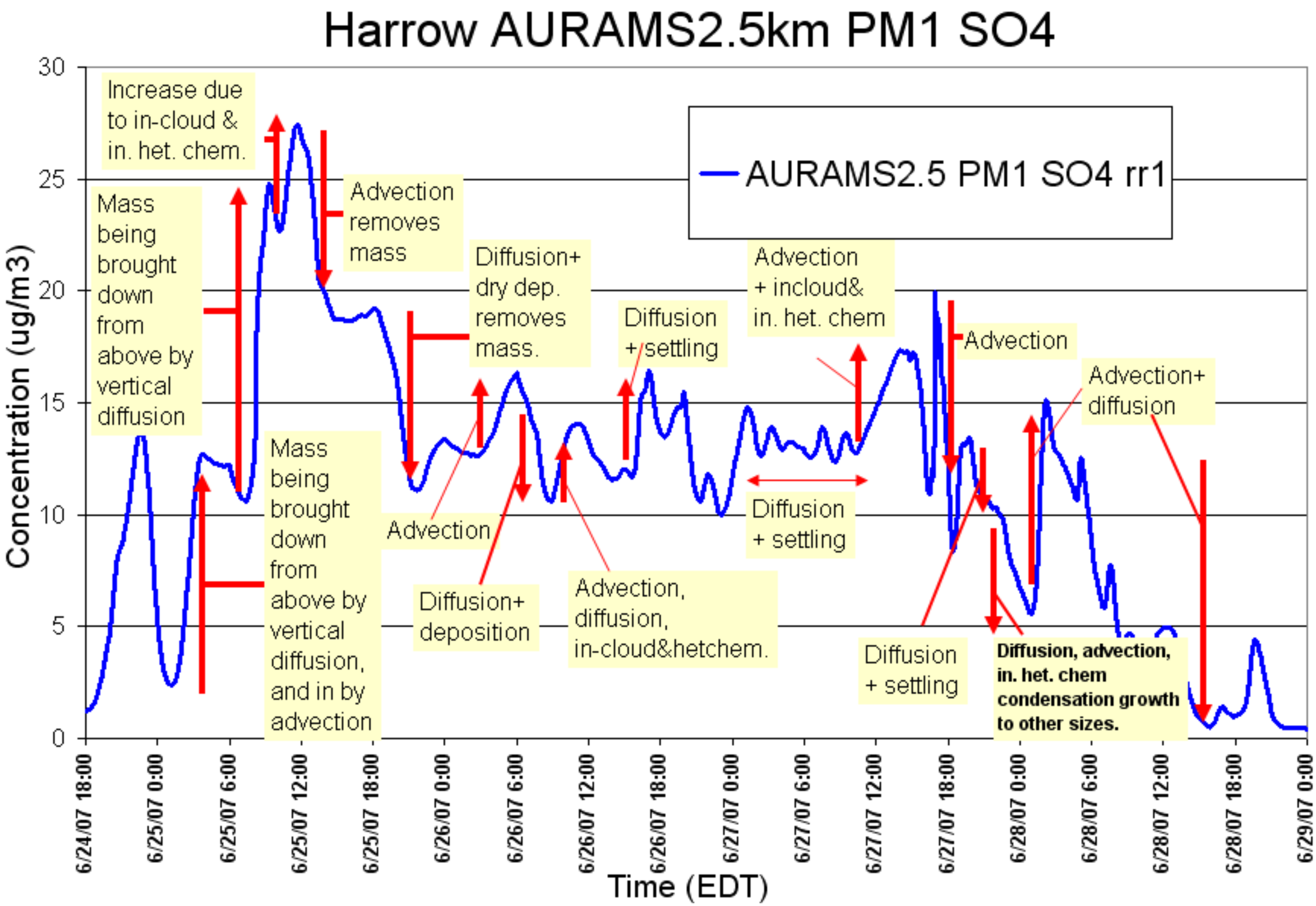
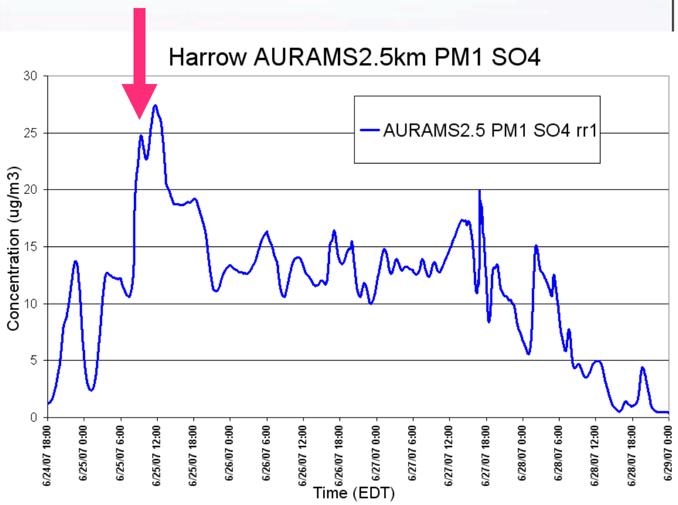


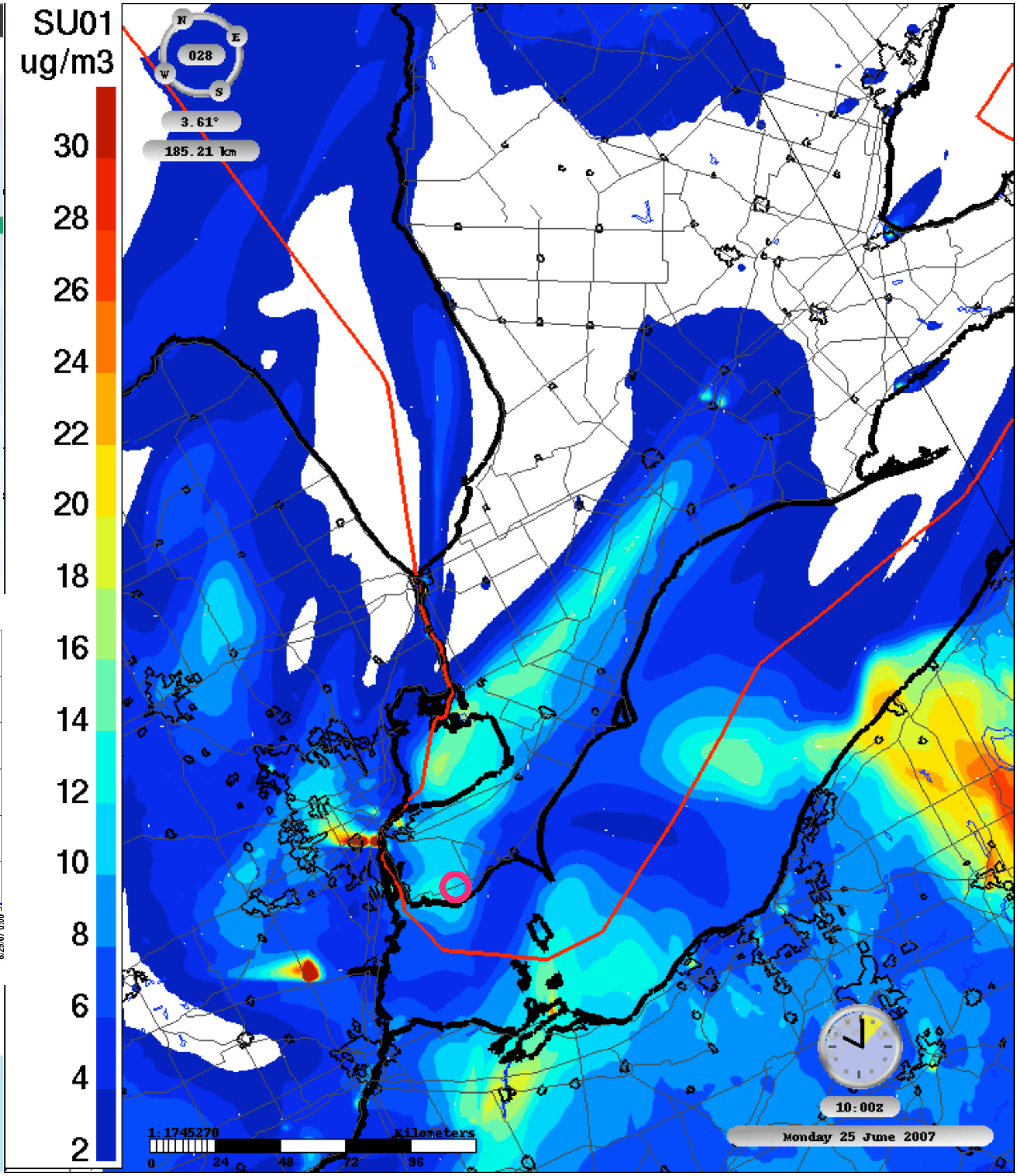
What created the (model) PM$_1$ SO$_4$?



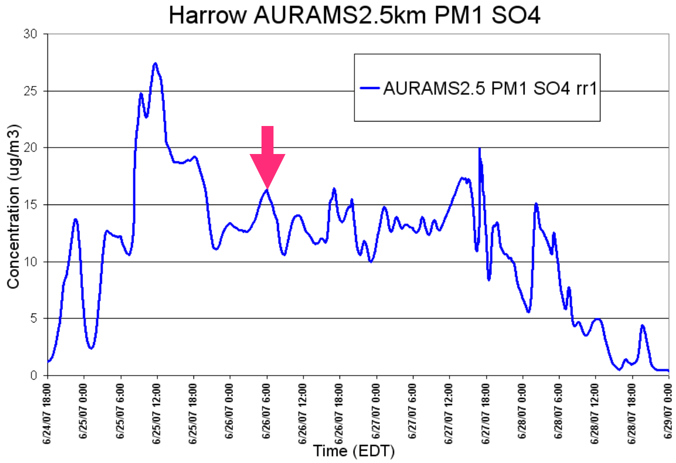
Ans.: mostly **transport** (advection, diffusion)

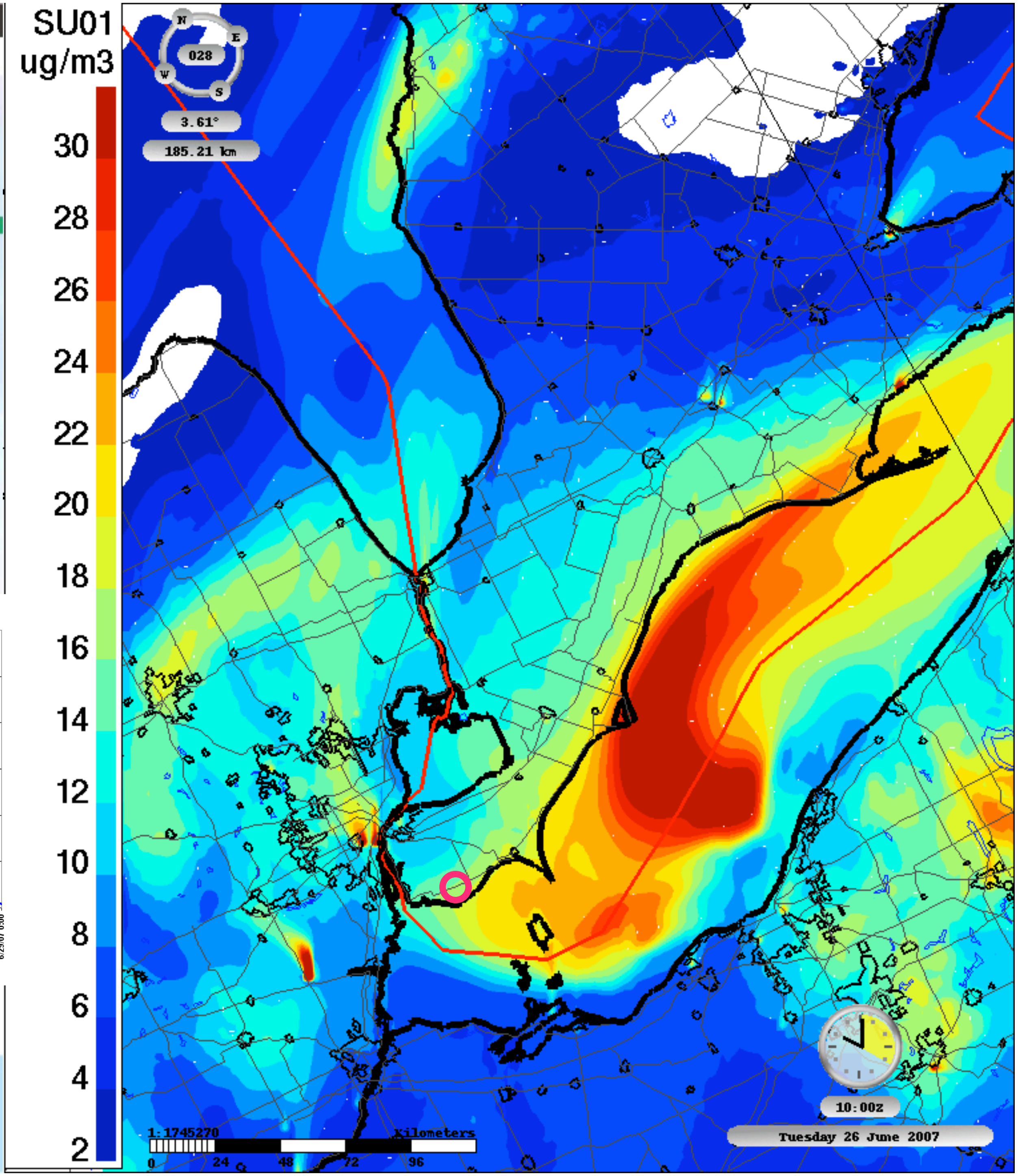
Increases on the 25th at Harrow: plumes from Cleveland crossing Lake Erie.





Increases on the 26th 10Z: Cleveland plume again.





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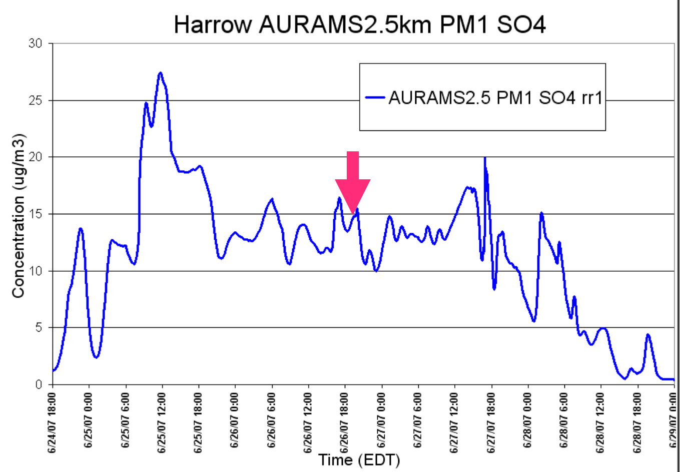
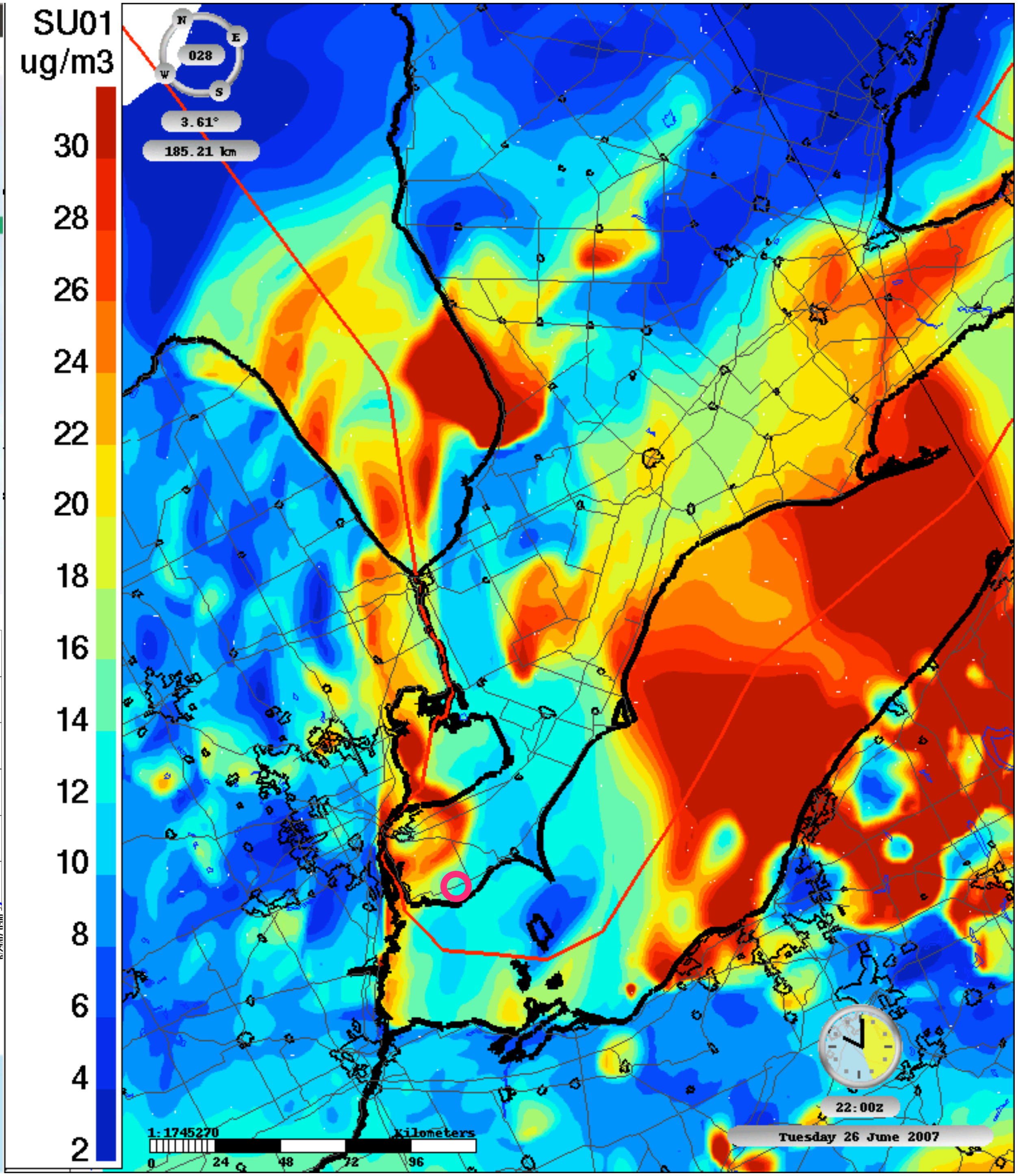
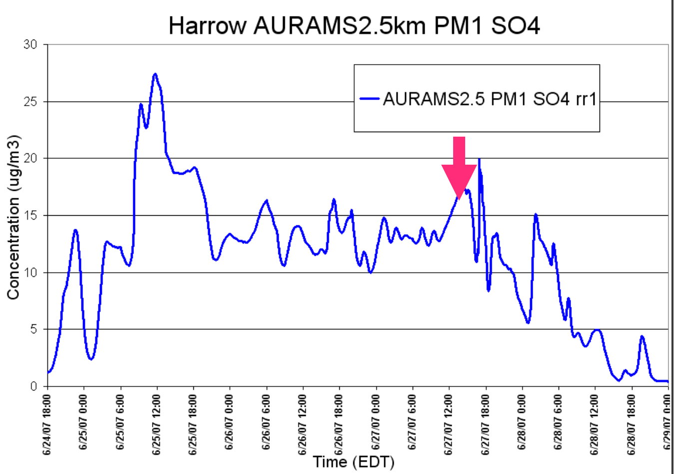
Increases on the 26th 22Z and thereafter: Detroit / Windsor; local emissions close to the measurement site.

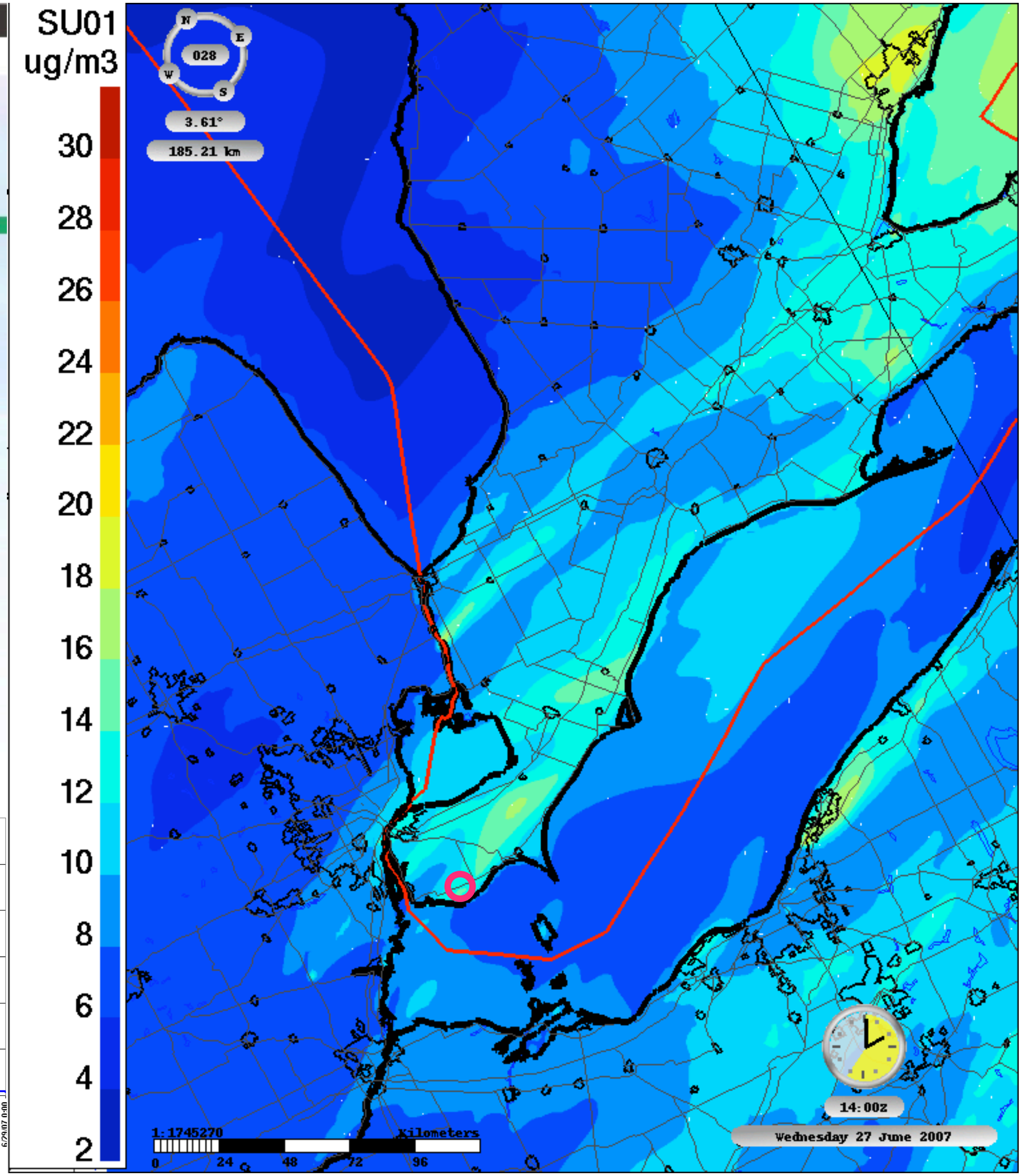




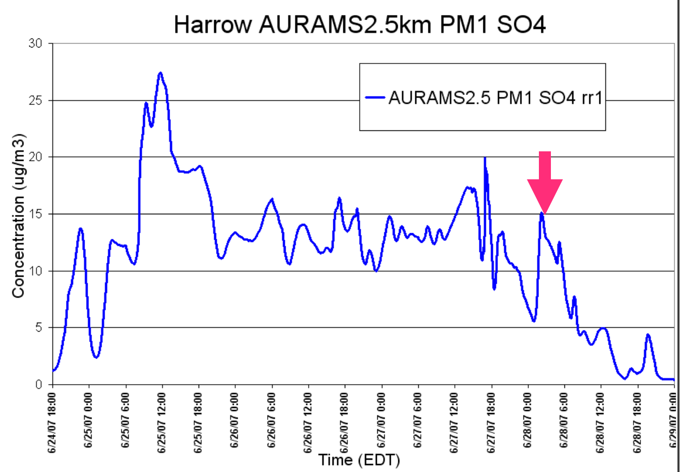
Environment Canada Environnement Canada

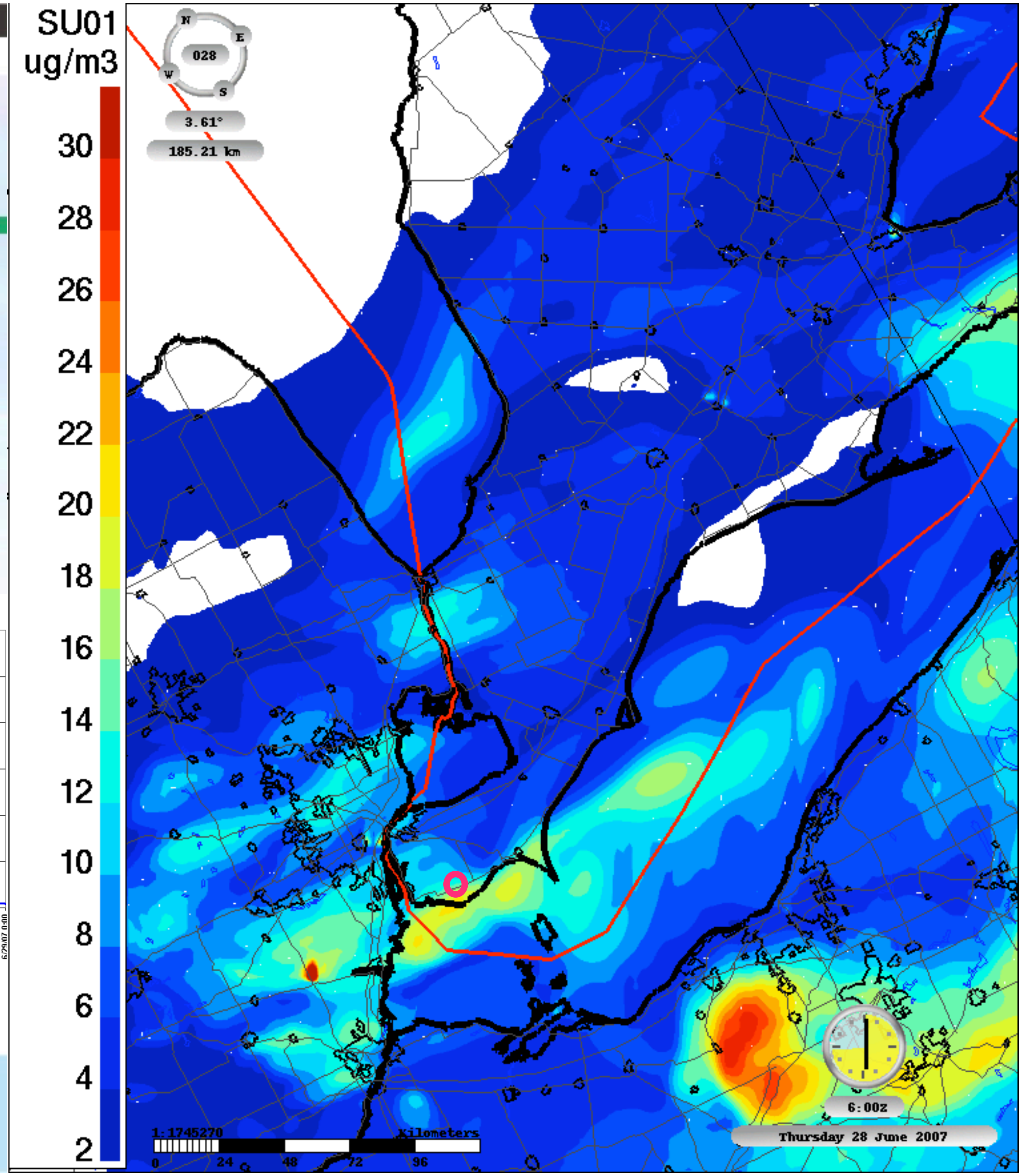
In cloud/het chem event: chemistry following fumigation of plume originating in point sources from South Detroit.

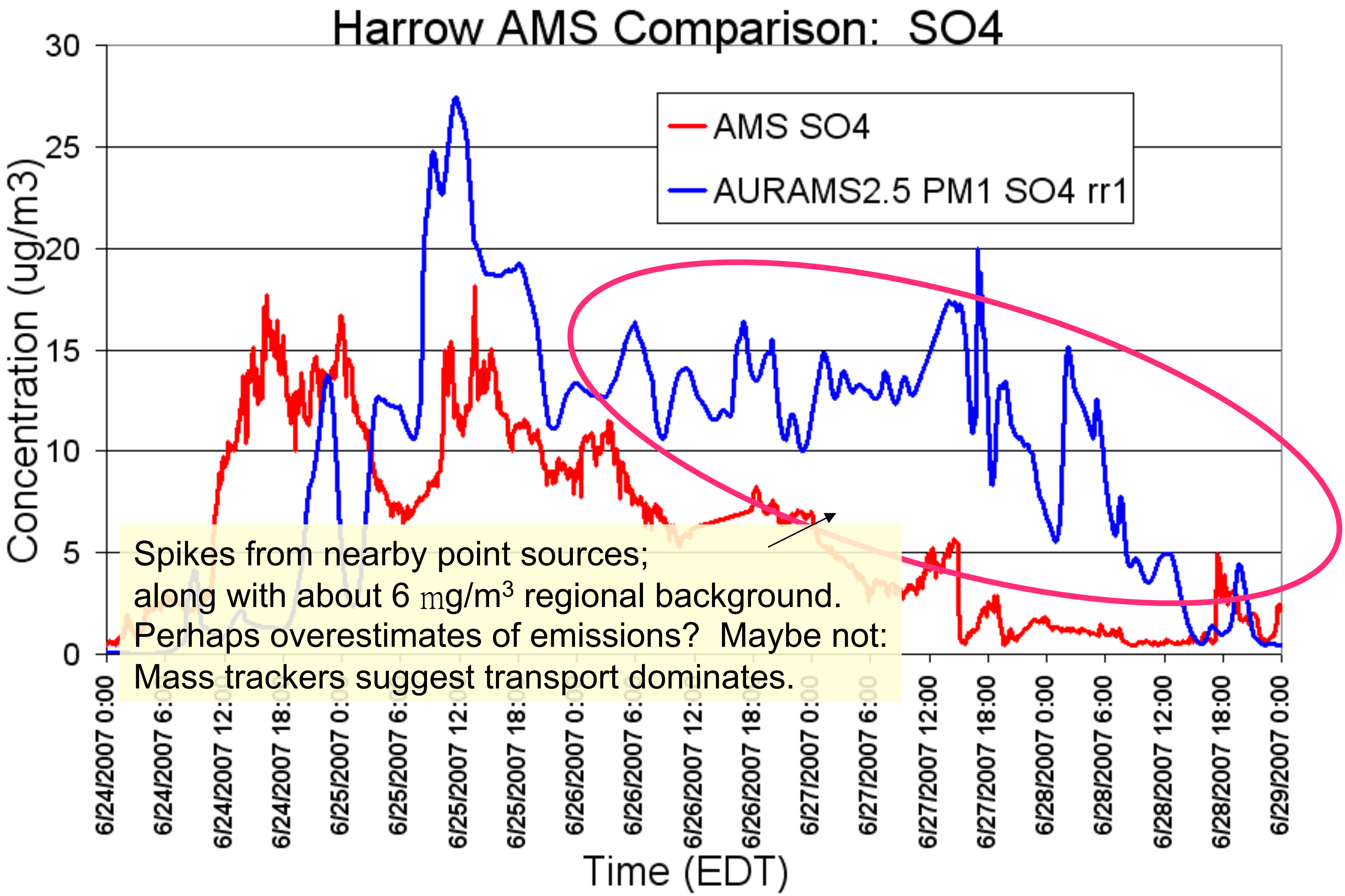




Advection event: point sources from S. Detroit.

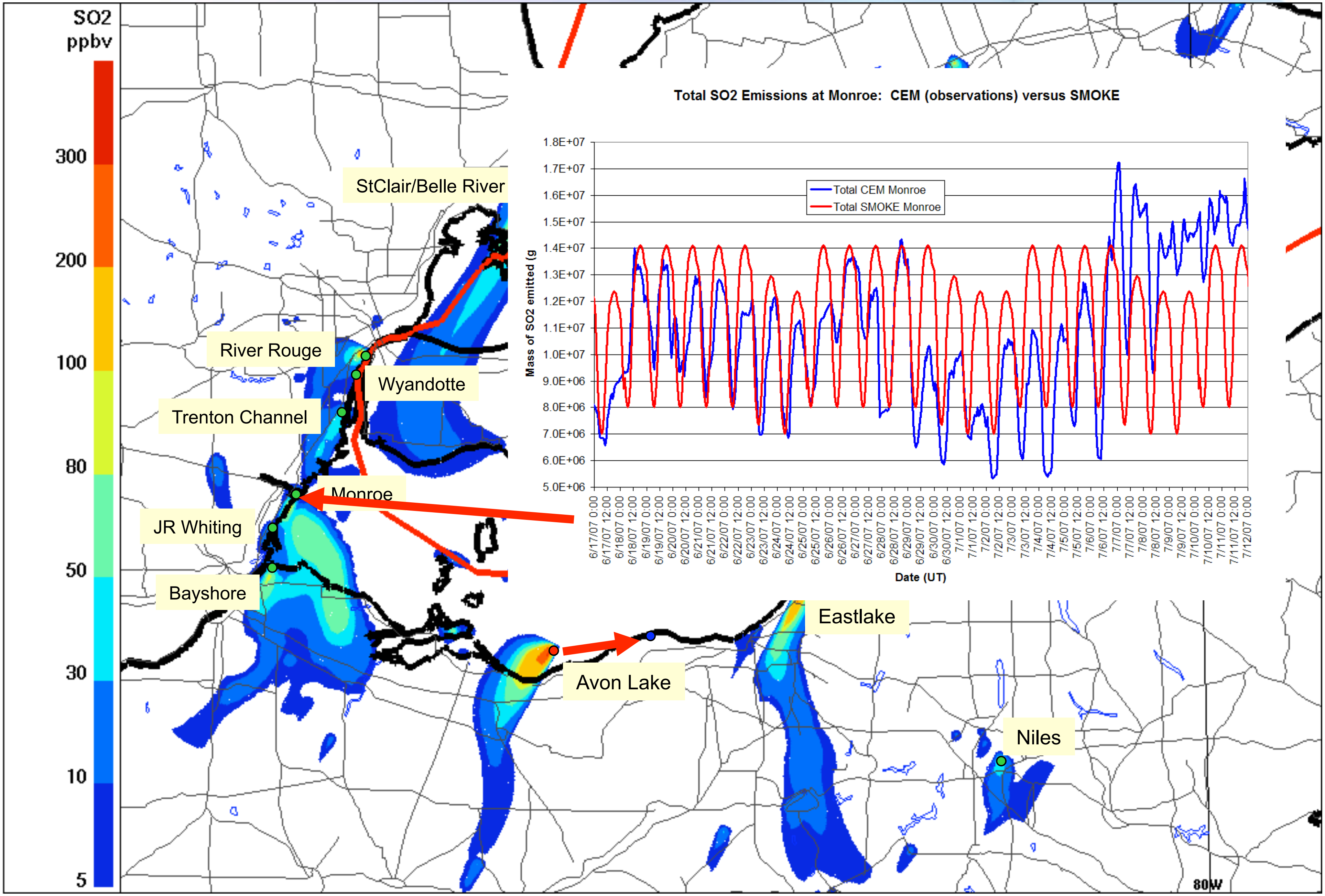






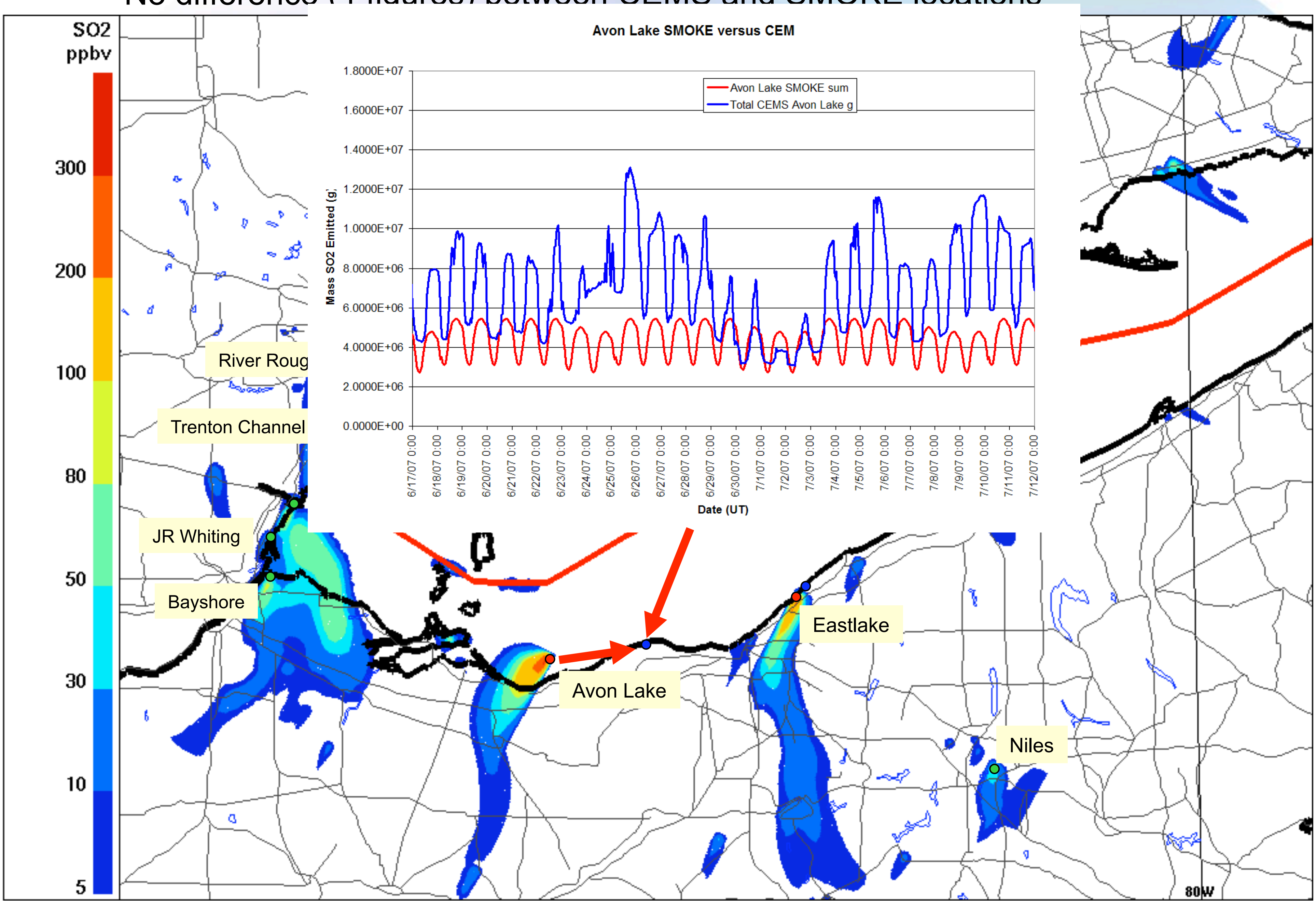
Spikes from nearby point sources;
along with about 6 mg/m^3 regional background.
Perhaps overestimates of emissions? Maybe not:
Mass trackers suggest transport dominates.

SMOKE output site locations ● CEMS (US EPA) site locations ●
No difference (4 figures) between CEMS and SMOKE locations ●

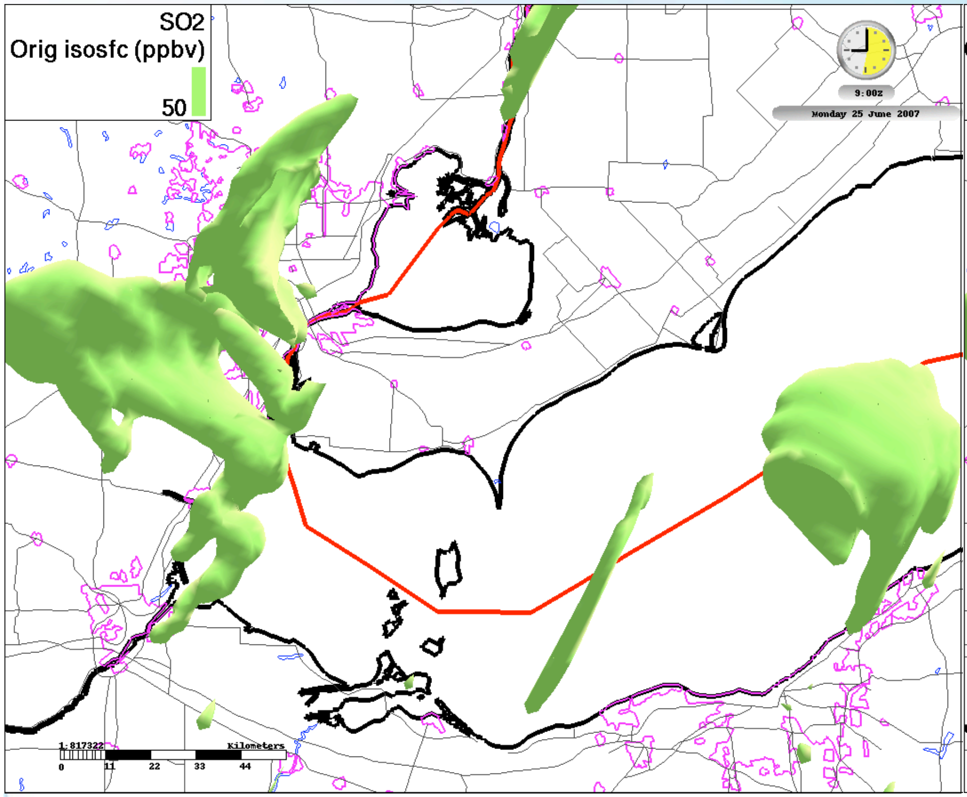


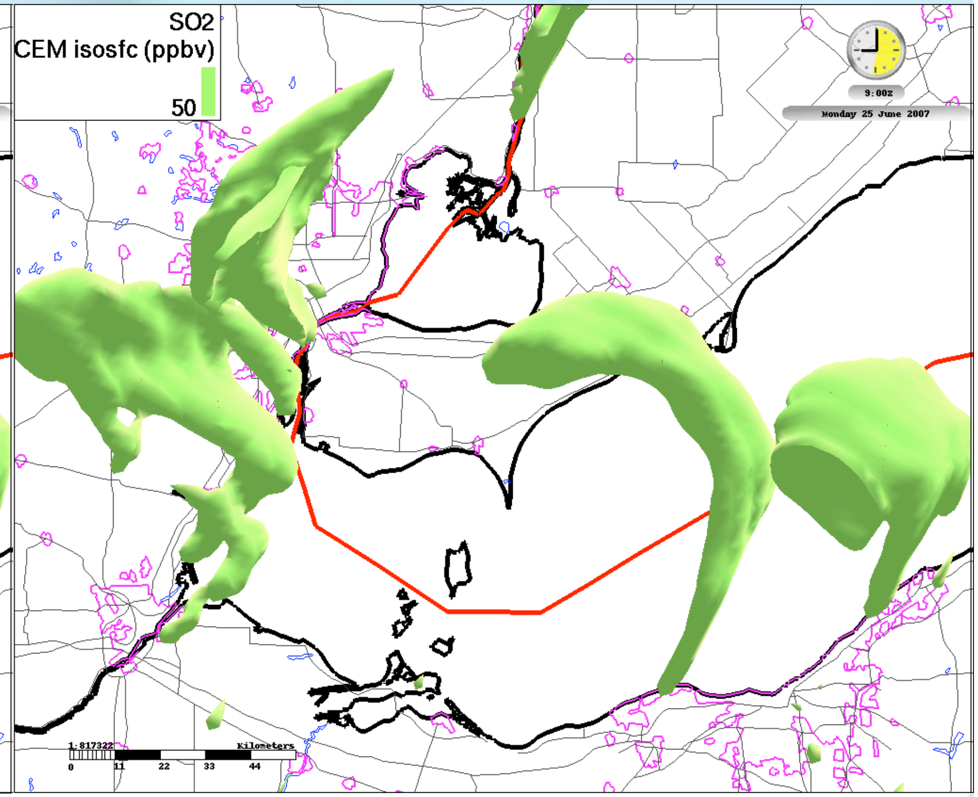
SMOKE output site locations • CEMS (US EPA) site locations •

No difference (4 figures) between CEMS and SMOKE locations •



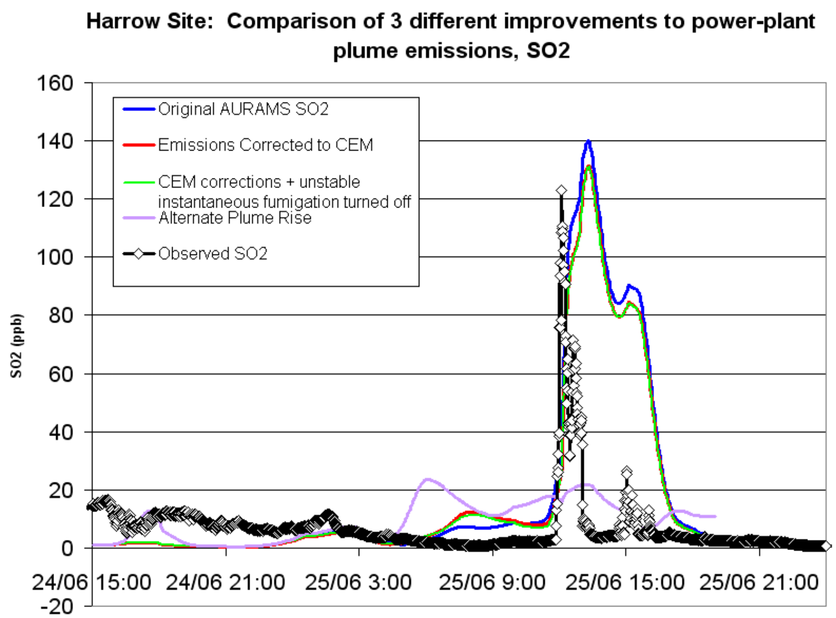
Comparison using a short rerun (24th, 16Z to 26, 0Z): SO_2 50 ppbv isosurfaces

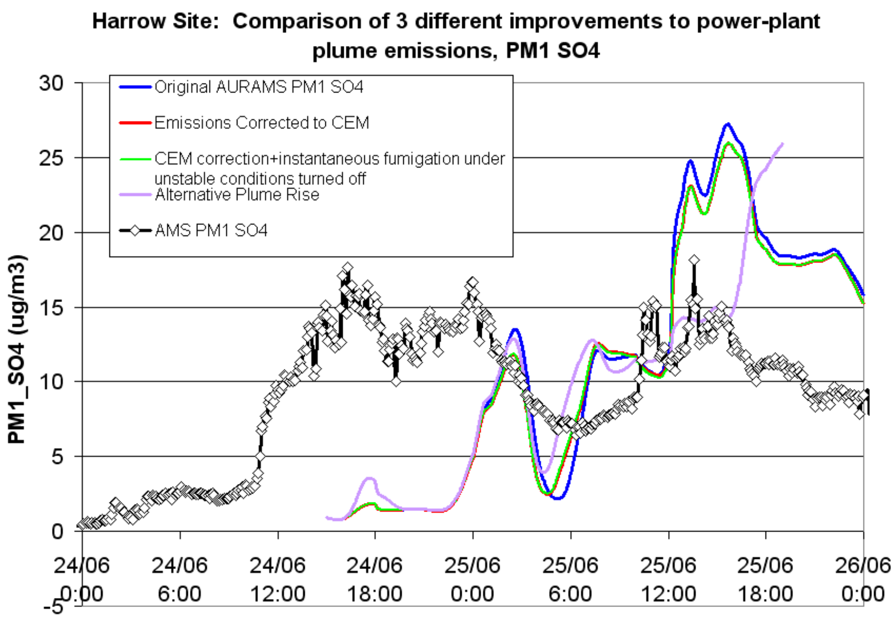




Time series comparison

- Using CEM emissions creates a slight improvement to the SO_2 and PM_1 SO_4.





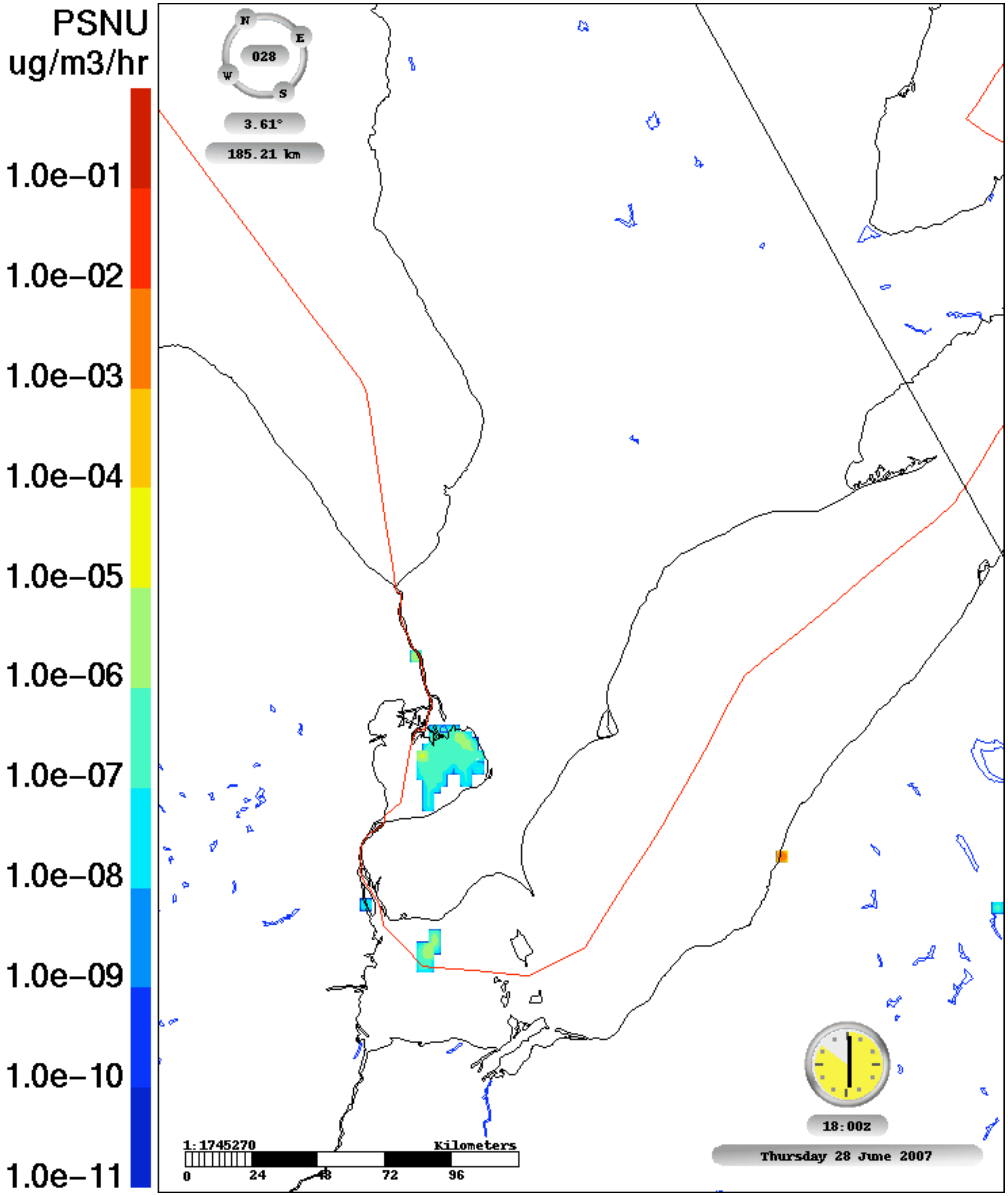
Blue line: SMOKE (emissions processing system) emissions, Red line: corrected to CEM emissions; Green line: instantaneous fumigation turned off; Mauve line: different major point source emissions algorithm.

Plume rise methodology has a bigger impact than correcting the emissions to CEM.
→Does seem to back up mass tracking finding of transport being the most important factor.

Other processes going on in the region

- From the above analysis, there's a lot of interesting "action" happening over the domain, aside from near Harrow or where the aircraft was flying.
- Looking at the mass trackers over the larger domain suggests some interesting things may be taking place.
- A few examples…

Large area extent nucleation events occur over the lakes.



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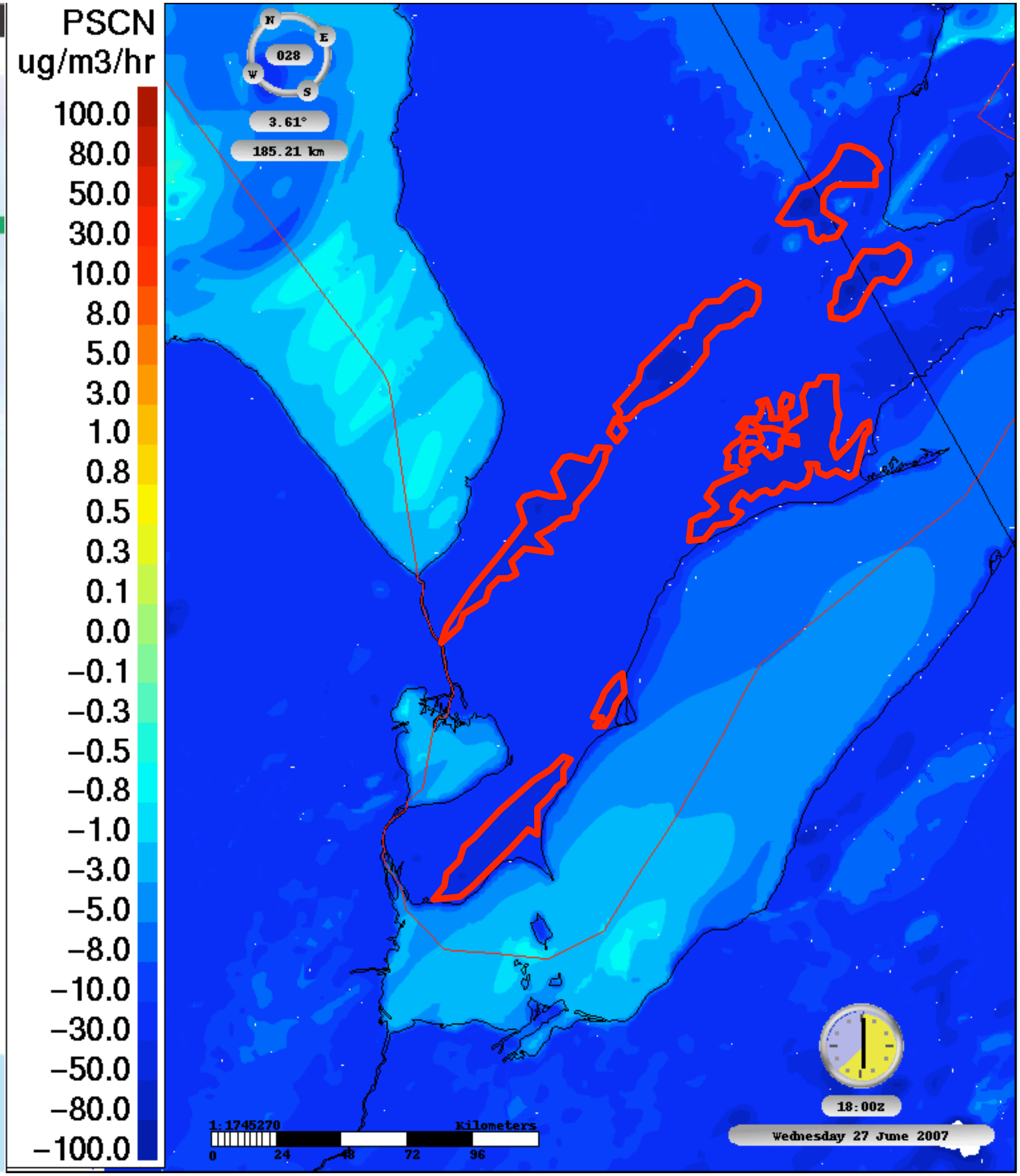
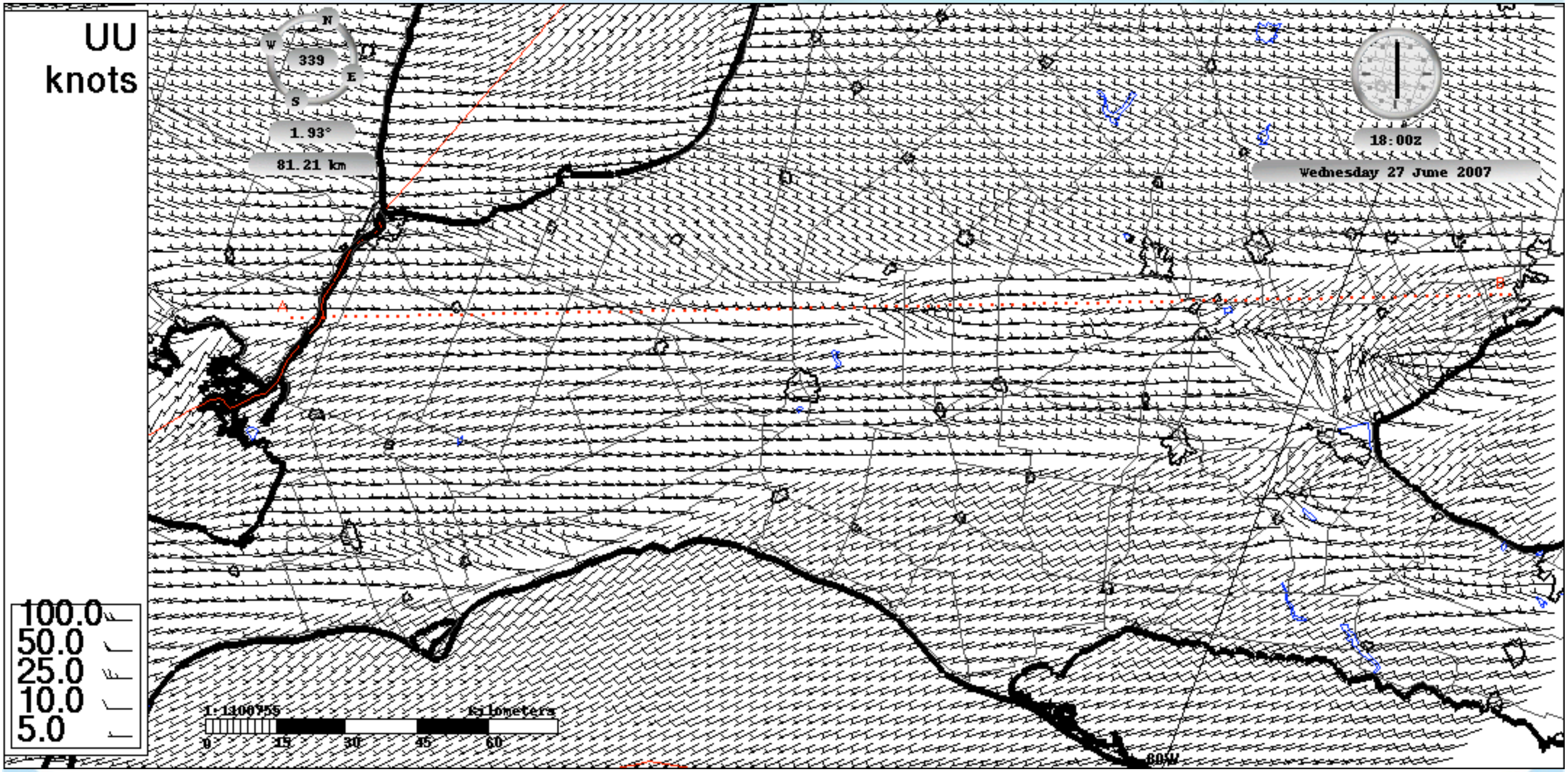
Condensation transferring PM_1 SO_4 mass to larger sizes due to bin transfer, along lake breeze convergence lines.

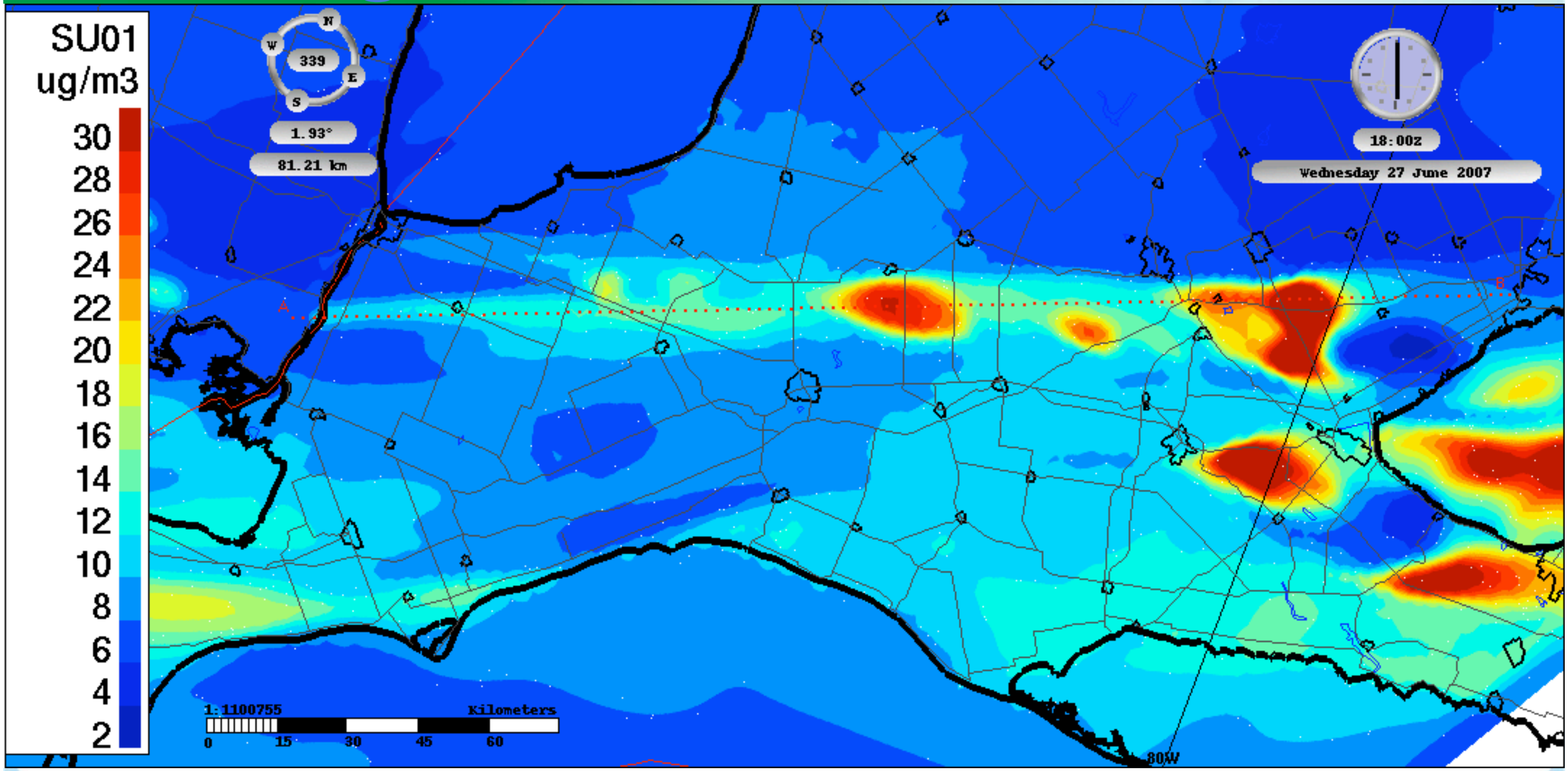
Darkest blue: fastest inter-bin transfer of mass out of PM_1 into larger size bins.



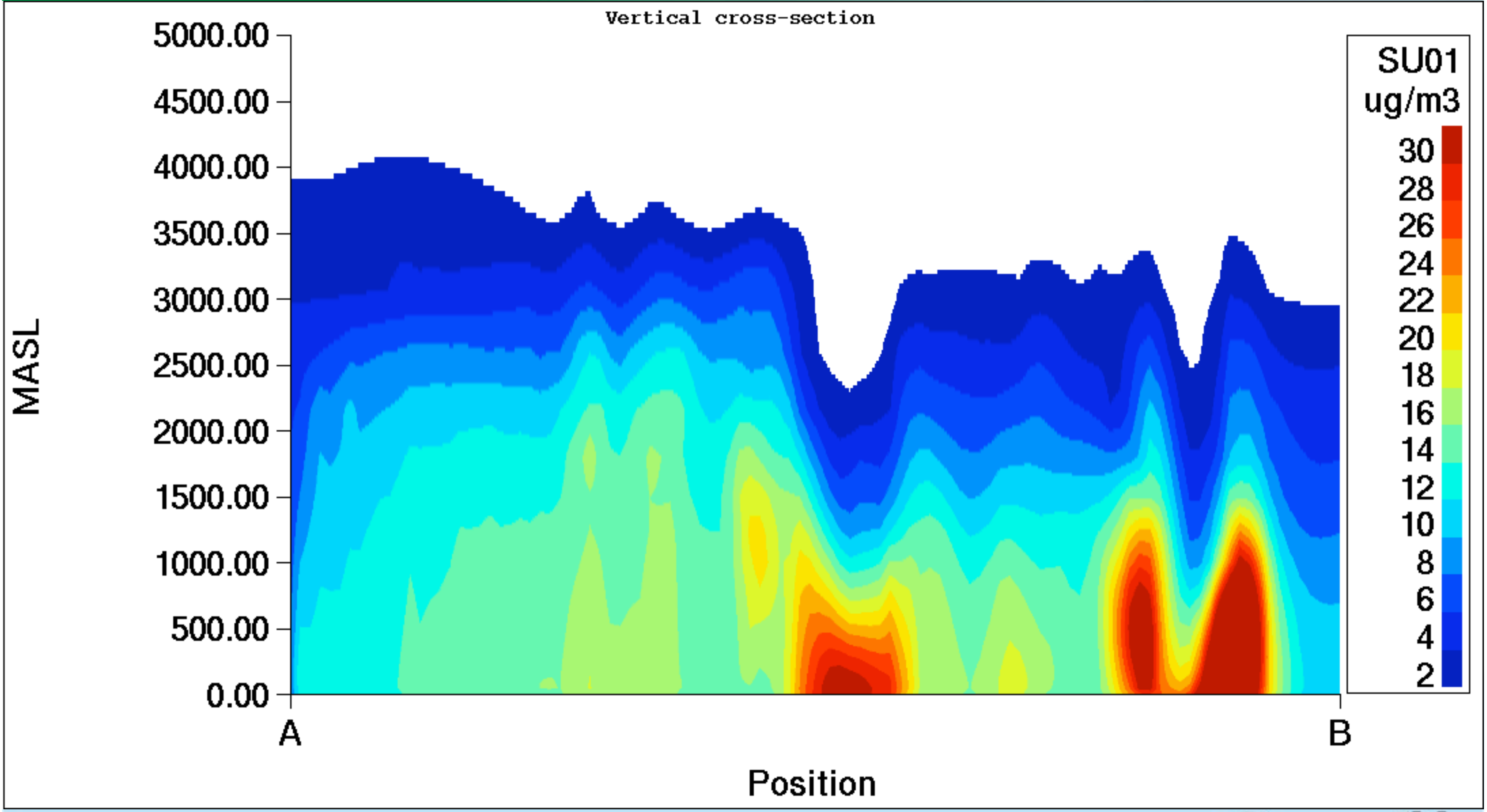
Other processes along that June 27th convergence line…



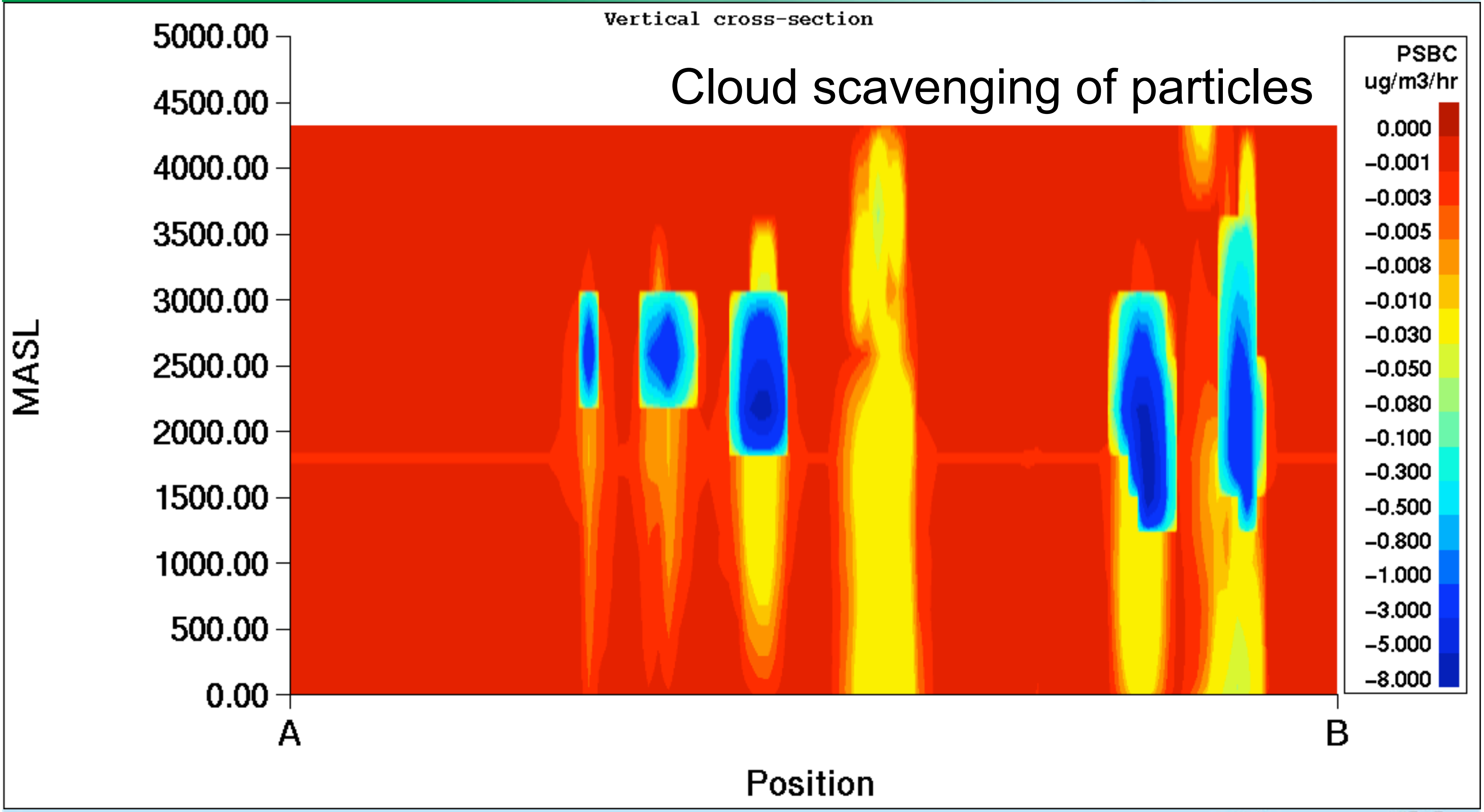
Other processes along that convergence line



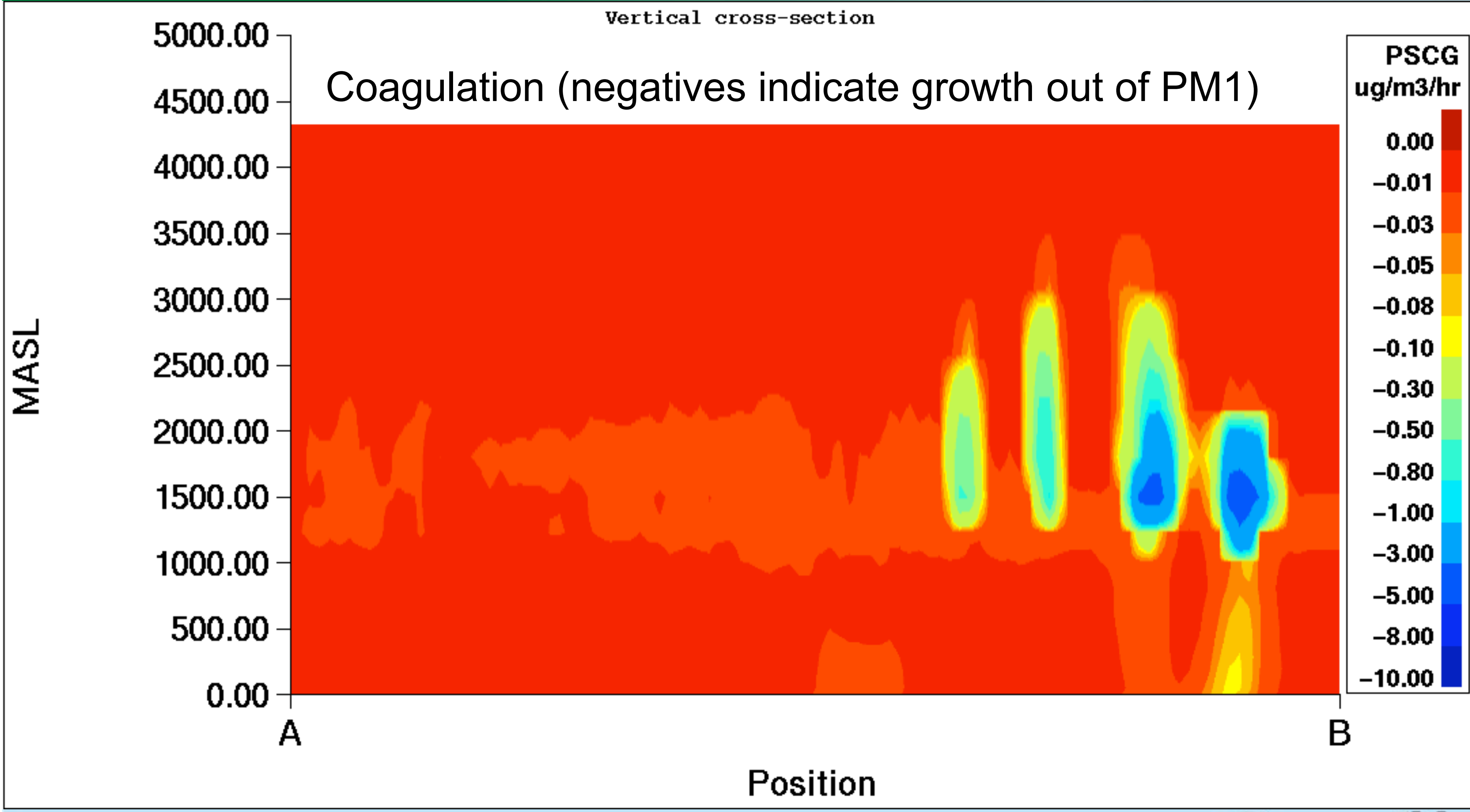
Other processes along that convergence line



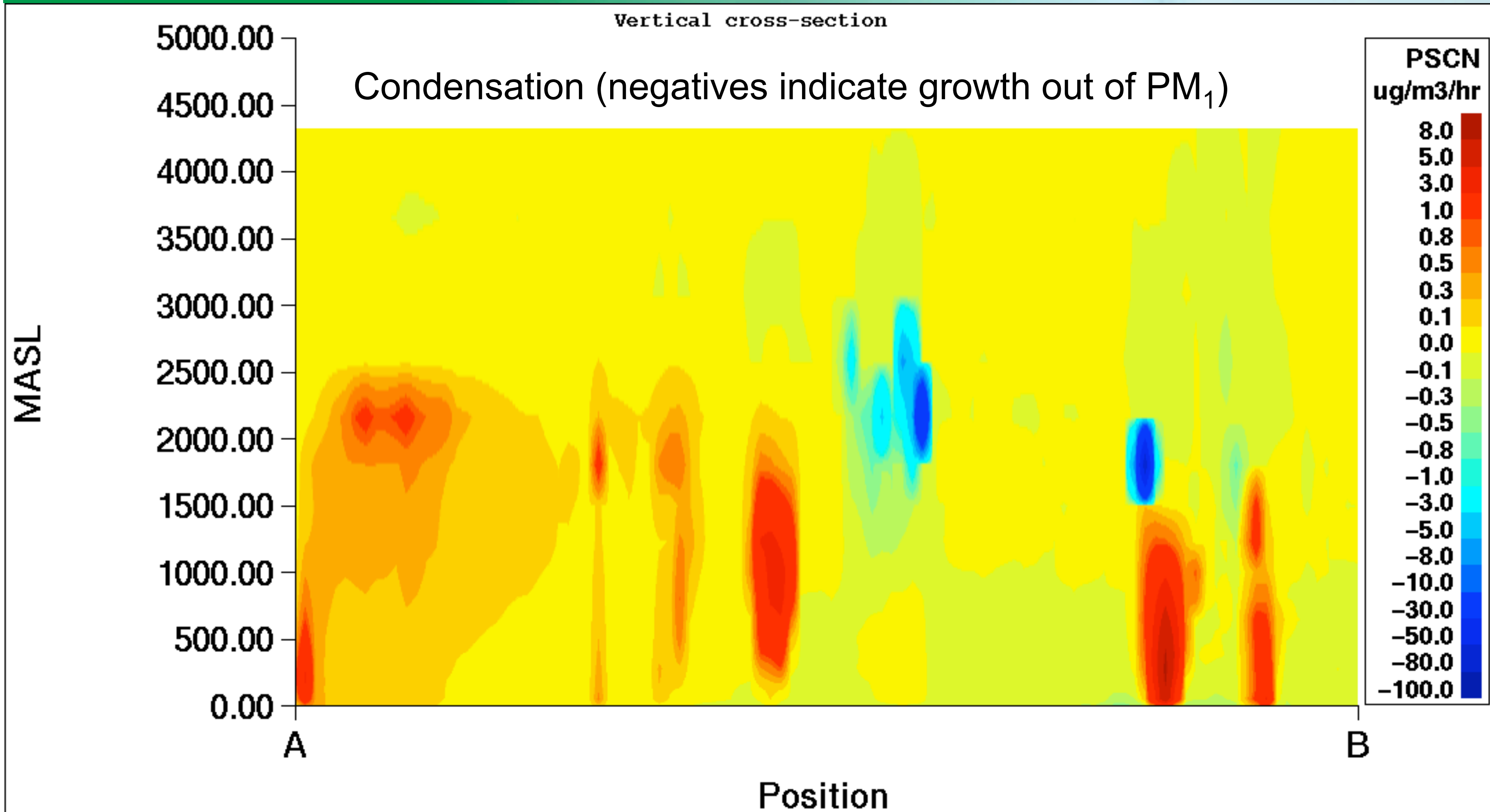
Other processes along that convergence line



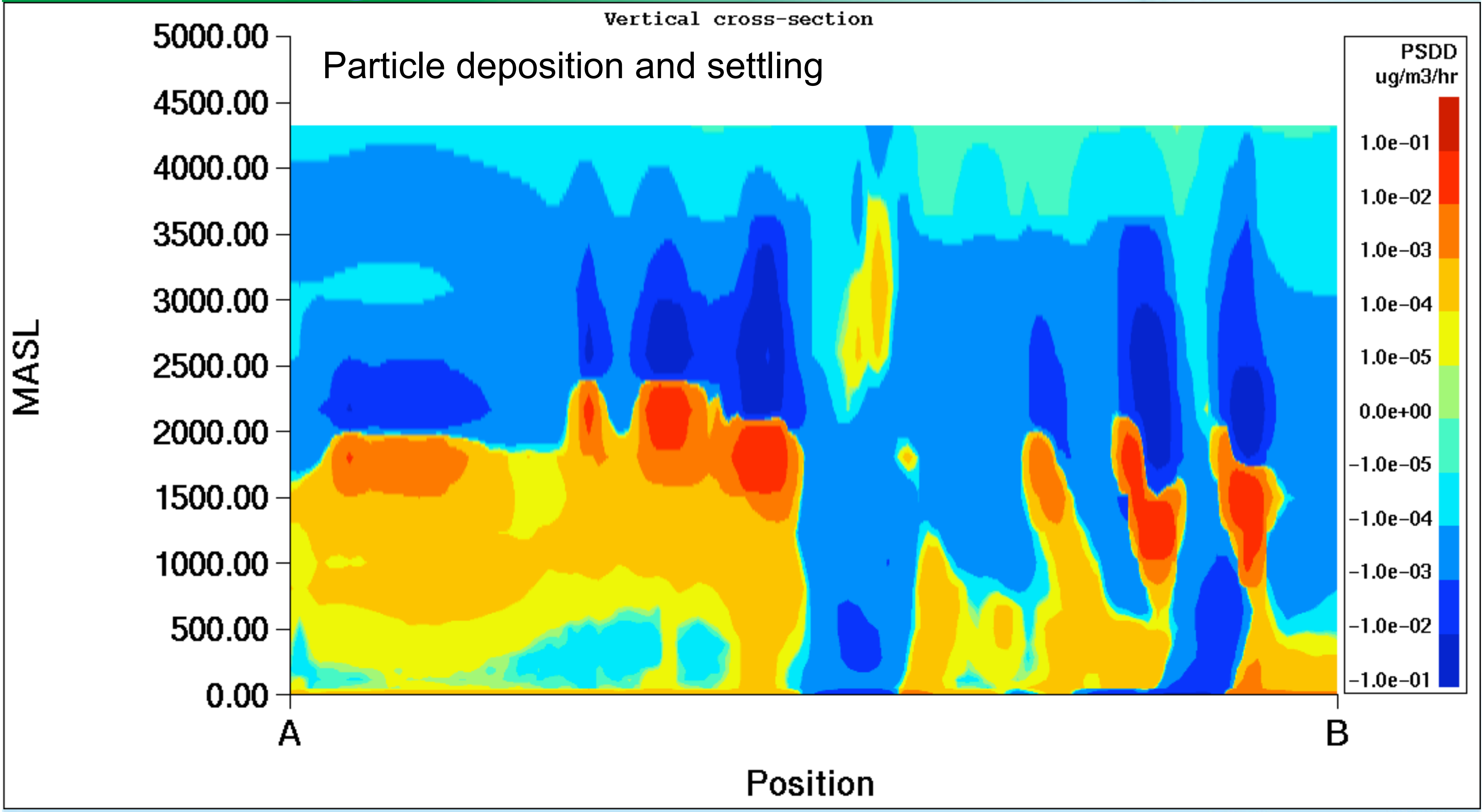
Other processes along that convergence line



Other processes along that convergence line



Other processes along that convergence line



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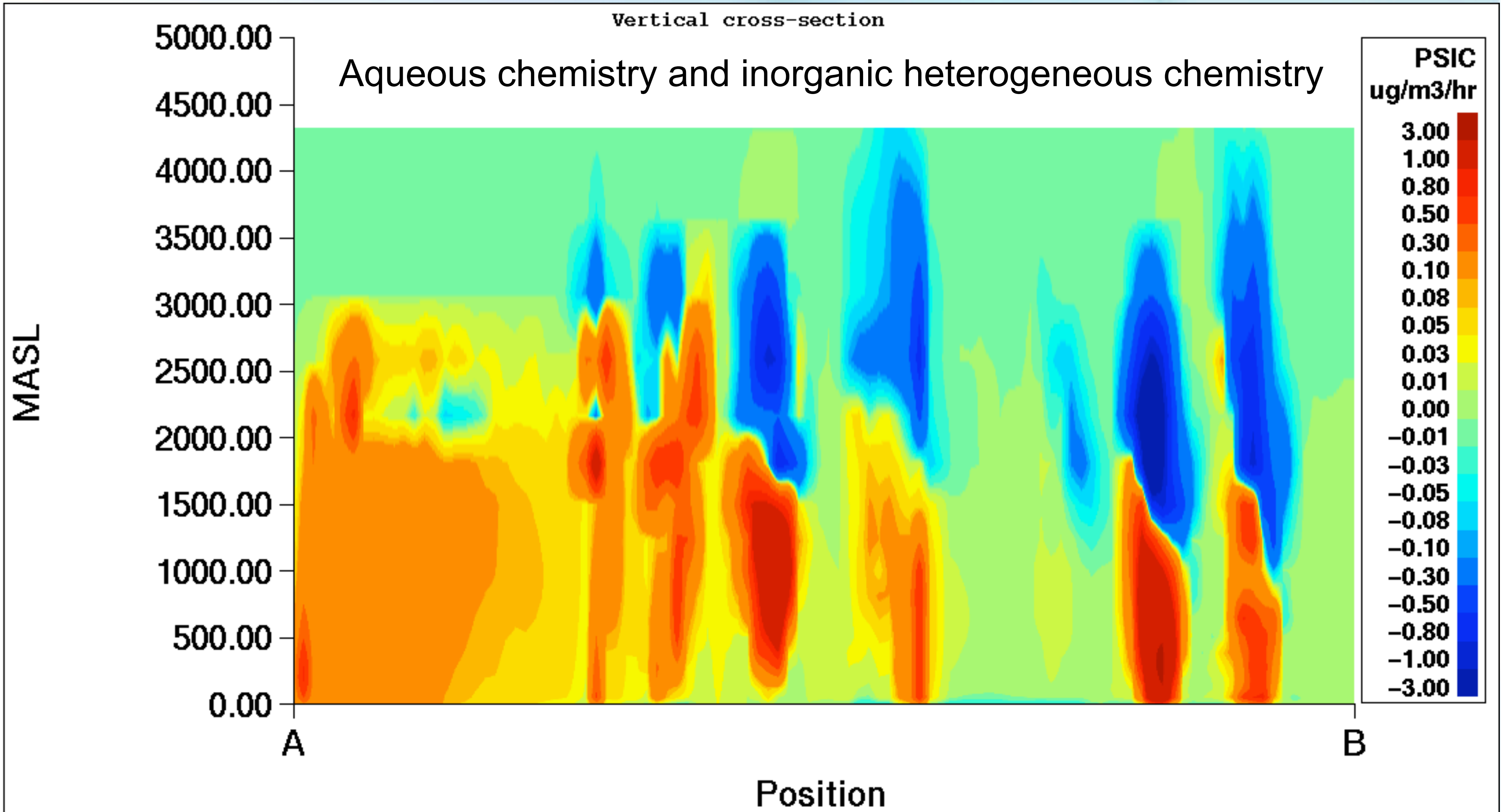

Conclusions

- High spatial and time resolution modelling is *difficult*:
  - Its hard to get the R^2 > .6, slope = 1.0 behavior of the coarser resolution version of the same model (compared to 24 hour averaged, one day in 3 or 6 network data).
  - Small errors in plume placement have a large effect at high resolution!
- Despite that (or bearing that in mind in interpreting the model output), you can learn useful things from the model:
  - Harrow: peaks timed well
  - Harrow: first episode biased high, probably due to major point source south of Detroit.
  - Aircraft: timing can be a few minutes off, and the emissions for a second power plant are likely too high.

  → *Strength of major point sources, and how their emissions are transported, should be re-examined.*

Conclusions

- Local circulation has a big impact on predicted concentrations! Mass tracking suggests that:
  - Cloud processes (rainout, aqueous chemistry) strongest in convective cells "kicked off" by surface-level convergence at lake breeze fronts.
  - Nucleation events strongest over lakes.
  - Fastest condensational transfer of mass from PM_1 to larger sizes occurs along surface frontal convergence lines.
  - Convergence lines "strengthen" many particle formation processes; fastest rates of change in lake-breeze fronts.

→ *Drop by the poster session for more info!*