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

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BAQS-Met: Border Air-Quality Study-Meteorology

BAQS-Met: June 20- July 10, 2007
Point Source Emissions
nominally for the year 2000

NO_x ~ 500K t/y

SO_2 ~ 600K t/y
Creating lake breeze fronts at the surface.

How do these affect the chemistry?

Relatively cold air falls over the lakes...

The local meteorology: synoptic flow interacting with lake breeze fronts.

Creating lake breeze fronts at the surface.

...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/GEM-LAM
(prognostic meteorological model)

GEM-MACH15 Interface

Most of which comes from AURAMS (2 bins instead of 12, emissions module improved relative to AURAMS, inter-bin mass transport improved, advection package in GEM).

GEM’s Physics package

A Unified Regional Air-Quality Modelling System (AURAMS)

- National Emission Inventories (Cdts, U.S.)
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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-evolved 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.

SMOKE
point mobile area biogenies

PM2.5, PM10, etc., Concentration Fields

A sectional model, 12 size bins, 8 particle species.
GEM nested grids for AURAMS input

15 km resolution

2.5 km resolution
AURAMS Nested Modelling Domains

42 km, 15 min. step

15 km, 15 min. step

2.5 km, 2 min. step

All model values shown here are from the high res run.

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

- Palmyra (PAL)
- Croton (CRO)
- Sombra (SOM)
- Lighthouse Cove (LIG)
- Merlin (MER)
- Wheatley (WHE)
- Staples (STA)
- Paquette Corners (PAQ)
- Leamington (LEA)
- Ridgetown
- Bear Creek
- U of Windsor

AMS located here.
Comparison to surface observations: Harrow supersite.

- AURAMS’ PM$_1$-SO$_4$: model values are aggregation of first 6 particle bins + 0.042 of the 7$^{th}$ bin: equivalent to AMS size range.
Comparison to surface obs: Harrow.

Harrow AMS Comparison: PM1-SO4

Two distinct periods of enhanced PM$_1$ SO$_4$

Though the magnitudes of the events are sometimes biased...
Stats (10 minute averages matched)

- R: 0.3964
- Best fit:
  model = 0.5933
  obs + 1.556
- Mean bias:
  \(-3.243 \times 10^{-3} \) \(\text{ug/m}^3\)
- Mean error:
  3.376 \(\text{ug/m}^3\)

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)

Model leads obs by 10 minutes.
Example: Flight 15 (R=0.67)

Model SO$_2$ precursor is biased high.
12/8/09

Flight Model is 12 minutes late.

PM$_1$ SO$_4$

Flight 9 June-26-2007

SO$_4$ (ug/m$^3$)

20:10:00 20:40:00 21:10:00 21:40:00 22:10:00 22:40:00 23:10:00

1st Model peak

Observed peak

2nd Model peak

Model is 12 minutes late.

1st peak in model is not present in the observations. Where is this coming from, in the model?  A major point source (coal-fired power plant) south of Detroit.

2nd peak in model is aged plume from Cleveland.

SO$_2$

Flight 9 June-26-2007

20:10:00 20:40:00 21:10:00 21:40:00 22:10:00 22:40:00 23:10:00

SU01 ug/m$^3$ sfc

T p W thlo o
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 (24$^{th}$ 18:00 EDT – 29$^{th}$ 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.

### AURAMS 2.5km change in PM1 SO4 Mass at Harrow, AURAMS 2.5

<table>
<thead>
<tr>
<th>Process Description</th>
<th>Color</th>
</tr>
</thead>
<tbody>
<tr>
<td>Advection</td>
<td>magenta</td>
</tr>
<tr>
<td>Coagulation</td>
<td>cyan</td>
</tr>
<tr>
<td>Diffusion + Surface Emission</td>
<td>red</td>
</tr>
<tr>
<td>Nucleation</td>
<td>orange</td>
</tr>
<tr>
<td>Condensation</td>
<td>green</td>
</tr>
<tr>
<td>In-Cloud + Inorg. Het. Chem</td>
<td>light green</td>
</tr>
<tr>
<td>Settling + Dry Dep.</td>
<td>blue</td>
</tr>
<tr>
<td>Below Cloud Processes</td>
<td>gray</td>
</tr>
</tbody>
</table>

**Legend**:
- **Red**: Diffusion adds mass.
- **Blue**: Settling + Dry Deposition removes mass.

**Note**: The graph shows a significant increase in PM1 SO4 mass around 6/27/2007, which looks like a Rorschach “ink-blot” test.

**Comment**: Diffusion (red) adds mass, and Settling + Dry Deposition (blue) removes it. Add the two together, and the two are close to being balanced.
What created the (model) PM$_1$ SO$_4$? 

**AURAMS 2.5km change in PM1 SO4 Mass at Harrow, AURAMS 2.5**

- **Advection**
- **Condensation**
- **Nucleation**
- **In-Cloud + Inorg. Het. Chem**
- **Coagulation**
- **Below Cloud Processes**
- **Diff., sfc emis., dry dep., settling**

**Two events where diffusion brings down mass from above, overwhelming dry dep and settling.**

**8 - 9am, 25th**

**Second event is partially offset by advection reducing mass.**
What created the (model) PM$_1$ SO$_4$?

AURAMS 2.5km change in PM1 SO4 Mass at Harrow, AURAMS 2.5

- Nucleation
- Coagulation
- Condensation
- Below Cloud Processes
- Diff+dep+advect
- In-Cloud + Inorg. Het. Chem
What created the (model) PM$_{1}$ SO$_4$?

AURAMS 2.5km change in PM1 SO4 Mass at Harrow, AURAMS 2.5
What created the (model) PM$_1$ SO$_4$?

AURAMS 2.5km change in PM1 SO4 Mass at Harrow, AURAMS 2.5
What created the (model) PM$_1$ SO$_4$?

Observations courtesy Greg Evans, Cheol-Heon Geong, U of T.
What created the (model) PM$_2.5$ SO$_4$?

Harrow AURAMS2.5km PM1 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.
Increases on the 26th 22Z and thereafter: Detroit / Windsor; local emissions close to the measurement site.
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/m³ 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

Bayshore
Avon Lake
Eastlake
Niles
Monroe
Trenton Channel
StClair/Belle River
River Rouge
Wyandotte
JR Whiting
Bayshore
Avon Lake
Eastlake
Niles
SMOKE output site locations • CEMS (US EPA) site locations •
No difference (4 figures) between CEMS and SMOKE locations •

Avon Lake SMOKE versus CEM

Date (UT)

Mass SO2 Emitted (g)

Avon Lake SMOKE sum
Total CEMS Avon Lake g
Comparison using a short rerun (24\textsuperscript{th}, 16Z to 26, 0Z): \(\text{SO}_2\) 50 ppbv isosurfaces
Time series comparison

- Using CEM emissions creates a slight improvement to the SO$_2$ and PM$_{10}$ SO$_4$.

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.
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

Cloud scavenging of particles

Vertical cross-section

PSBC
ug/m³/hr

0.000
-0.001
-0.003
-0.005
-0.008
-0.010
-0.030
-0.050
-0.300
-0.800
-1.000
-3.000
-5.000
-8.000
Other processes along that convergence line

Coagulation (negatives indicate growth out of PM1)
Other processes along that convergence line

Condensation (negatives indicate growth out of PM$_1$)
Other processes along that convergence line

Particle deposition and settling
Other processes along that convergence line

Aqueous chemistry and inorganic heterogeneous chemistry
Conclusions

• High spatial and time resolution modelling is difficult:
  – It’s hard to get the $R^2 > 0.6$, 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!