Process Analysis Evaluation of Global and Regional Ozone Models

Identifying Sources of Uncertainty

Barron H. Henderson; Joseph P. Pinto; Chris Emery

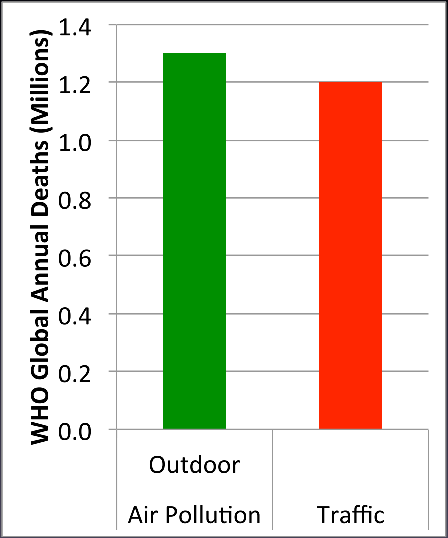
University of Florida

2013-10-30

Summary

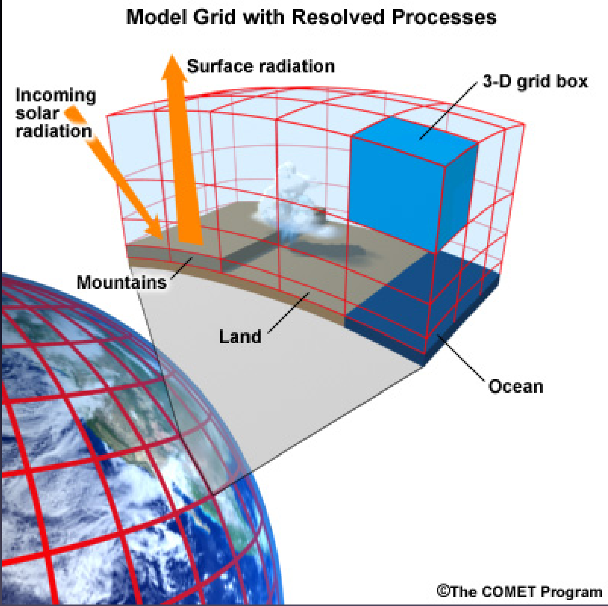
- As Oreskes (1994) and later Beck (2002) have demonstrated, atmospheric models are "open systems" that have "essentially unknowable" inputs
- Can have a wide variety of inputs
  - Generated by different groups Minimum level of detail
  - Come from models with their own uncertainty
- Easily suffer from compensating errors
- Getting the "right answer" for the "wrong reasons"
- Model Performance Evaluations look at the result
- Process Analysis examines the processes
  - that are typically lost
  - useful in identifying important processes
  - useful to constraining development

Air pollution kills people

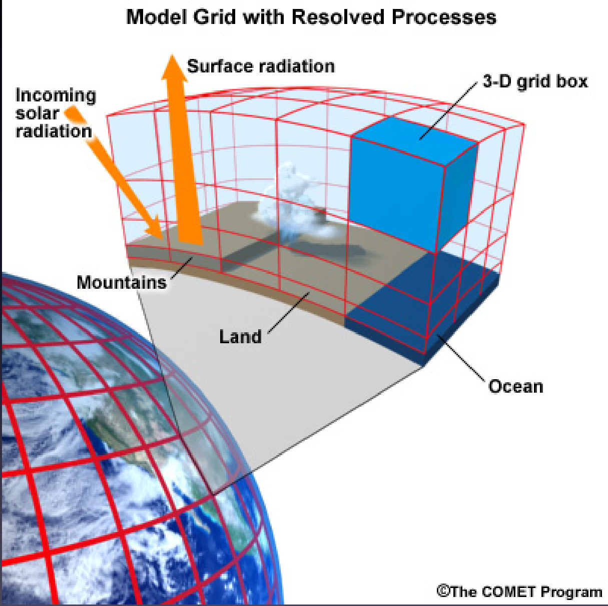


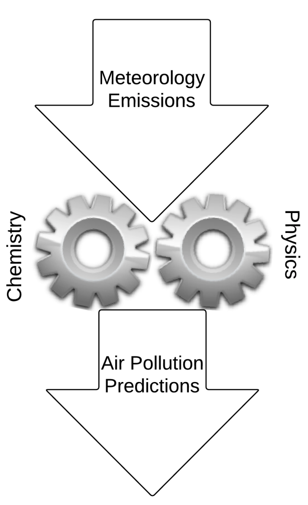
- Data from the World Health Organization
- Internationally: Easy to accept – developing countries have more pollution.
- Developed world too – "Four times more people die in the San Joaquin Valley from air pollution than they do from traffic fatalities." – Jared Blumenfeld, EPA Regional Administrator
- IARC classifies air as a carcinogen

We simplify the real world for solution

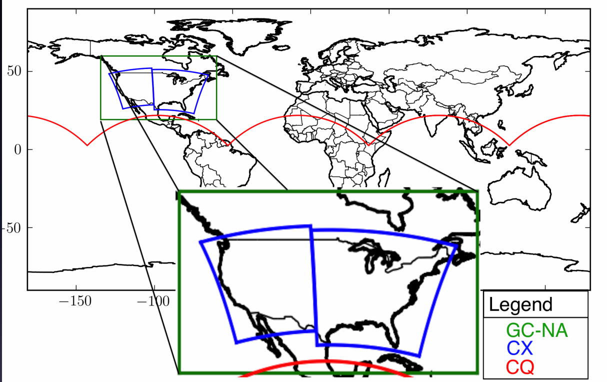


We simplify the real world for solution



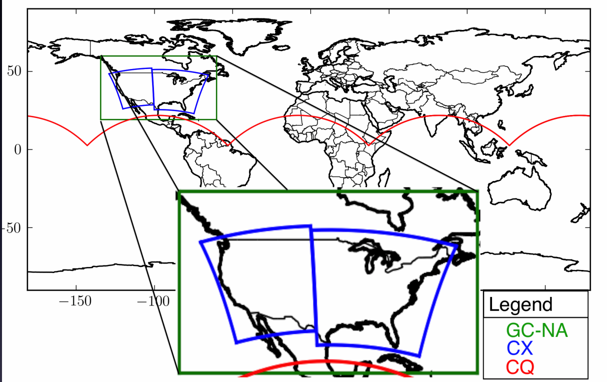


We simplify the real world for solution



- Several modeling systems simulated the globe or northern hemisphere (Zhang et al. 2011; Emery et al. 2012; Mathur unpublished.)

We simplify the real world for solution



- Several modeling systems simulated the globe or northern hemisphere (Zhang et al. 2011; Emery et al. 2012; Mathur unpublished.)
- Evaluation looked only at the continental United States

Models generally agreed

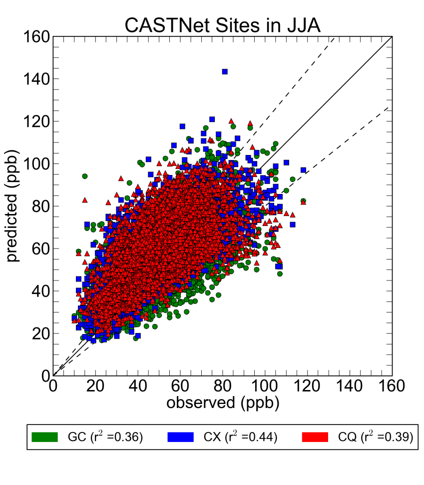


Figure 1 : Time paired predictions vs observations

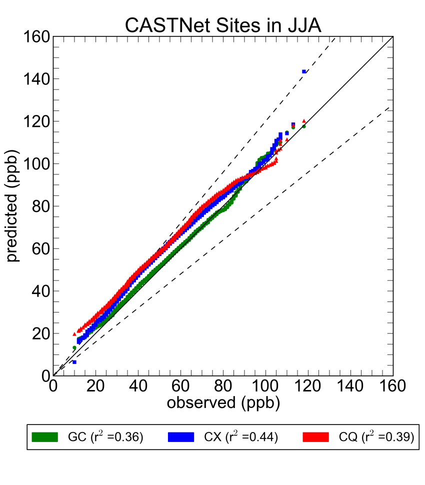


Figure 2 : Rank paired predictions vs observations

Models generally agreed mostly

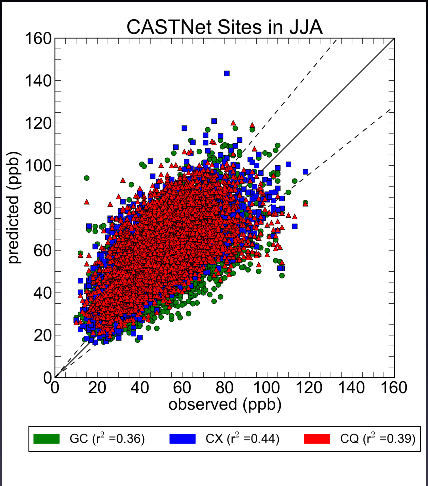


Figure 1 : Time paired predictions vs observations

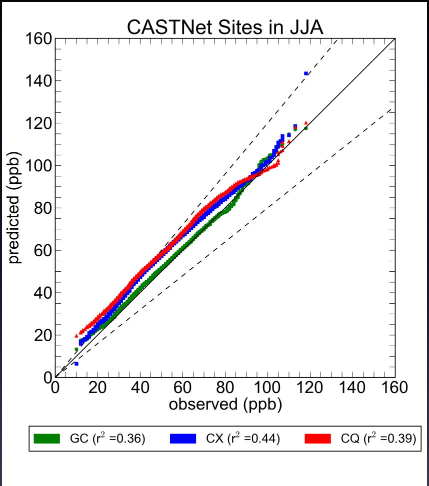
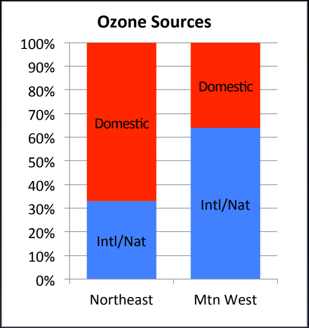


Figure 2 : Rank paired predictions vs observations

Where did it come frome?



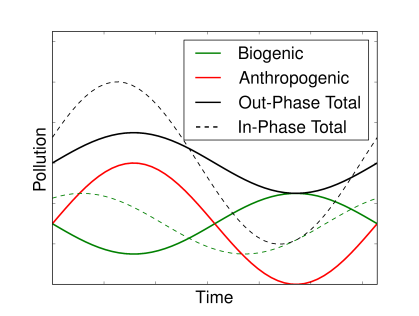


Figure 3 : Conceptual contributions of “background” air to total.

Figure 4 : Conceptual anthropogenic (red) and biogenic (green) contributions: in-phase contributions (dashed); out-of-phase (solid).

- Conceptually we can ask, do the models agree on how much biogenic emissions contribute to total ozone?
- Mostly, Henderson et al., 2012

What differences are there?

Problems: Compensating isoprene nitrate issue led to

1. Better rank-paired performance for GEOS-Chem compared to CAMx in the East
2. Less correlation between background and total
3. Still other differences.

Two options:

1. Wait for results to disagree and diagnose the problem then?
2. Systematically compare processes

Other inter-comparisons show differences and call for details

1. AQMEII Phase II calls for process-based comparisons
2. HTAP shows model differences

Process Analysis Overview

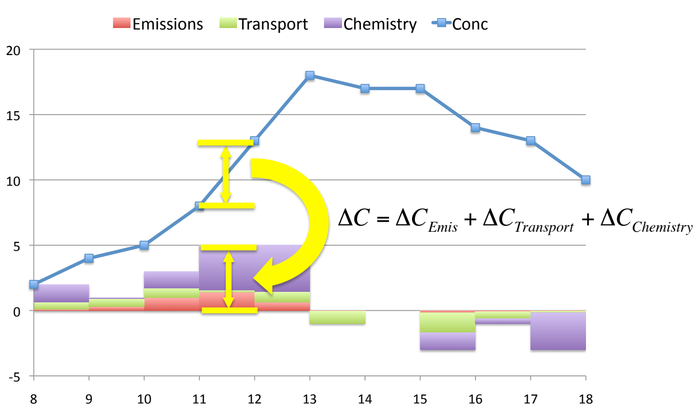


Figure 5 : Conceptual photochemical day

Process Analysis Overview

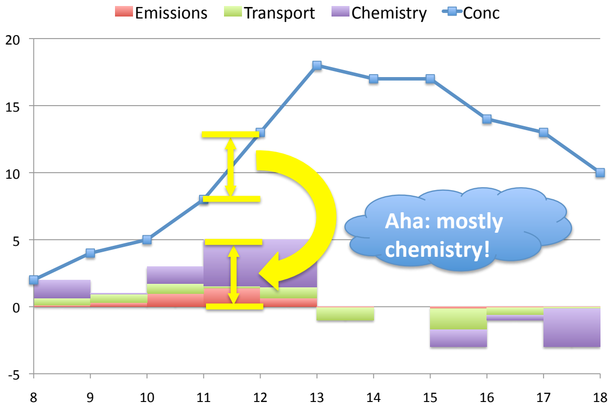


Figure 5 : Conceptual photochemical day

Process Analysis Overview

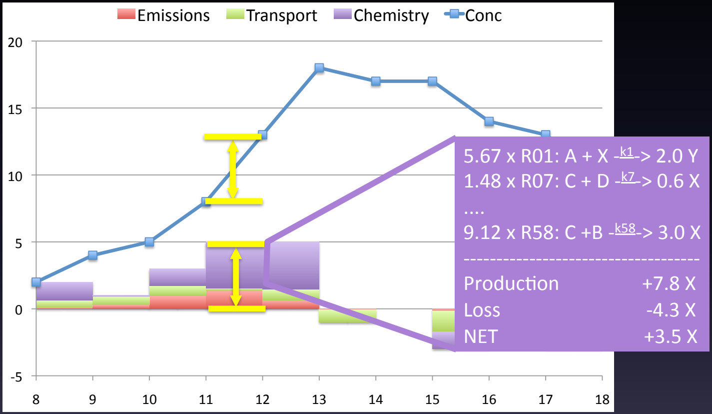


Figure 5 : Conceptual photochemical day

Implementation: Integrated Process Rates

Implemented:

- Transport
- Convective Mixing
- Wet deposition
- Emissions[a]
- Dry deposition[a]
- Heterogeneous Chemistry
- Gas-phase Chemistry

---

[a]Gas-phase emissions and dry deposition are solved either in the chemical solver or in asymmetric convection routines. Separation within convection has not yet been implemented.

Implementation: Integrated Process Rates

Implemented:

- Transport
- Convective Mixing
- Wet deposition
- Emissions[a]
- Dry deposition[a]
- Heterogeneous Chemistry
- Gas-phase Chemistry

[a]Gas-phase emissions and dry deposition are solved either in the chemical solver or in asymmetric convection routines. Separation within convection has not yet been implemented.

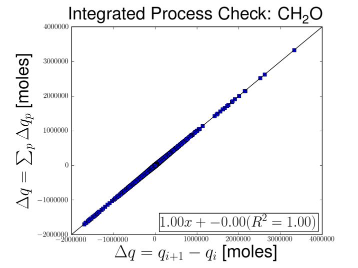


Figure 6 : Illustrative processes summation check where *p* is chemistry, transport, deposition, etc. Formaldehyde process sum compared to instantaneous species change in moles.

Implementation: Integrated Reaction Rates

- Sparse Matrix Vector Gear: available
- Kinetic Pre-Processor
  - Rosenbrock (coming soon)
  - LSODES (available)

Implementation: Integrated Reaction Rates

- Sparse Matrix Vector Gear: available
- Kinetic Pre-Processor
  - Rosenbrock (coming soon)
  - LSODES (available)

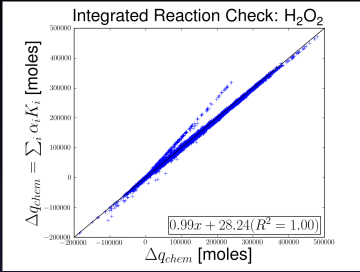


Figure 7 : Illustrative reaction sum check. Hydrogen peroxide sum of reactions vs chemistry process.

$$\Delta q_{chem} = \sum_i \left(\alpha_i k_i \prod_j [Rct]_j \right) \quad (1)$$

Implementation: Integrated Reaction Rates

- Sparse Matrix Vector Gear: available
- Kinetic Pre-Processor
  - Rosenbrock (coming soon)
  - LSODES (available)
- To do: incorporate species specific error correction from SMV-Gear or switch to Rosenbrock

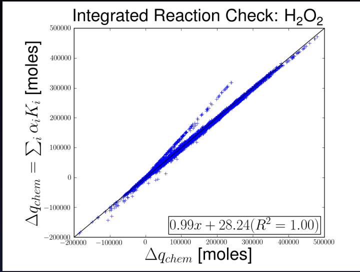


Figure 7 : Illustrative reaction sum check. Hydrogen peroxide sum of reactions vs chemistry process.

$$\Delta q_{chem} = \sum_i \left(\alpha_i k_i \prod_j [Rct]_j \right) \quad (1)$$

Allows for development of chemical indicators

- Sillman Ratio (eq 3): Ratio of radical losses (L) via non-nitrogen pathways to nitrogen pathways
  - In approximation, greater than 0.35 is NO_x limited

$$\frac{L}{L_{\mathrm{NO_x}}} - 1 = \frac{L_{\mathrm{HO_x}}}{L_{\mathrm{NO_x}}} \approx \frac{P(\mathrm{H_2O_2})}{P(\mathrm{HNO_3})} \quad (2)$$

- We can use processes or these chemical metrics to identify regions of interest for further study.

Isoprene emissions

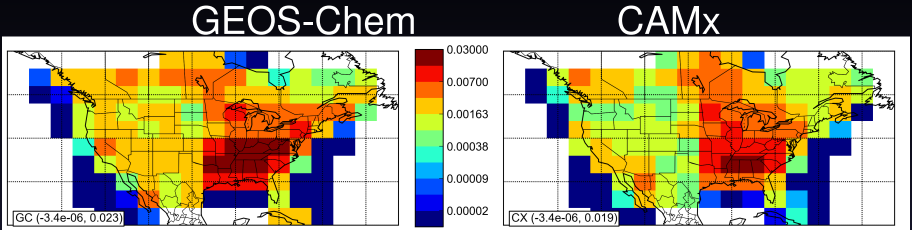


Figure 8 : GEOS-Chem (left) and CAMx (right) isoprene emissions integrated throughout their planetary boundary layers.

- GEOS-Chem's isoprene emissions are higher than CAMx's in the east
  - Consistent with Carlton and Baker ES&T 2011
  - GEOS-Chem uses MEGAN which emits more isoprene than BEIS, which was used by CAMx

Sillman Ratio

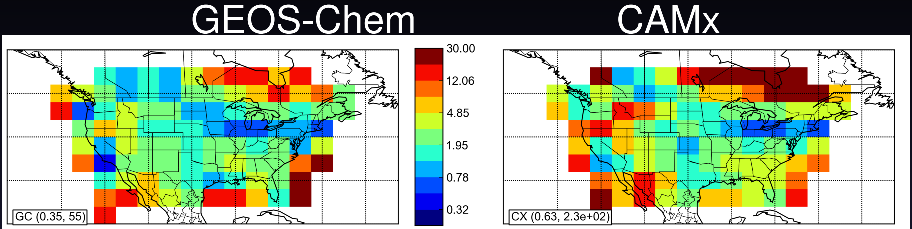


Figure 9 : GEOS-Chem (left) and CAMx (right) SILLMAN integrated throughout their planetary boundary layers.

- Many of the same features as seen in OPE
- Along US northern boundary, differences are more clear
- Recall that >0.35 is NO_x sensitive: the differences here are shades of NO_x limited

Focus Area

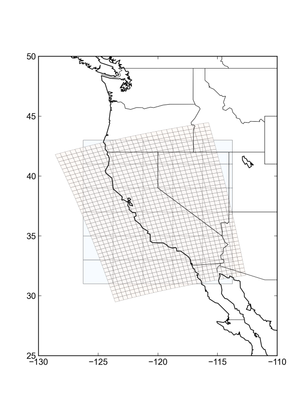
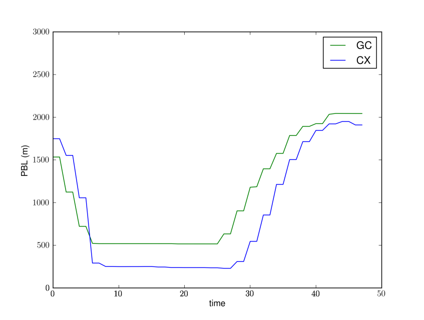


Figure 10 : Region for further analysis.

- Focus area selected for:
  - Isoprene emission discrepancy
  - Previous findings that western bias may be attributable to BC
  - Large populations exposed on the western seaboard

Planetary Boundary Layer Height (focus)



- Note the earlier rise and lower peak for GEOS-Chem
- The CAMx PBL is diagnosed from vertical diffusivity using ENVIRON's vertavg algorithm.

Figure 11 : GEOS-Chem (green) and CAMx (blue) planetary boundary layer height averaged within the focus area.

ISOP time series

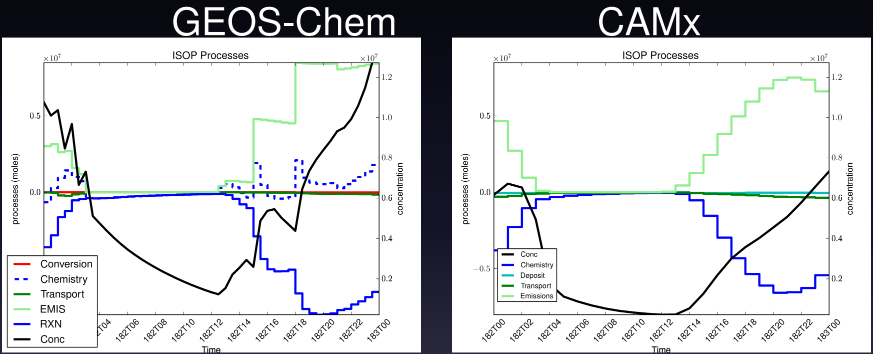


Figure 12 : Time series ISOP plots for GEOS-Chem (left) and CAMx (right) on 2006-07-01 for the focus area.

- Note: The time step associated with averaged met/biogenics.

NO_x time series

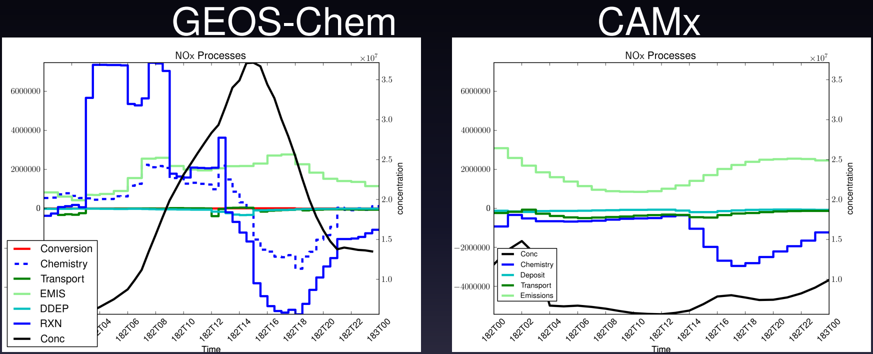
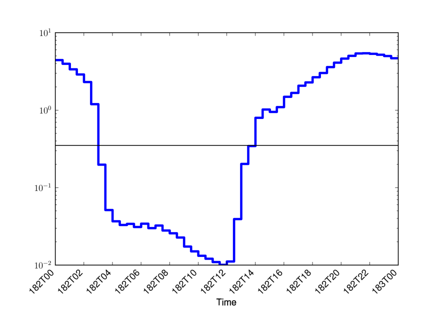


Figure 13 : Time series NO_x plots for GEOS-Chem (left) and CAMx (right) on 2006-07-01 for the focus area.

- Huh?

Sillman ratio time series

GEOS-Chem



CAMx

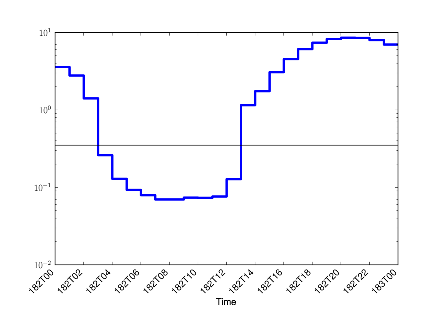


Figure 14 : Time series sillman ratio plots for GEOS-Chem (left) and CAMx (right) on 20060701 for the focus area. The black line marks the NO_x sensitive transition

- Note: timing of increasing OPE differs between the two models
- Could be an artifact of the timing and extent of the PBL rise

Adv. Sillman ratio time series

GEOS-Chem CAMx

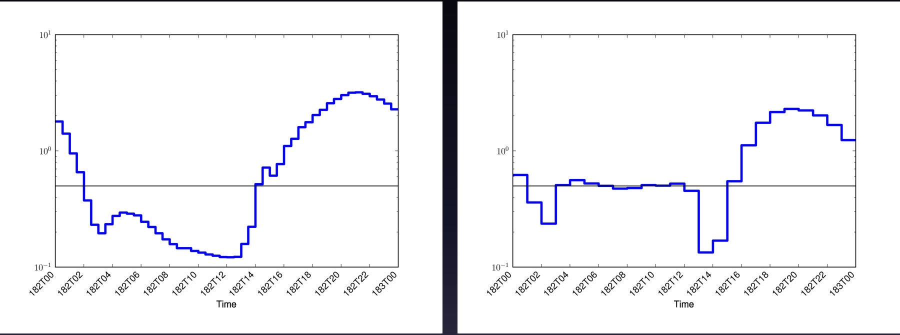


Figure 15 : Time series sillman ratio plots for GEOS-Chem (left) and CAMx (right) on 20060701 for the focus area. The black line marks the NO_x sensitive transition

- Woah! Warning, still very preliminary resutls.

Conclusions and Disclaimer

1. These results are still under development and may change
    1. Post processing of PBL should be further reviewed
    2. Night-time IRR may not be sampled correctly
        1. Seems to only effect photolysis IRR, which are not in Sillman ratios
        2. But could affect advanced Sillman ratio
2. Even so, we see interesting patterns that are likely robust
    1. Explainable given known differences in wildfires and emission inventories
    2. These results were also consistent with a fixed-top analysis (0-1km AGL; not shown)

Future work

1. Process-based Analysis allow for:
    1. rapid identification of model discrepancies
    2. rapid model development
2. As researchers implement regional scale models for new territories, comparison with established global models provides:
    1. process-level benchmarks for the regional scale models
    2. a means of providing feedback to the global scale model about processes that need updating

Acknowledgements

- US EPA OAQPS: Farhan Akhtar, Heather Simon, Norm Possiel
- US EPA NCEA: Joseph P Pinto
- ENVIRON: Chris Emery
- Peking University: Lin Zhang
- Supported in part by (1) an appointment to the Research Participation Program at EPA/NERL administered by the Oak Ridge Institute for Science and Education; (2) startup at the University of Florida

Chemistry and Physics Options

	CX[1]	CQ[2]	GC-NA[3]	GC[4]
Model	CAMx	Hemispheric CMAQ	Nested North America GEOS-Chem	GEOS-Chem
Resolution	12 x 12km	108 x 108km	1/2° x 1/3°	4° x 5°
Meteorology	WRF		GEOS5	
Chemistry	Carbon Bond[5]		8-02-03	9-01-03[6]
Boundaries	GC 2° x 2.5°	N/A	GC 2° x 2.5°	N/A
Biogenic	BEIS		MEGAN	
Lightning	Scaled with Koo profile	N/A	LTDIS scaled with Pickering 1997 profile	
Wildfires	SmartFire daily	N/A	GFED monthly average	

1) Emery et al. AE 2012; 2) Simulations in development; 3) Zhang et al. JGR 2011; 4) Developed for this work; 5) Hemispheric CMAQ nitrates updated to account for isoprene nitrates; 6) Updates in chemistry will decrease NOx loss to isoprene nitrates

Planetary Boundary Layer Height (spatial)

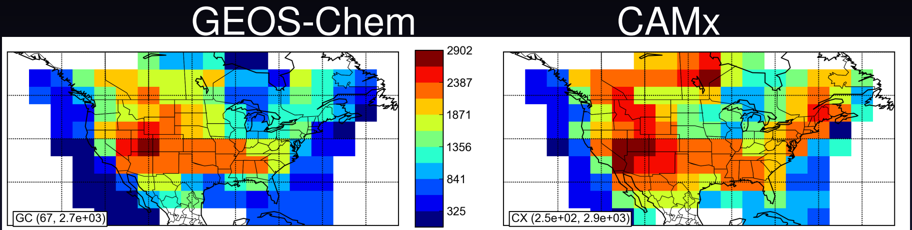


Figure 16 : GEOS-Chem (left) and CAMx (right) planetary boundary layer heights.

- The CAMx PBL is diagnosed from vertical diffusivity using ENVIRON's vertavg algorithm.
- Note relatively good agreement in the southeast