Use of Air Quality Forecasting as a Diagnostic Tool over the Northeastern US

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Outline

- Introduction about model-based air quality forecasting
- An example – Evaluation of isoprene predictions on a diurnal scale
- Diagnostic Analyses
- Results
- Conclusions

Introduction

- The Bureau of Air Quality Analysis and Research (BAQAR) within the Division of Air Resources at New York State Department of Environmental Conservation (NYSDEC) has been performing air quality forecast over the Northeast US using CMAQ at 12 km grid resolution since June 2005.
- Model inputs and outputs of these simulations have been (and continue to be) archived.
- The overall objective is to assess the utility of the forecast-based simulations as a diagnostic tool by using a combination of analyses on the response of the model to biogenic, anthropogenic and secondary species.
- To this end, we chose to evaluate the isoprene predictions, along with other supporting species, on a diurnal scale between June-August 2005.

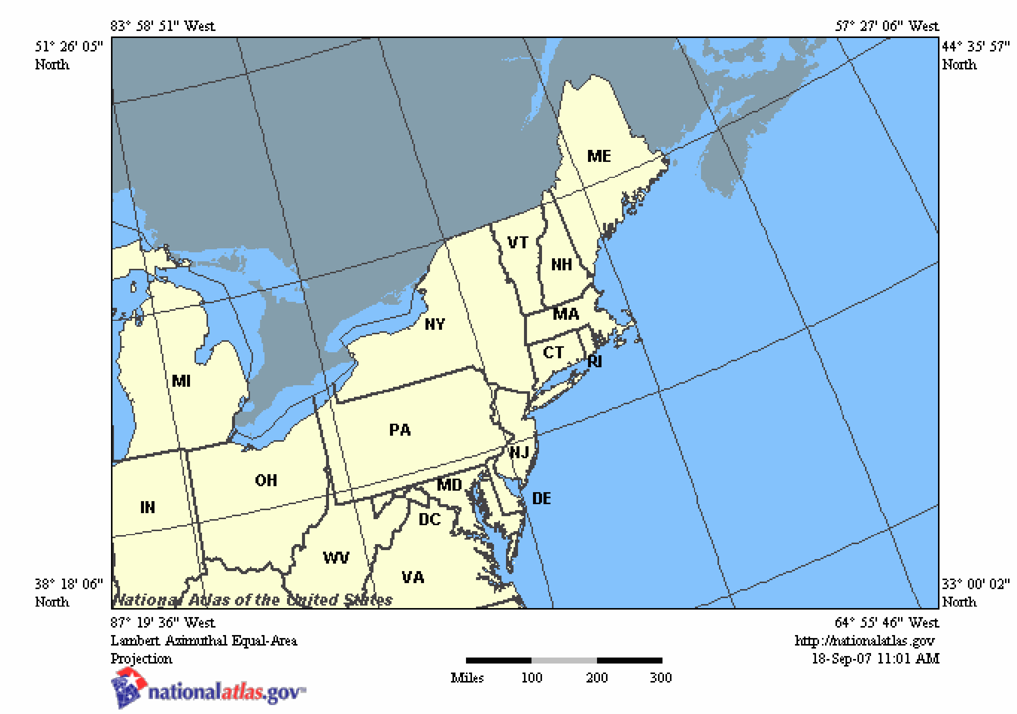
Short Overview of NYSDEC Forecast System

- Follows the air quality forecast system used by EPA/NOAA.
- For the summer 2005 simulations, it consisted of the ETA meteorological model, the PREMAQ emissions and meteorological preprocessor and the CMAQ (v4.4) model.
- Each CMAQ simulation was performed for 48 hours starting at 12:00 noon Greenwich Mean Time (GMT).
- Simulations initialized using default initial conditions (IC) on June 1, 2005. Everyday thereafter used modeled concentration fields from the previous day as IC.
- Time-invariant boundary conditions were used.

Approach

- Modeled isoprene concentrations from the *CONC* file for layer 1 (~35m thick) for June-Aug of 2005 were used in the analysis.
- Measured concentrations were obtained from the EPA Air Quality System (AQS) for sites in the Northeastern US (ME, VT, NH, NY, MA, CT, RI, PA, NJ, MD, DE and DC).

- Hourly concentrations of ethylene (anthropogenic and biogenic origin), ozone (product of photochemical reaction) and nitrogen dioxide (anthropogenic and chemical production) were also obtained, where available, for the same sites, to serve as supplemental information in the evaluation of model performance.



Analysis

- Utilized data from June 12-August 31, 2005 in the analyses, with June 1-11 as spin-up period to avoid effect of IC.
- Compared hourly model predictions with measured concentrations.
- Estimated correlation, normalized mean gross error and normalized mean bias for the entire summer period.
- Examined diurnal profiles averaged over the entire summer period.
- Reran the simulations for the same period using process analysis to confirm initial hypothesis.

Model Performance for Isoprene over the Summer Period (June 12-August 31,

59% data missing

State	Site ID	No.of Pairs	Obs.Mean (ppb)	Pred.Mean (ppb)	Correlation, r	RMSE[a] (ppb)	No.of Pairs, where Obs.> 0	NMGE[b] (%)	NMB[c] (%)
CT	090019003	1616	0.477	0.231	0.446	0.718	1334	75.7	-53.1
	090031003	1729	0.495	0.263	0.43[illegible]	0.589	1714	73.8	-46.9
	090090027	1865	0.260	0.284	0.525	0.433	1691	82.6	7.7
D.C	110010043	1520	0.402	0.959	0.404	1.210	1348	165.3	128.4
ME	230052003	1831	0.183	0.219	0.255	0.516	1113	102.9	-11.6
	230090102	1630	0.311	0.106	0.318	0.420	1337	81.2	-67.4
	230313002	1602	0.435	0.383	0.441	0.644	1437	76.1	-13.2
MD	240053001	803	0.429	0.524	0.544	0.575	794	75.1	22.0
MA	250092006	1454	1.001	0.468	0.418	1.158	1428	72.1	-53.3
	250094004	1606	0.491	0.196	0.351	0.805	1501	85.6	-60.2
	250130008	1625	0.818	0.446	0.393	0.917	1615	71.5	-45.5
	250154002	824	1.989	0.526	0.380	2.943	820	79.3	-73.6
NH	330111011	1515	0.885	0.765	0.580	1.022	1504	63.8	-13.9
NJ	340070003	1810	0.397	1.180	0.524	1.323	1803	214.3	196.9
	340210005	1796	0.439	0.697	0.502	0.716	1783	102.8	58.4
	340230011	1583	0.783	1.141	0.542	1.236	1575	88.4	45.3
NY	360050083	1526	0.769	0.300	0.272	0.9[illegible]	1525	78.2	-61.0
PA	420010001	1722	0.479	1.370	0.634	1.995	1644	211.2	185.2

58% data missing

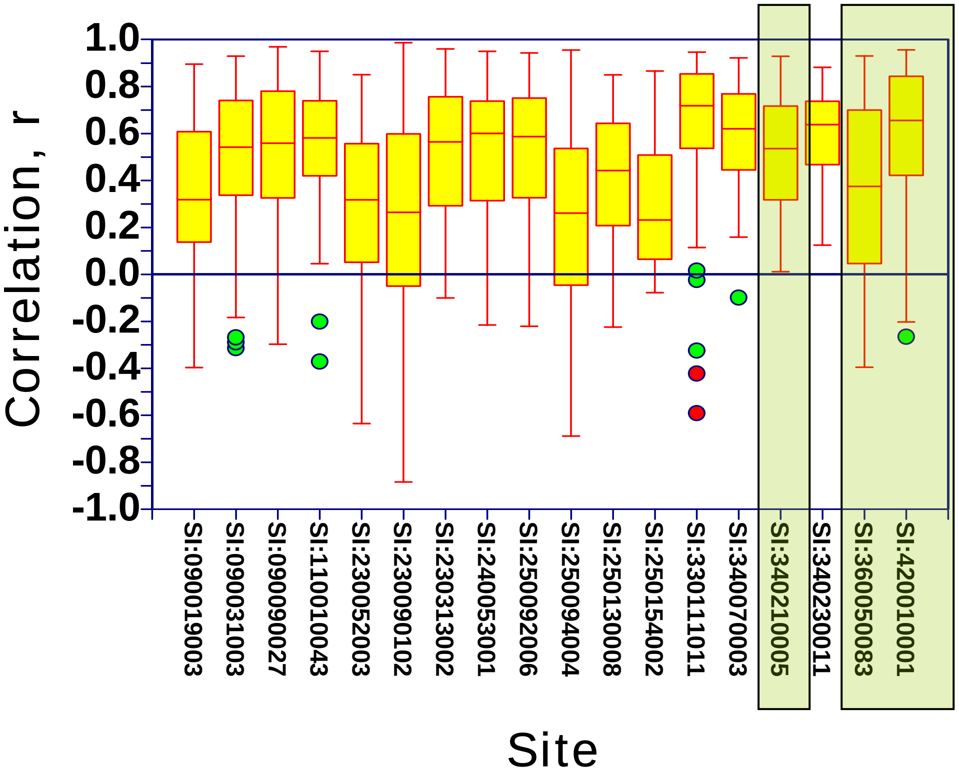
[a] RMSE: Root Mean Square Error = $\sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(C_{pred}-C_{obs}\right)^2}$

[b] NMGE: Normalized Mean Gross Error = $\frac{100*\frac{1}{n}\sum ABS\left(C_{pred}-C_{obs}\right)}{\frac{1}{n}\sum C_{obs}}$

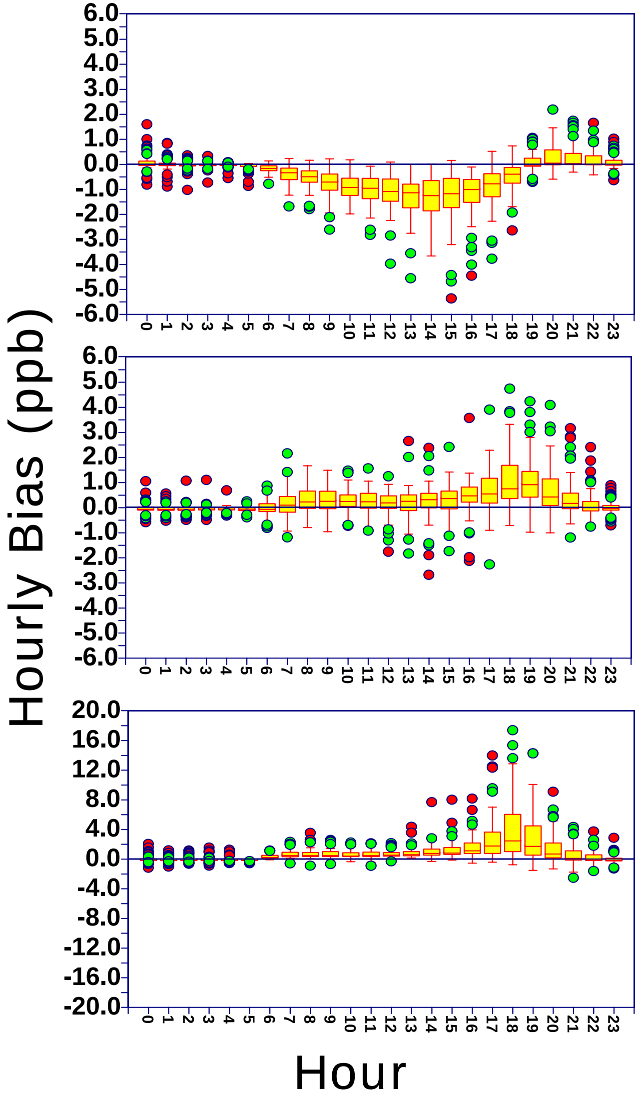
[c] NMB: Normalized Mean Bias = $\frac{100*\frac{1}{n}\sum\left(C_{pred}-C_{obs}\right)}{\frac{1}{n}\sum C_{obs}}$

Distribution of Daily Correlations by Site (Isoprene)

- Correlation between predicted and measured hourly isoprene concentrations each day.
- Only days with 18 hours or more of valid pairs of data were included (i.e., at the least, 75% data availability each day) .
- Median daily correlations ranged from 0.24 to 0.72



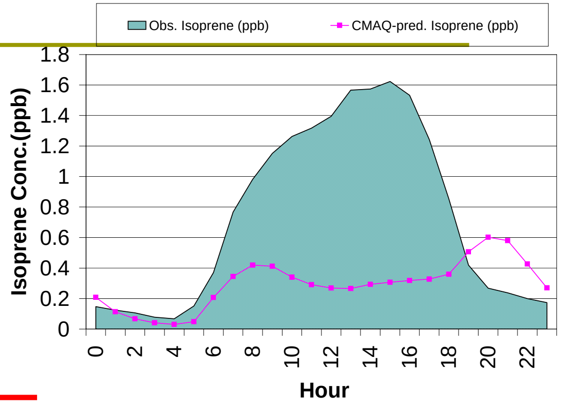
Hourly Bias of Isoprene (Predicted – Measured) at Selected Sites



Site 360050083

New York Botanical Gardens (NYBG)

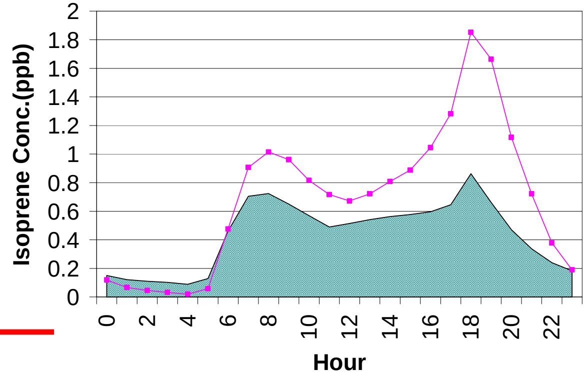
Urban



Site 340210005

New Jersey, Riders College (NJRC)

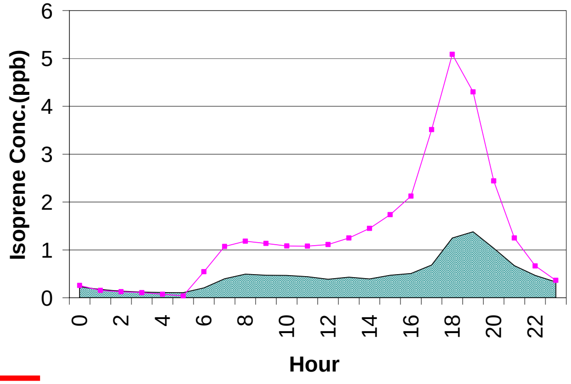
Suburban



Site 420010001

Pennsylvania NARSTO Site at Arendtsville (PANARSTO)

Rural



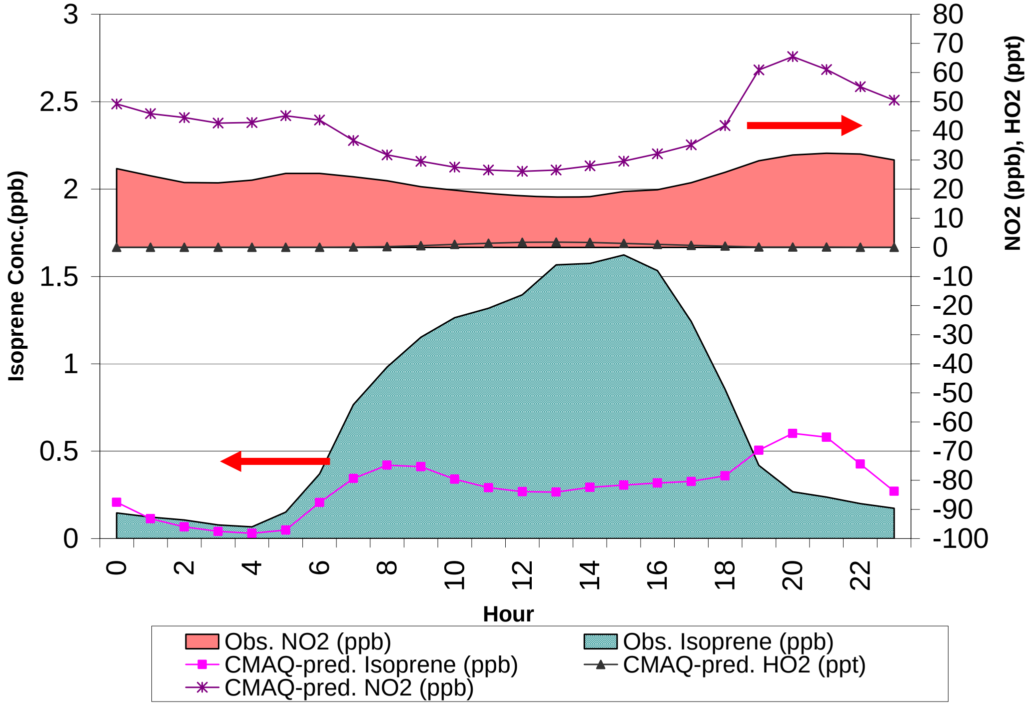
Questions of Interest

- Why do we see dual peak in predicted concentrations when the measurements do not show such a pattern ?
- What are the reasons for agreement/ disagreement between measurements and predictions?
- The potential solution lies in examining possible sources and sinks
  - Sources: Biogenic sources, addition of isoprene by convection/advection from adjacent cells
  - Sinks: Consumption by reaction; Depletion by convection/advection to adjacent cells; Currently in CMAQ, dry deposition is not a sink for isoprene.
- The dual peak could be interpreted as a “depression” during the mid-day hours.
- In CMAQ-CB4, consumption of isoprene is through a set of 5 reactions

Reactions Consuming Isoprene

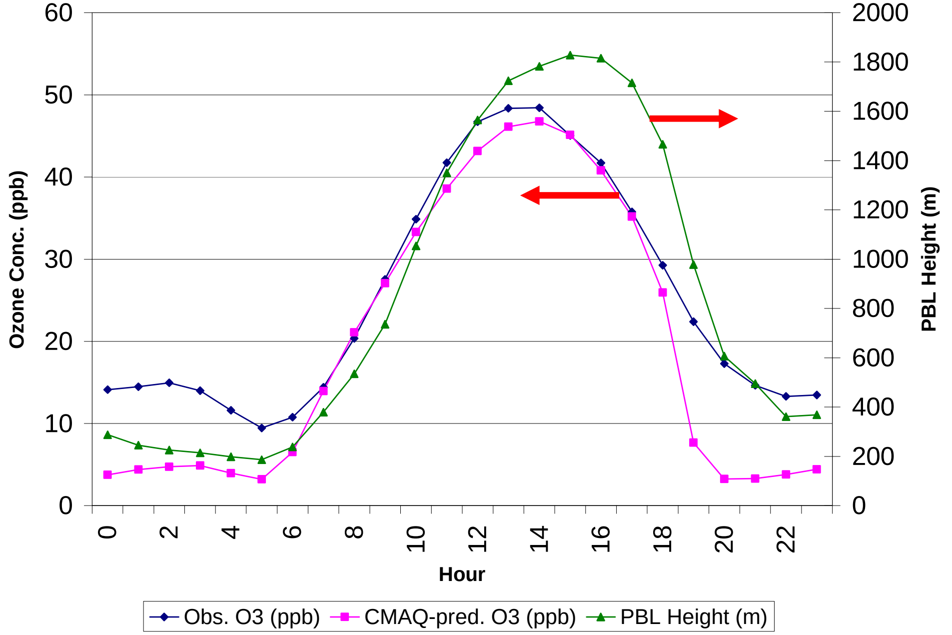
Rx #	**Reac. 1**	**Reac. 2**	**ISPD**	**FORM**	**XO2**	**HO2**	**PAR**	**OH**	**Others**	**Rate Const. (mol/cc)$^{-2}$sec^{-1}**
75	ISOP	O	0.75	0.5	0.25	0.25	0.25	---	0.25C2O3	3.6E-11
76	ISOP	OH	0.912	0.629	0.991	0.912	---	---	0.088 XO2N	2.54E-11 * exp(407.6/T) ***[9.97E-11] @ 25 °C***
77	ISOP	O3	0.65	0.6	0.2	0.066	0.35	0.266	0.15ALD2 + 0.066CO + 0.2C2O3	7.86E-15 * exp(-1912/T) ***[1.29E-17] @ 25 °C***
78	ISOP	NO3	0.2	---	1	0.8	2.4	---	0.8 NTR + 0.2NO2 + 0.8ALD2	3.03E-12 * exp(-448/T) ***[6.74E-13] @ 25 °C***
94	ISOP	NO2	0.2	---	1	0.8	2.4	---	0.8 NTR + 0.2NO + 0.8ALD2	1.49E-19

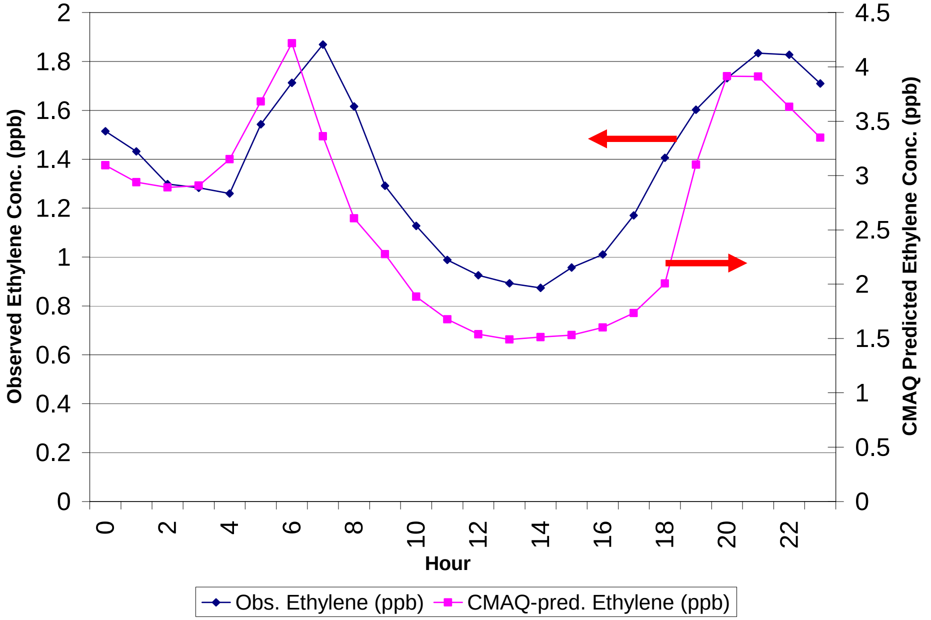
R76 typically dominates during day time; R78 dominates @ night time



NYBG (360050083), Urban

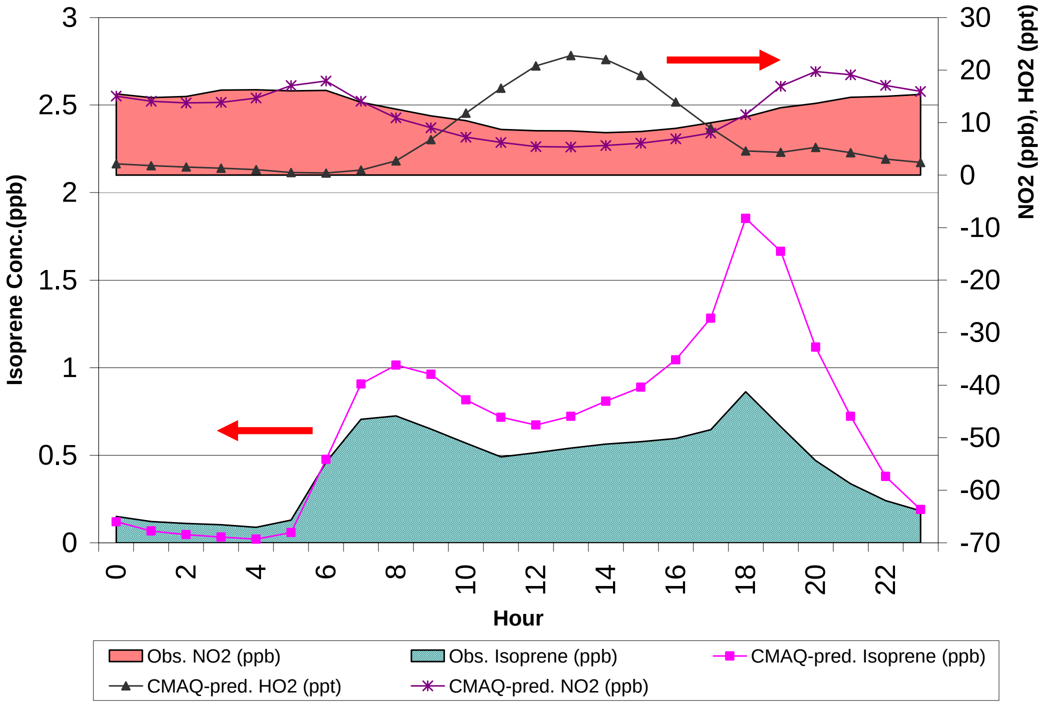
- NO_2 concentrations over-predicted
- Consistent with predicted night-time O_3 being lower than observed (due to scavenging by NO_x).
- PBL height seems reasonable for summer
- Ethylene, while over-predicted, appears to track measured profile

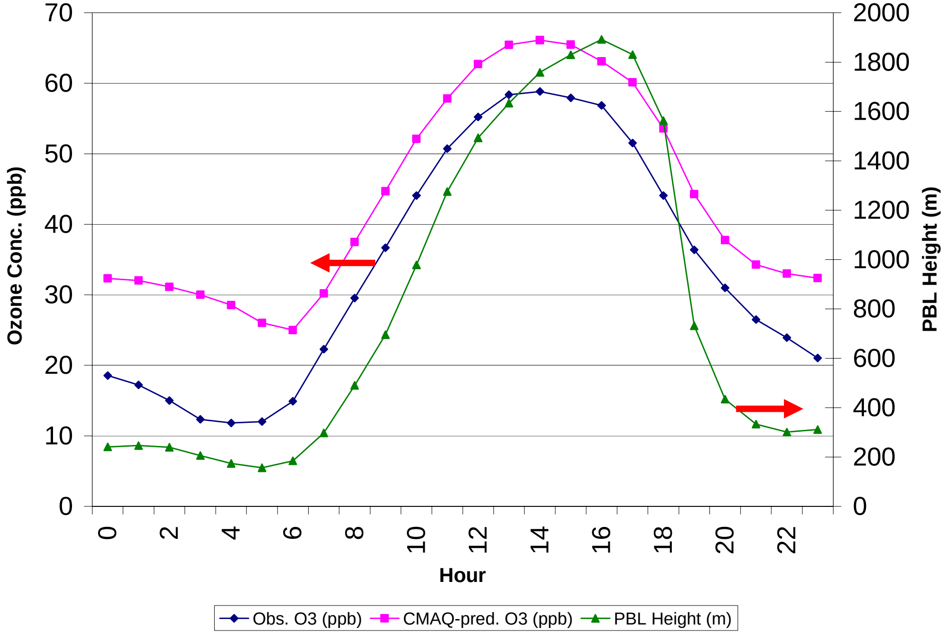


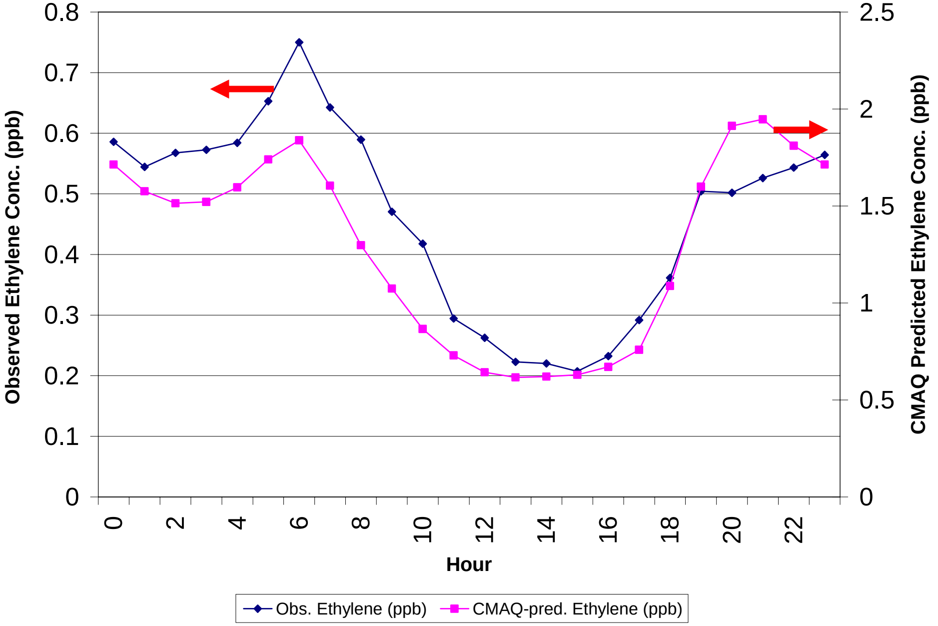


NJRC (340210005), Suburban

- Predicted Isoprene tracks measured profiles
- Isoprene emissions appear to be overestimated

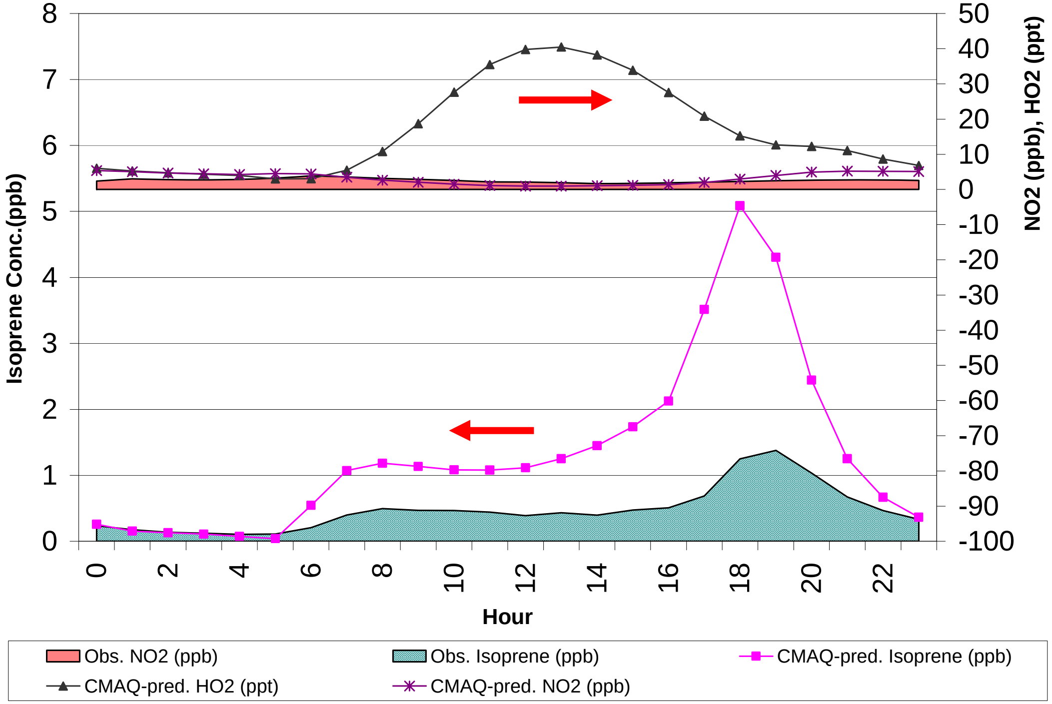


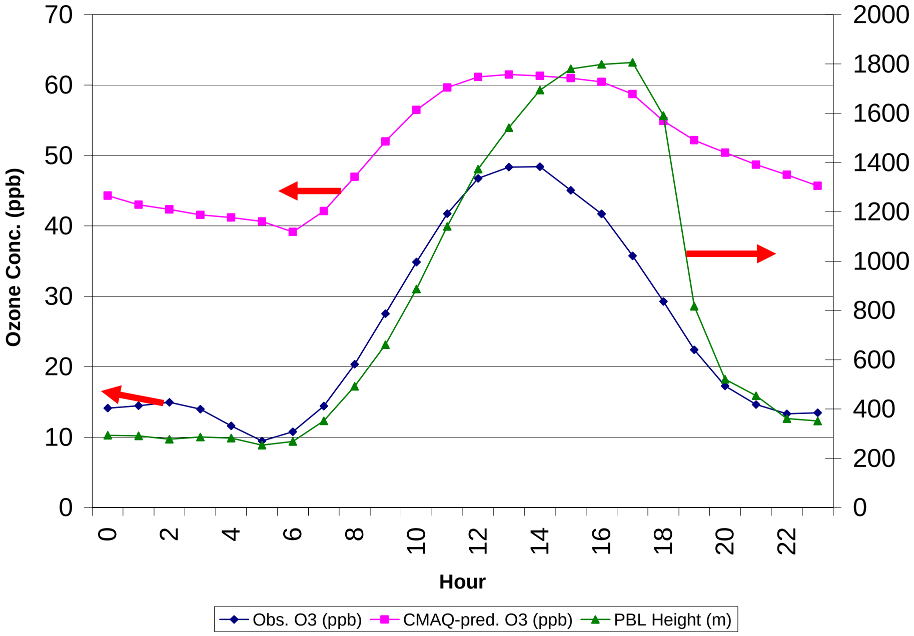


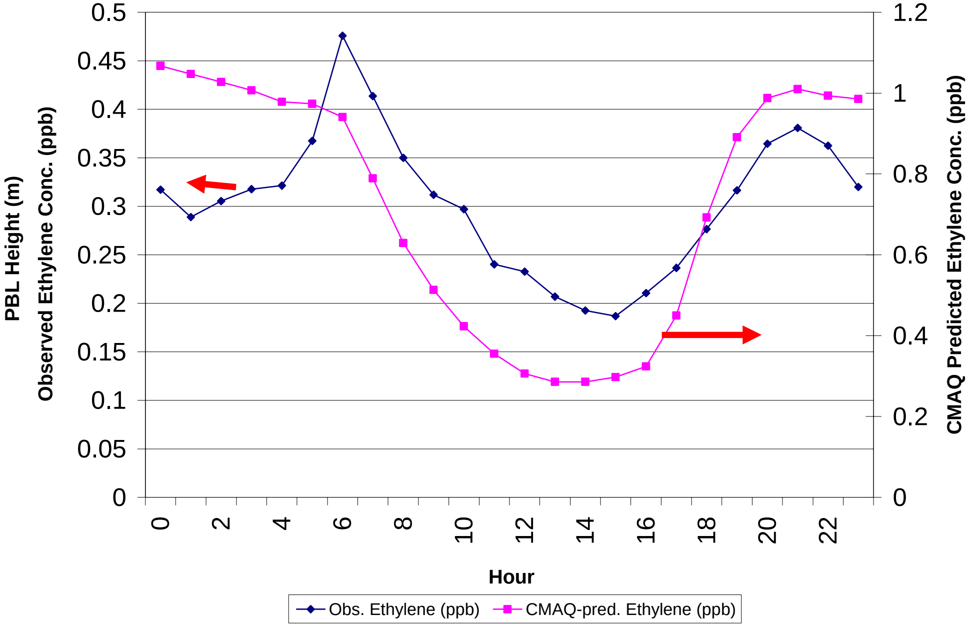


PANARSTO (42001001), Rural

- ☐ Predicted Isoprene tracks measured profiles
- ☐ Isoprene emissions appear to be overestimated

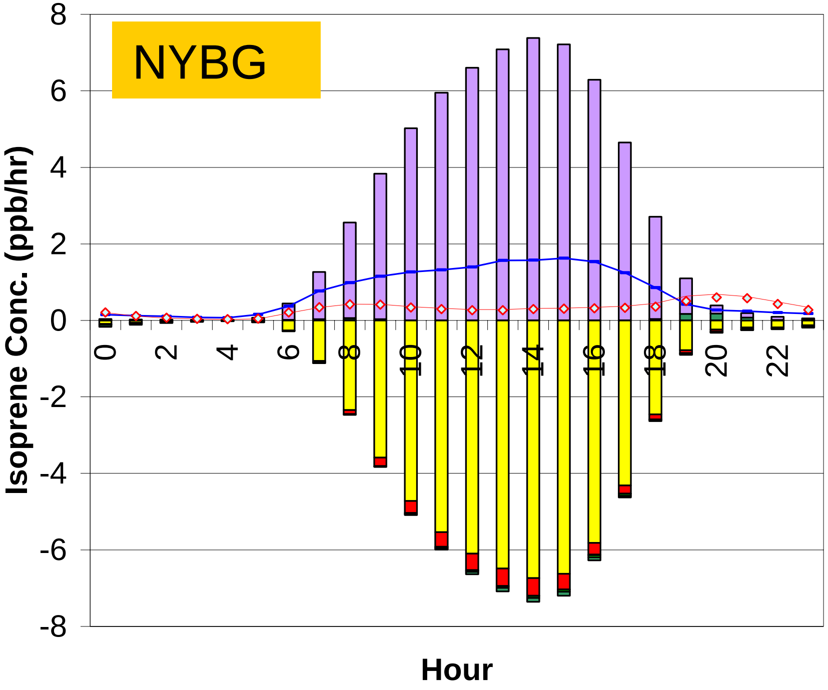


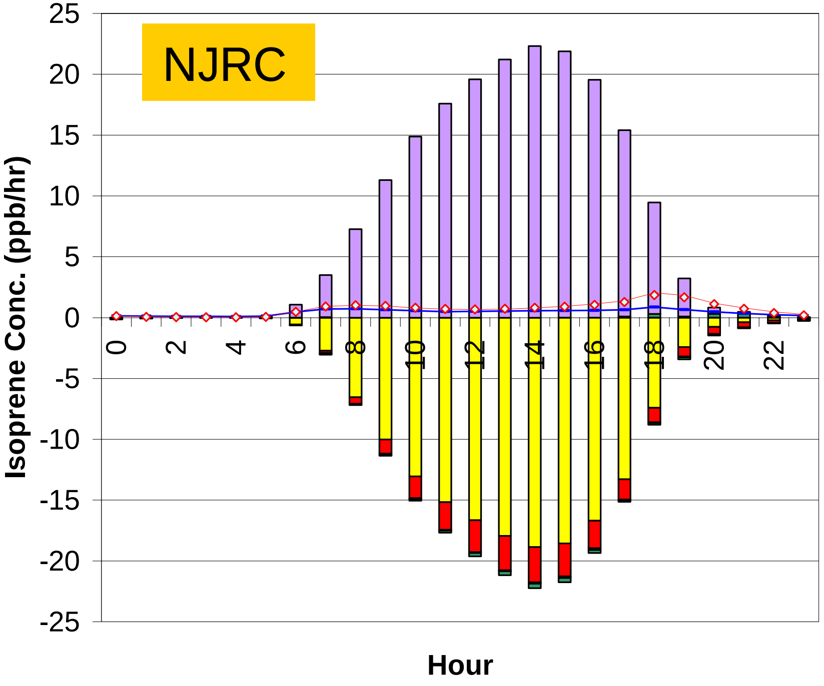


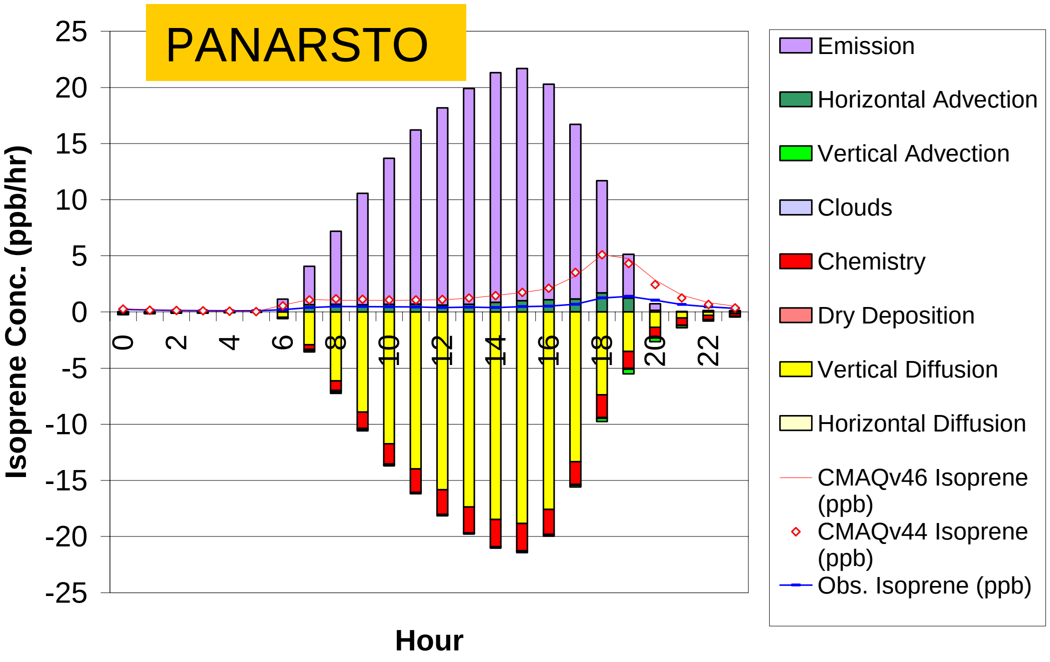


What does Process Analysis show ?

- For surface layer, emission is major source; vertical diffusion and chemistry are the dominant loss processes.
- For upper layers (not shown), chemistry is the dominant loss process







Day time profile

- At NYBG, over-prediction of isoprene loss during mid-day hours could be mostly explained by chemical reactions (~0.5 ppb/hr), probably due to overestimation of NOx. However, it is also quite possible that the model is underestimating additional sources (emissions, mixing from above?)
- For NJRC and PANARSTO, the process analysis confirmed emissions to be the major source. Mixing processes are likely modeled satisfactorily, given the similarity of observed vs. predicted profiles of isoprene, ozone and ethylene at these two sites. It is likely emissions are overestimated resulting in a positive bias.

Evening/Night time peak?

- Evening peak was present at all three sites (8-9 pm at NYBG, 6 pm at other two sites).
- While horizontal advection from adjacent cells contributed to some addition, it was not a major source.
- Night-time isoprene loss is subject of extensive research
  - Reaction of ISOP + NO_3 (Starn et al., 1998)
  - Vertical diffusion (Sillman et al, 2002; Hurst et al., 2001)
  - Reaction with OH (Hurst et al., 2001)
  - Other unknown (as yet) mechanisms (Goldan et al., 1995)
- Starn et al. (1998) observed that rapid isoprene loss was associated with conditions when $[O_3]*[NO_2] > 300$ ppb^2. At NYBG, while observed conditions fall in this range (dropped from 657 to 382 ppb^2, suggesting the possibility of loss by ISOP + NO_3 rxn), CMAQ predictions showed < 200 ppb^2 (supporting the argument that NO emissions were overestimated)
- Cause of peak at other two sites is not clear at this time.

Conclusions

- Archived forecast simulations presents a unique opportunity to evaluate the model under varied atmospheric conditions. While this specific example could have also been done using retrospective modeling, the presence of archived simulations allows us to look at other seasons/years of interest.
- Overall, the model appeared to track diurnal profile at most sites.
- Analysis revealed that the causes of model performance differ by site.
  - Possible overestimation of NOx emissions at NYBG
  - Possible overestimation of isoprene emissions at NJRC and PANARSTO

References

- Goldan, P. D., W. C. Kuster, F. C. Fehsenfeld and S. A. Montzka (1995). "Hydrocarbon measurements in the southern United States: The Rural Oxidants in the Southern Environment (ROSE) Program 1990." Journal of Geophysical Research 100(25): 945-963.
- Hurst, J. M., D. J. Barket, O. Herrera-Gomez, T. L. Couch, P. B. Shepson, I. Faloona, D. Tan, W. Brune, H. Westberg, B. Lamb, T. Biesenthal, V. Young, A. Goldstein, J. W. Munger, T. Thornberry and M. A. Carroll (2001). "Investigation of the nighttime decay of isoprene." Journal of Geophysical Research 106(D20): 24335-24346.
- Sillman, S., M. A. Carroll, T. Thornberry, B. K. Lamb, H. Westberg, W. H. Brune, I. Faloona, D. Tan, P. B. Shepson, A. L. Sumner, D. R. Hastie, C. M. Mihele, E. C. Apel, D. D. Riemer and R. G. Zika (2002). "Loss of isoprene and sources of nighttime OH radicals at a rural site in the United States: Results from photochemical models." Journal of Geophysical Research 107(D5).
- Starn, T. K., P. B. Shepson, S. B. Bertman, D. D. Riemer, R. G. Zika and K. Olszyna (1998). "Nighttime isoprene chemistry at an urban-impacted forest site." Journal of Geophysical Research 103(D17): 22437-22448.