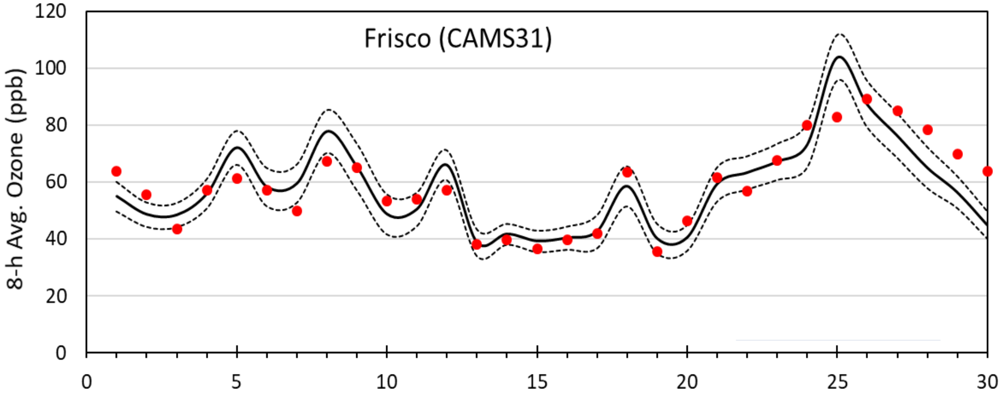
DDM Enhancements in CAMx: Local Chemistry Sensitivity and Deposition Sensitivity



Uncertainty in CAMx 8-h O_3 due to emissions, deposition, boundary concentrations and chemistry for June, 2012

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Alan M Dunker

AQRP project 18-007

Texas Air Quality Research Program Workshop, 21 August 2019

RAMBØLL Bright ideas. Sustainable change.

Acknowledgments and Disclaimer

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Dr. Bonyoung Koo contributed to the project design

Results shown today are draft and subject to change

Objective

Respond to the following AQRP research priority

Sensitivity of modeling results to uncertainties in model inputs. Develop new tools and methodologies or find innovative ways to apply existing tools, such as DDM, HDDM, Process Analysis, etc., to estimate the sensitivities of photochemical grid modeling results to uncertainties in model inputs. Projects should focus on the development of tools and applications that are easily portable and scalable. i.e., tools and applications that can be easily utilized by the modeling community for practical research and policy development purposes

Previous Work – for Context

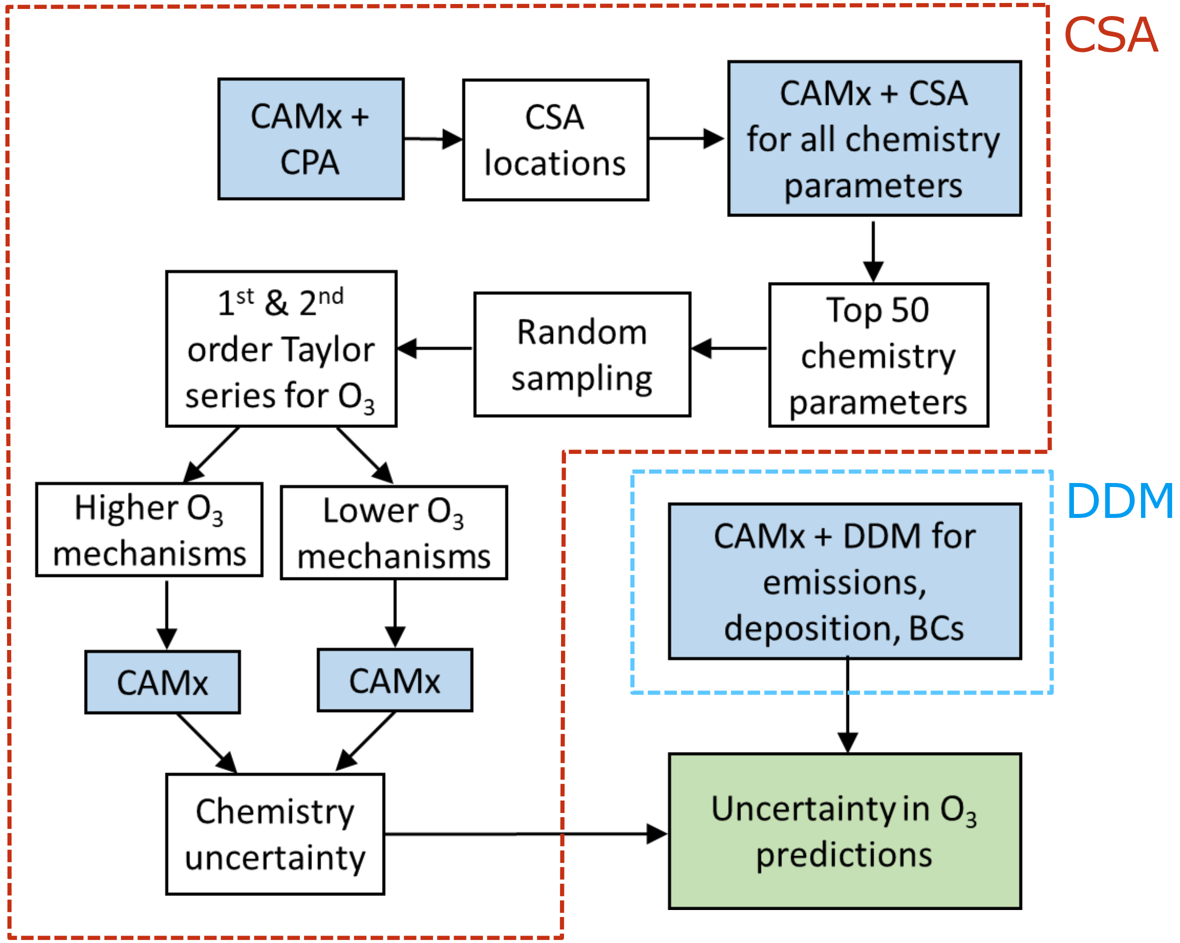
Beddows et al. (2017) performed extensive uncertainty analysis of a 1 month CMAQ simulation for London

They constructed a reduced-form model using 576 CMAQ simulations equivalent (computationally) to a 48-year simulation

They quantified uncertainties due to emissions, boundary concentrations, deposition and chemical reaction rates in CB05

Beddows, A.V., Kitwiroon, N., Williams, M.L., Beevers, S.D., 2017. Emulation and sensitivity analysis of the Community Multiscale Air Quality Model for a UK ozone pollution episode, Environ. Sci. Technol., 51, 6229-6236.

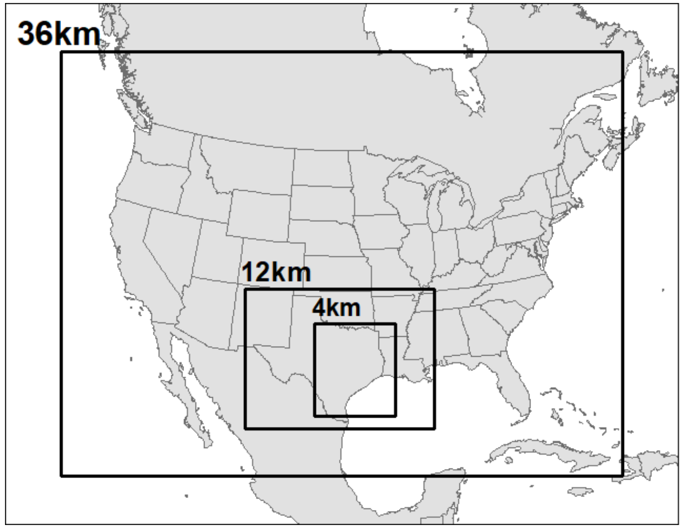
Method Outline



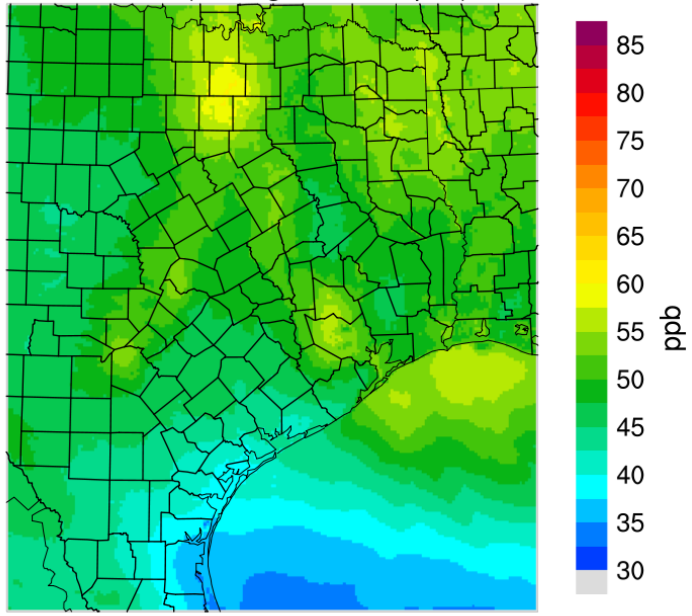
- Use DDM to estimate O_3 uncertainty due to emissions, BCs, deposition
    - deposition sensitivity new in CAMx
- Use CSA to estimate O_3 uncertainty due to chemistry
    - CSA is new for this project
- Combining the uncertainties
    - variance (var) is [standard deviation]^2
    - standard deviation (σ) is given by:

$$[var(chem) + var(emis) + var(dep) + var(BCs)]^{0.5}$$

June 2012 Modeling Episode

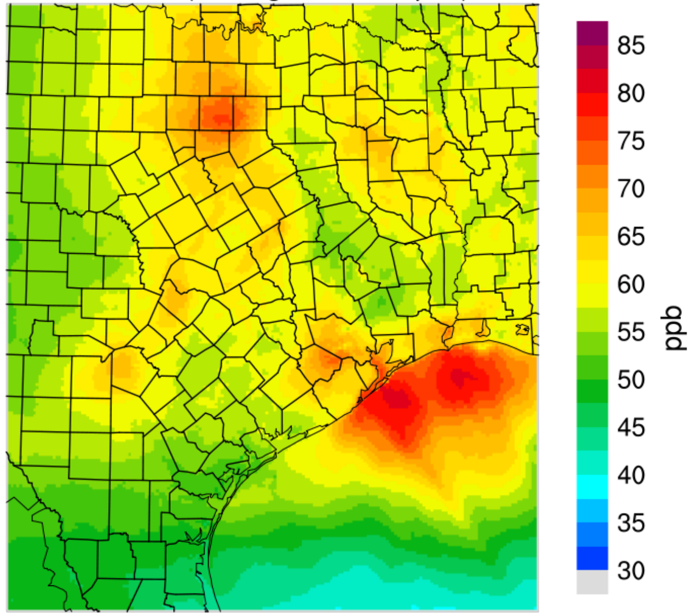


June MDA8 O_3
10 am to 6 pm



Min= 34.2, Max= 60.5

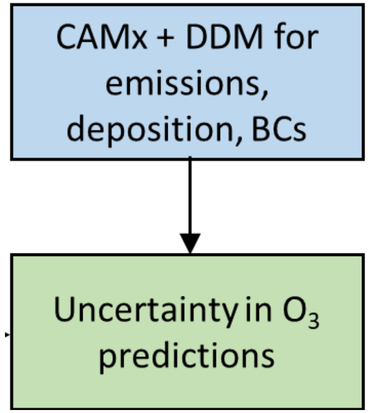
Top10 days
June 7-9 and 22-28



Min= 39.7, Max= 81.7

Modeling input data provided by the TCEQ

Estimated Input Uncertainties


CAMx + DDM for emissions, deposition, BCs

Uncertainty in O_3 predictions

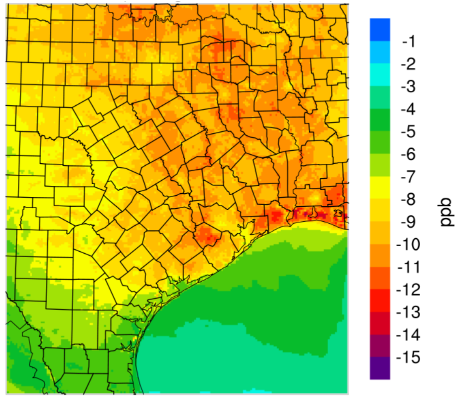
Model Input	Uncertainty Factor
DFW anthropogenic VOC emissions	1.35
DFW anthropogenic NOx emissions	1.3
DFW biogenic VOC emissions	1.5
DFW biogenic NOx emissions	2.
Non-DFW emissions of all species	1.4
Oceanic inorganic iodine (Ix) emissions	2.
Dry deposition velocity of O_3	2.
Dry deposition velocity of all species but O_3	2.
Boundary concentrations of O_3	1.25

- Input uncertainties estimated from literature review
- Uncertainties are assumed to be independent and lognormally distributed
- Combine factors with 1st order sensitivities [$S^{(1)}$] computed with DDM like so, for deposition:

$$var(dep) = \left[\frac{\ln 2}{2} S^{(1)}(Dep\ O_3)\right]^2 + \left[\frac{\ln 2}{2}\ S^{(1)}(Dep\ other)\right]^2$$

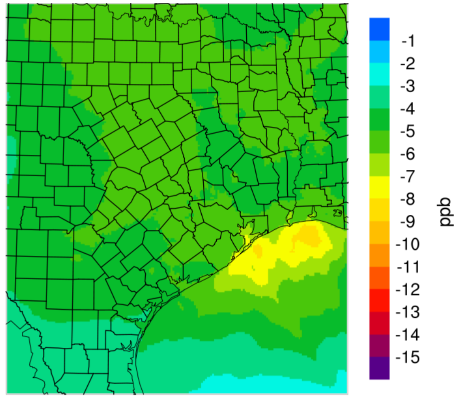
Uncertainty Due to Ozone Deposition

Top10 O_3 sensitivity to O_3 deposition



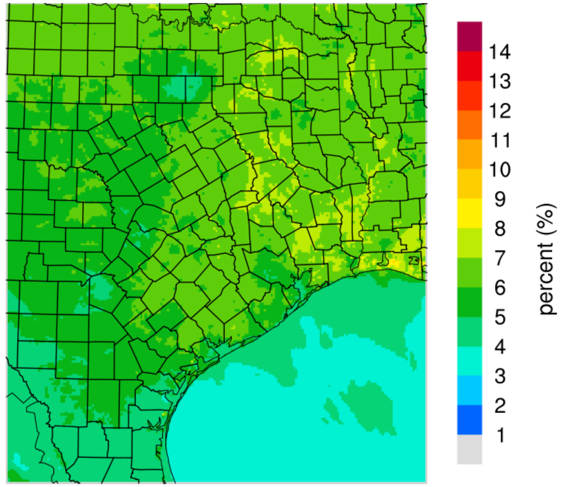
Min= -14.7, Max= -2.9

Top10 O_3 sensitivity to deposition of species other than O_3



Min= -8.8, Max= -2.6

std dev (%) of Top10 O_3 due to deposition



Min= 3.4, Max= 9.0

- Generally, deposition of O_3 more influential than deposition of all other species combined
- The near-shore gulf of Mexico is an exception to investigate

$$var(dep) = \left[\frac{\ln 2}{2} S^{(1)}(Dep\ O_3)\right]^2 + \left[\frac{\ln 2}{2}\ S^{(1)}(Dep\ other)\right]^2$$

Chemistry Sensitivity Analysis

CAMx DDM can compute

- 1st order sensitivity to rate constant
- 1st order sensitivity to stoichiometric coefficient
- 2nd order sensitivity to rate constant

Parameters in CB6r4

- 230 rate constants
- 764 product coefficients
- 452 uncertain product coefficients
- 230 + 452 = too many

CAMx Process Analysis

- Report information for CAMx grid cells
- Single grid cells or sub-domains

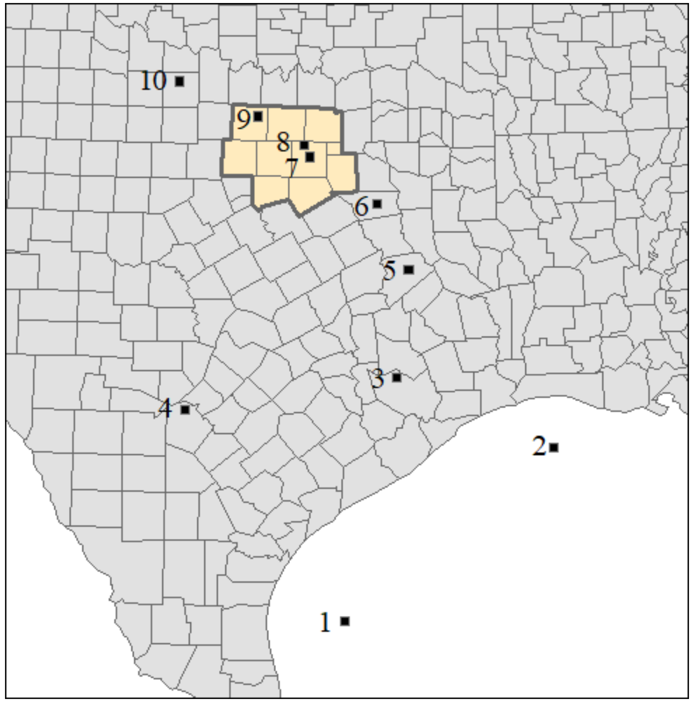
Chemistry Sensitivity Analysis (CSA)

- Apply DDM like Process Analysis, i.e., for subdomains and only to parameters in the chemistry
- Local sensitivity, i.e., no communication between grid cells
- Like running many constrained box models
- Implemented as a CAMx “probing tool” and configured at run-time
- Simpler to use than running many box models

CSA Locations

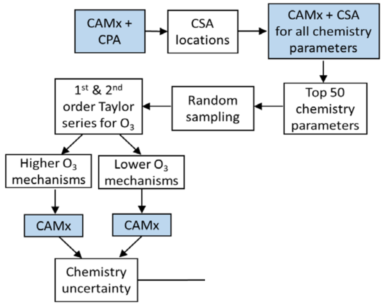
10 locations selected by reviewing CAMx Chemical Process Analysis (CPA) output (extra slides)

Each location is a block of 3 x 3 x 7 grid cells from surface to ~500 m



ID	Location	Description
10	Archer County	Rural, lower BVOC emissions
9	Wise County	Rural, Barnett Shale natural gas production
8	Carrollton (Dallas County)	Urban, outside core area
7	Dallas (Dallas County)	Urban, central core
6	Henderson County	Rural, higher BVOC emissions; higher isoprene fraction
5	Houston County	Rural, higher BVOC emissions; lower isoprene fraction
4	San Antonio (Bexar County)	Urban, outside core area
3	Houston (Harris County)	Urban, outside core area
2	Eastern Gulf of Mexico	Oceanic, net O3 production tendency
1	Western Gulf of Mexico	Oceanic, net O3 destruction tendency

CSA Used to Create Alternative Chemical Mechanisms



CB6r4 Parameters X Uncertainty in Parameters X Random Numbers = Alternative to CB6r4 X CSA Sensitivity to Parameters

Σ = O_3 productivity (ppb/h)

1. Construct N alternatives to CB6r4 using N sets of random numbers
2. Rank mechanisms by O_3 productivity (ppb/h)
   - use 1st order CSA O_3 sensitivity and 2nd order for Top5
3. Select 3 alternatives near +1σ: Hi1, Hi2, Hi3
4. Select 3 alternatives near −1σ: Lo1, Lo2, Lo3
5. Conduct CAMx simulations using 6 alternative mechanisms
   - the Top50 parameters are perturbed

Estimating Chemical Mechanism Parameter Uncertainties

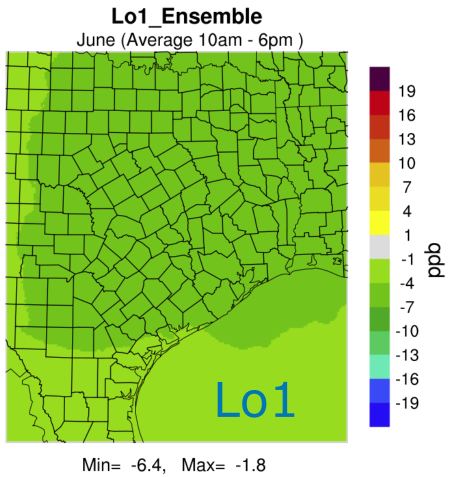
- Rate constant uncertainties
    - NASA JPL evaluation lists many inorganic reaction
    - IUPAC evaluation discusses some reactions
    - Others estimated for this work
    - Factors range from 1.05 to 10.
- Stoichiometric coefficient uncertainties
    - Not aware of any previous estimates
    - Estimated uncertainties of 1.15, 1.3 or 1.5
    - Many (but not all) integer coefficients are certain
    - Some coefficients are correlated (e.g., by N-balance) which we accounted for in the Monte Carlo analysis
- Not considered by this analysis
    - Excluded reactions
    - Unknown chemistry

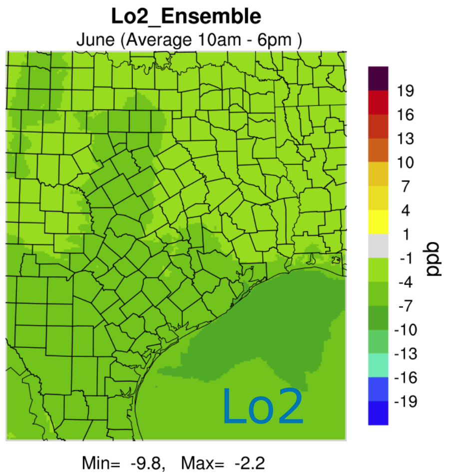
Top 20 CB6r4 Parameters Contributing to Ozone Uncertainty

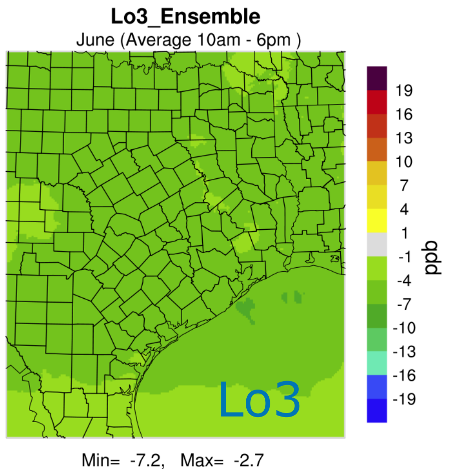
Parameter	Reaction	Cumulative Variance (%)
k1	NO2 = NO + O	26.5
k55	PAN = NO2 + C2O3	41.3
k3	O3 + NO = NO2	55.6
k63	PANX = NO2 + CXO3	63.7
k25	HO2 + NO = OH + NO2	71.0
k54	C2O3 + NO2 = PAN	77.3
k53	C2O3 + NO = NO2 + MEO2 + RO2	81.9
k45	NO2 + OH = HNO3	84.5
k62	CXO3 + NO2 = PANX	87.2
ROR-228	XPAR = 0.874 ROR +0.874 XO2 + 4 others	89.4
k89	ROOH + OH = 0.540 XO2H + 3 others	90.3
k61	CXO3 + NO = NO2 + ALD2 + XO2H + RO2	91.1
k129	PAR + OH = XPAR	91.8
XO2H-130	ROR = 0.940 XO2H + 8 others	92.5
XO2-228	XPAR = 0.874 ROR +0.874 XO2 + 4 others	93.2
k13	O3 + HO2 = OH	93.6
k76	XO2H + HO2 = ROOH	94.1
k72	MEO2 + HO2 = 0.9 MEPX + 0.1 FORM	94.4
k201	OPAN = OPO3 + NO2	94.8
k223	INO3 = I + NO3	95.1

- Top10 account for 89% of variance and Top20 for 95%
- 17 of the Top20 are reaction rates and 3 are stoichiometric coefficients
- NO_2 photolysis ranked top even with small uncertainty (factor 1.1)
- Influential parameters related to:
    - NO-NO_2-O_3 photo-stationary state
    - NO_2 availability (NOx recycling)
    - Radical production
    - Iodine availability

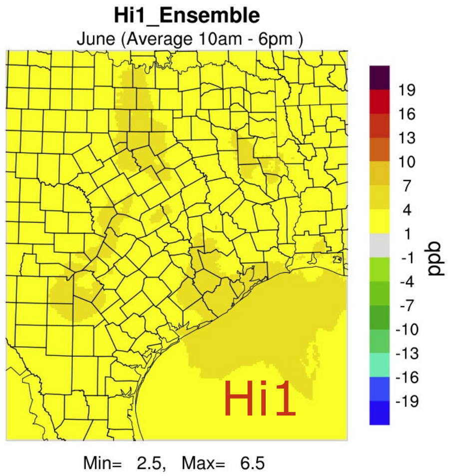
CB6r4 Hi & Lo Simulations – Top 10 Days

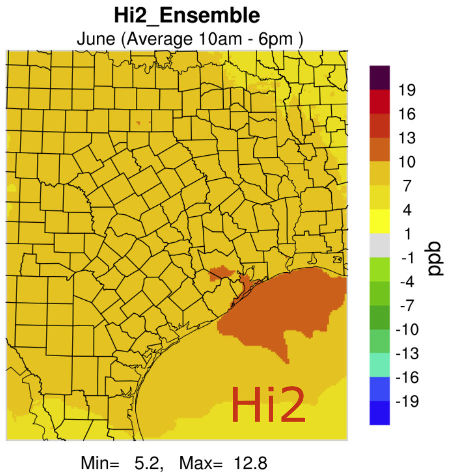


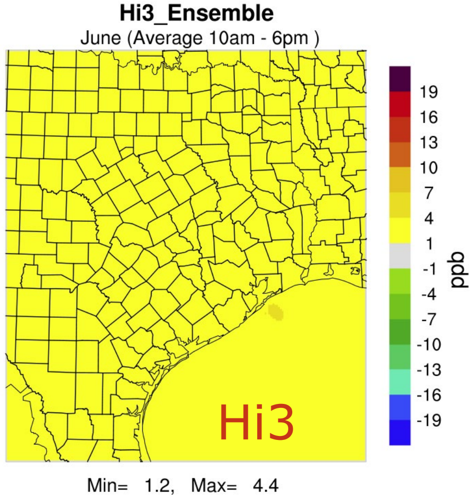


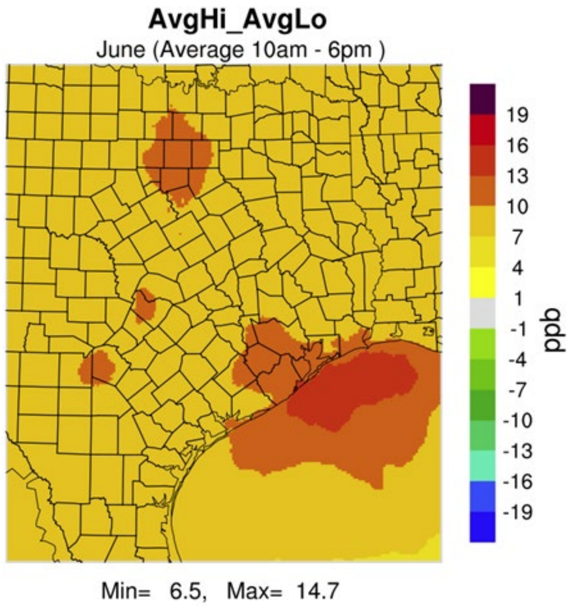


O_3 difference (ppb) from the mean of 6 simulations (ensemble mean)









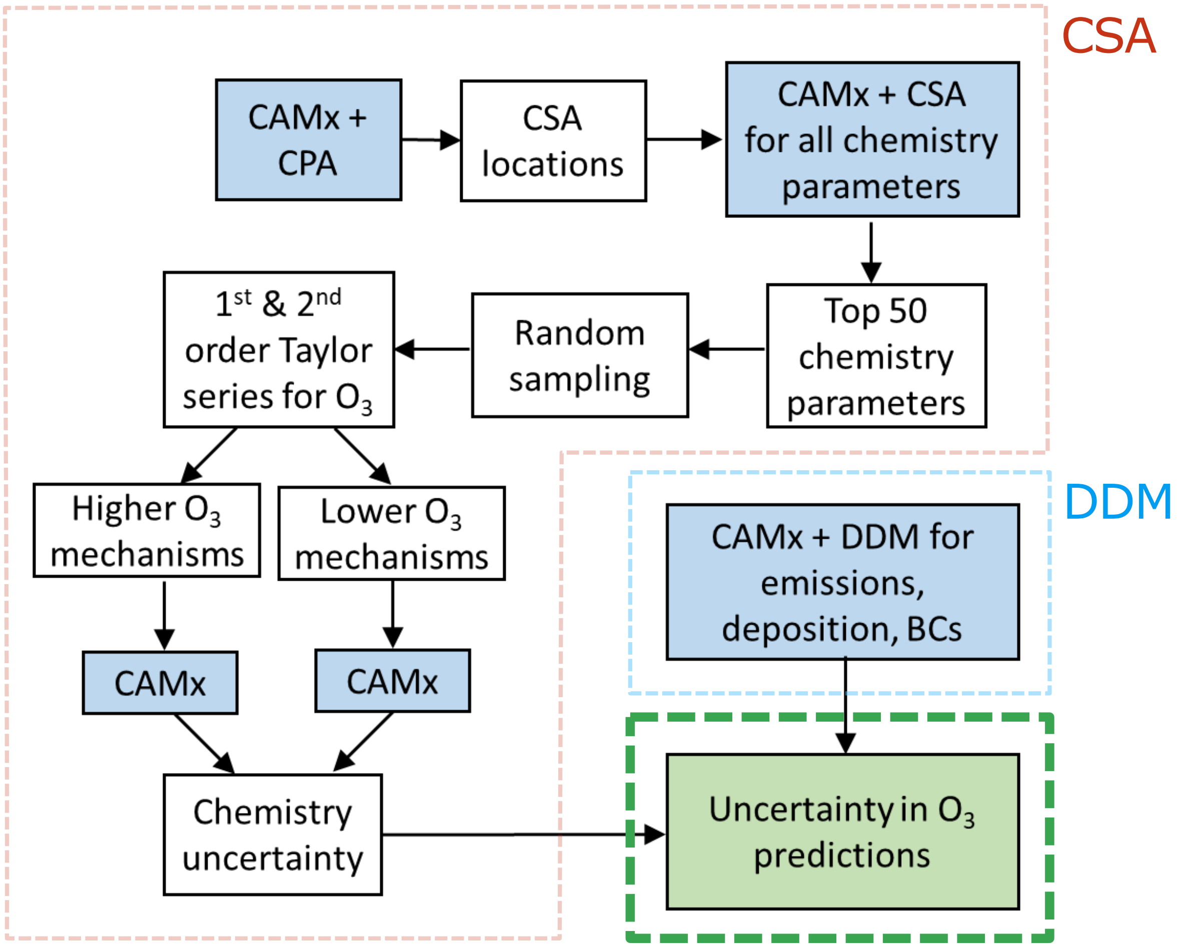
- Each simulation is unique showing the need for an ensemble
- Avg Hi – Avg Lo provides the uncertainty due to chemistry

$$var(chem) = \left(\frac{c_i(avgHi) - c_i(avgLo)}{2}\right)^2$$

Comments on CSA Analysis and CB6r4 Hi Lo Mechanisms

- Importance (ranking of each parameter) depends both on sensitivity and uncertainty
- Including 2nd order sensitivity for the Top 5 parameters did not change their ranking
- Top20 parameters accounted for 95% of the variance

Ozone Uncertainty



CSA

CAMx + CPA

CSA locations

CAMx + CSA for all chemistry parameters

Top 50 chemistry parameters

Random sampling

1st & 2nd order Taylor series for O_3

Higher O_3 mechanisms

Lower O_3 mechanisms

CAMx

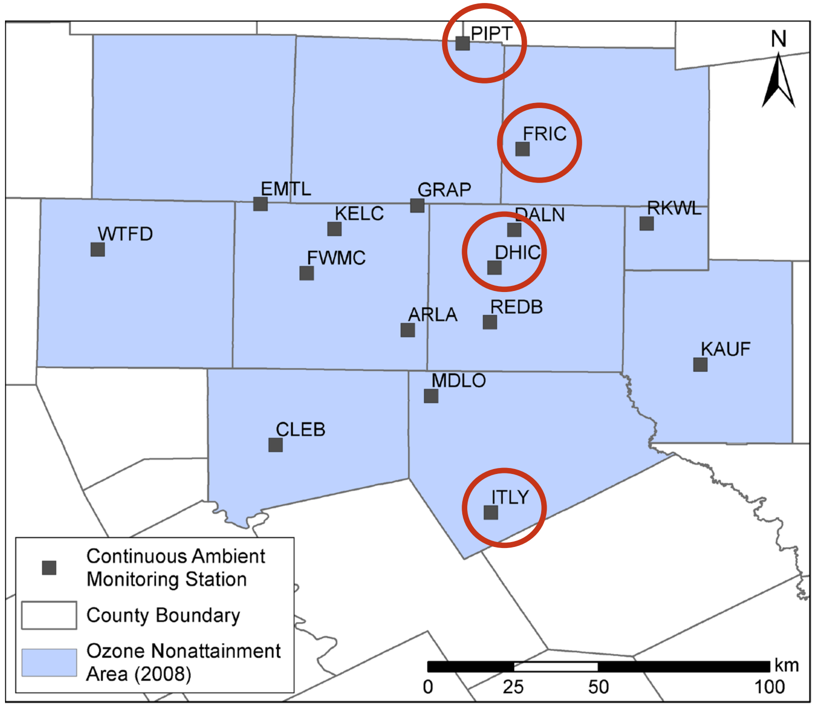
CAMx

Chemistry uncertainty

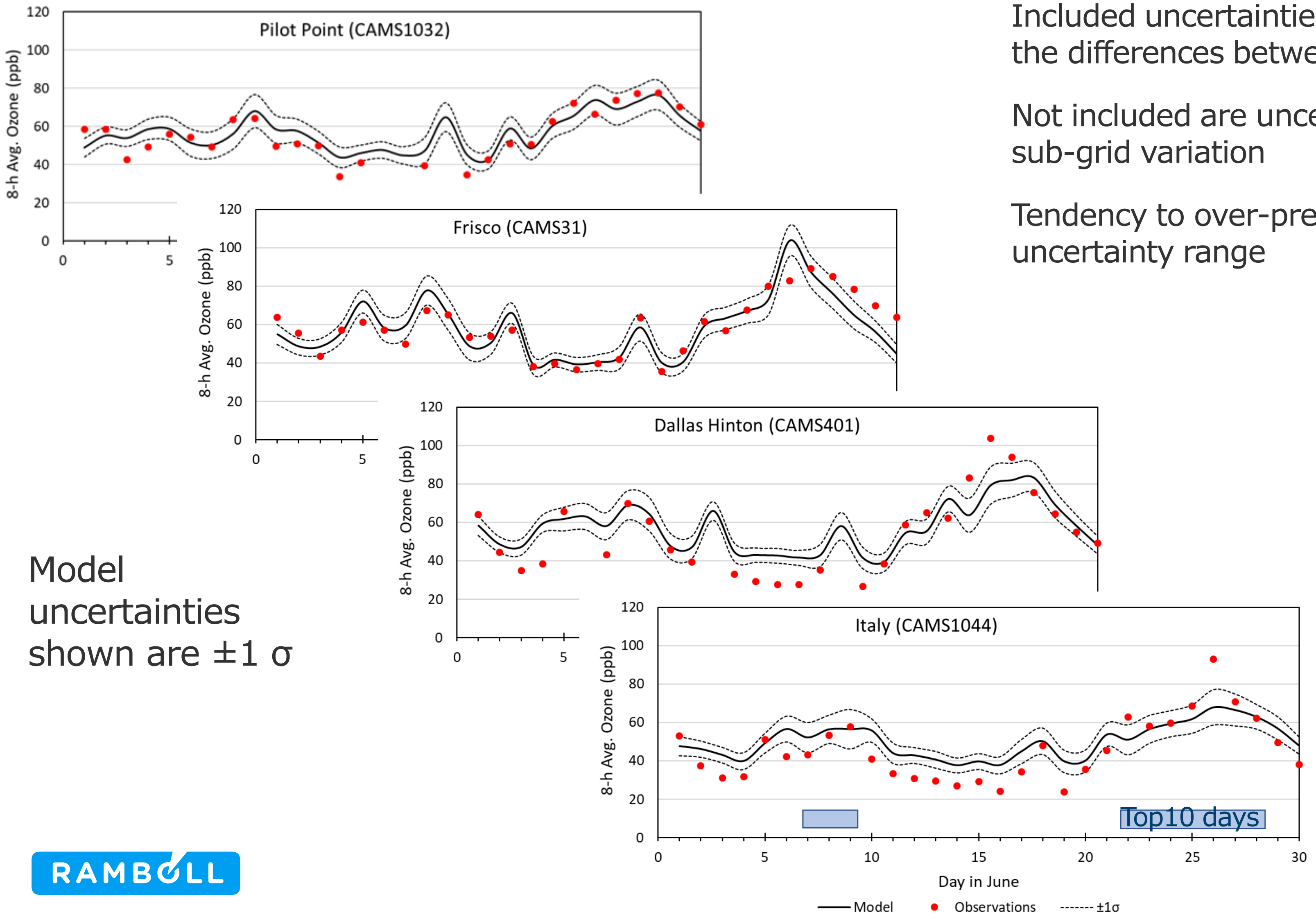
DDM

CAMx + DDM for emissions, deposition, BCs

Uncertainty in O_3 predictions



Ozone Uncertainty: Time Series



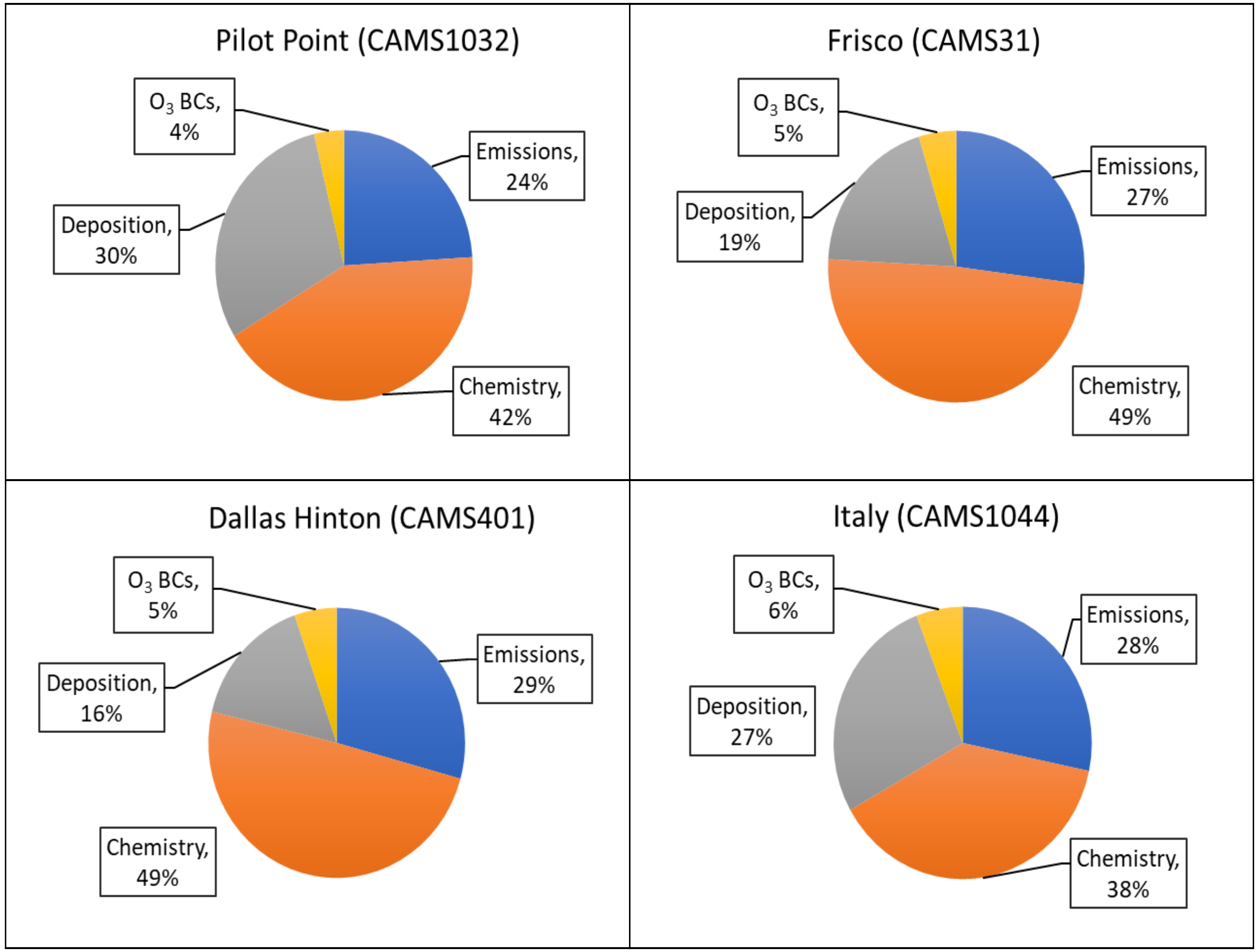
Included uncertainties can account for much but not all of the differences between observations and model results

Not included are uncertainties due to meteorology and sub-grid variation

Tendency to over-predict low days outside ±2 σ uncertainty range

Model uncertainties shown are ±1 σ

Ozone Uncertainty: Contributions



Pilot Point (CAMS1032)

Frisco (CAMS31)

Dallas Hinton (CAMS401)

Italy (CAMS1044)

Contributions (%) to total uncertainty in predicted O_3
Results are averages over June 2012

At all 4 sites, uncertainties in the chemistry contribute the most and uncertainties in O_3 BCs the least

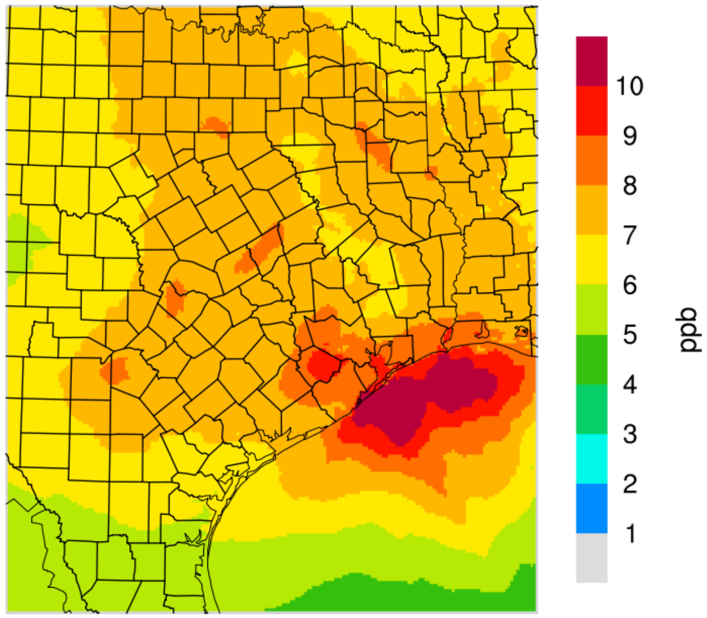
At the sites closest to downtown (Dallas Hinton and Frisco) emission uncertainties are more important than deposition uncertainties

At the outlying sites, deposition uncertainties are more important (Pilot Point) or nearly as important (Italy) as the emission uncertainties

Ozone Uncertainty: 4-km grid

O_3 uncertainty (1σ) Top10 days

ppb



Min= 4.5, Max= 10.9

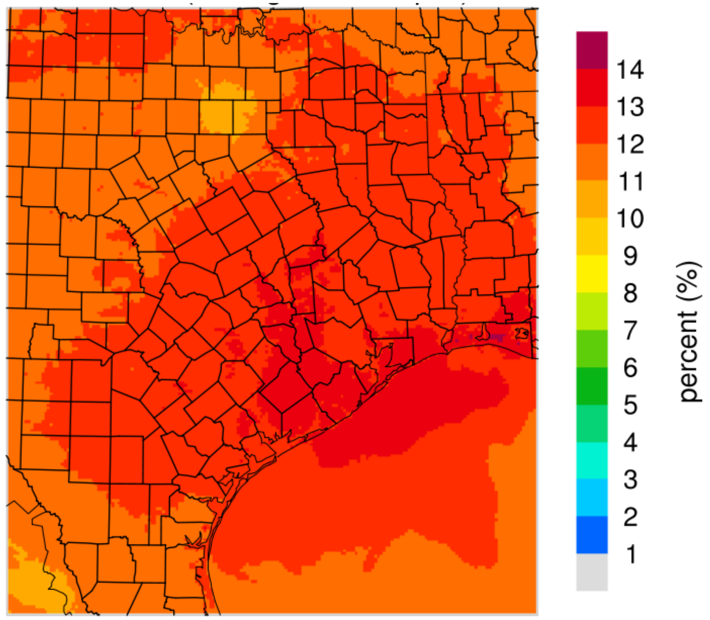
Total O_3 uncertainty (1 σ) is 10-11 ppb in the Gulf near Galveston and 7 ppb - 8 ppb in much of the rest of the domain (Top10 days)

As a percent of the O_3 concentration, the uncertainty is more uniform, 11% - 14% over the whole domain for the Top10 days and 9% to 13% for June average

The uncertainty varies from day to day at a fixed location.

O_3 uncertainty (1σ) Top10 days

percent



Min= 10.6, Max= 14.4

CSA is Computationally Efficient: No Super-computer Required

We simultaneously computed sensitivity to 697 chemical mechanism parameters (1 model run)

- 230 1st order rate constant sensitivities,
- 15 2nd order rate constants sensitivities
- 452 1st order sensitivities to a product stoichiometric coefficient.
- 59,942 individual sensitivities, i.e., the sensitivity of 86 CB6r4 species to 697 parameters
- 630 grid cells selected for analysis

CAMx simulation times

- without CSA required 1.3 hours/day
- with CSA required 2.5 hours/day (factor 1.9 longer)
- 12 CPU cores (Intel E5-2630 V2, 2.5 GHz)

Recommendations

- Use the Hi and Lo mechanisms in simulations with perturbed emissions
- Investigate the region of larger O_3 uncertainty over the Gulf near Houston
- Include uncertainty to inorganic iodine (Ix) emissions from the Gulf (this work is done)
- Include uncertainty due to meteorology
- CSA can identify the least influential chemical mechanism parameters to guide condensation for more efficient models

Extra Slides

Chemical Process Analysis (CPA)

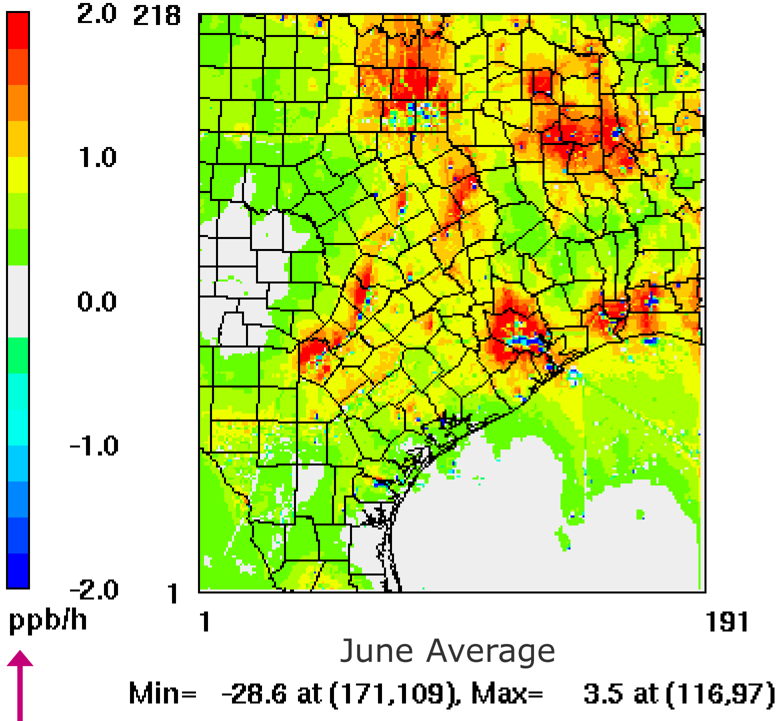
We used CPA to select locations for CSA

Method

- Turn on CPA in CAMx with 3-D output
- Average the CPA output over 4 surface layers (up to 250 m) to dilute the strong forcing by surface emissions
- Average the CPA output for June
- Review maps of CPA output and pick ~10 locations

PO3_netc

Net Ozone Production Rate

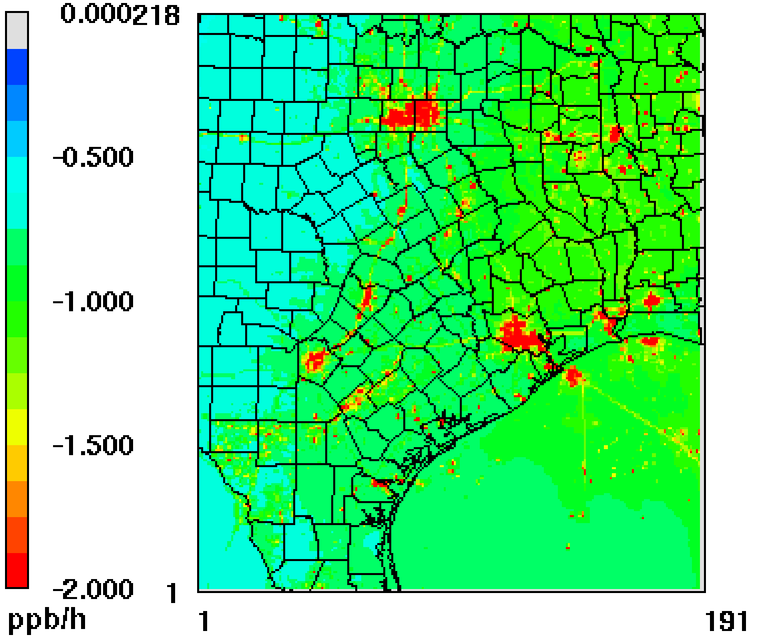


Units always ppb/h or fraction (dimensionless)

O_3 Destruction and Production

Destruction and production can be co-located because monthly average and also they can occur simultaneously. The major urban areas (Houston, Dallas, San Antonio; HGB, DFW, SAT) have localized VOC-sensitive ozone production. NOx-sensitive ozone production is widespread.

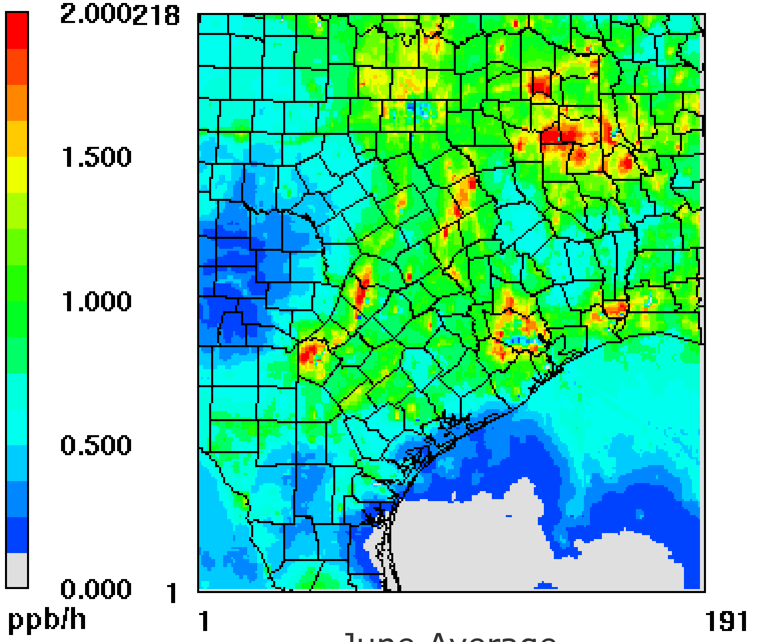
O3_dest



June Average

Min= -28.902 at (172,112), Max= 0.000 at (1,1

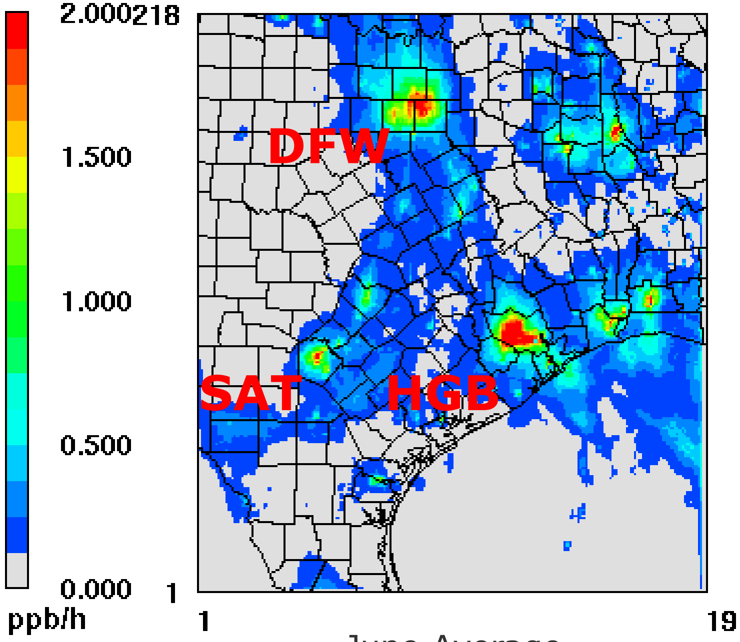
PO3_NOxsns



June Average

Min= 0.000 at (1,1), Max= 2.380 at (128,190)

PO3_VOCsns

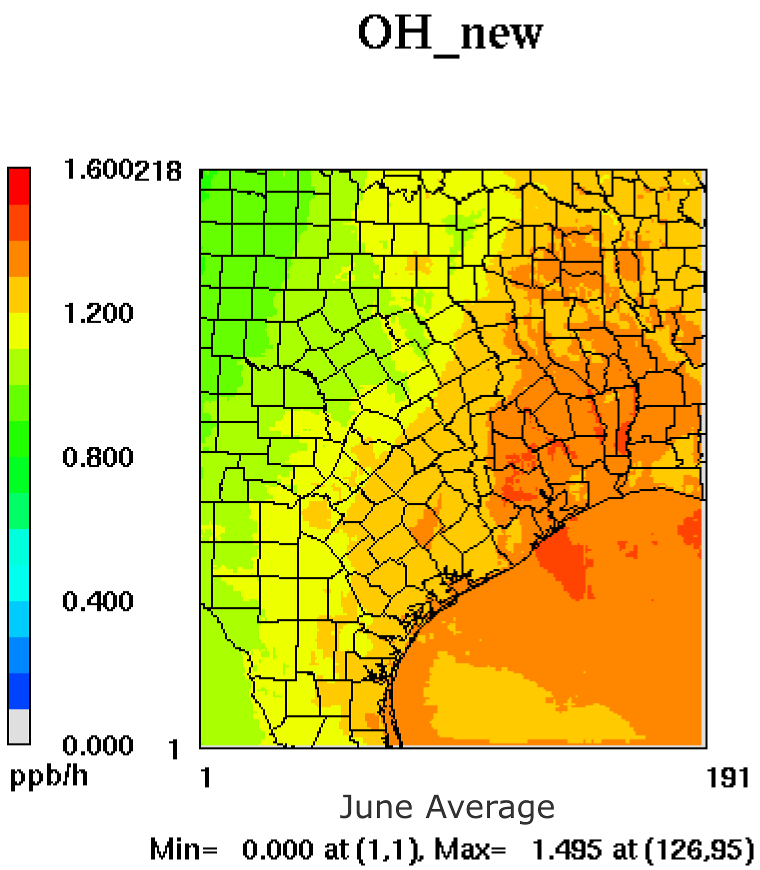


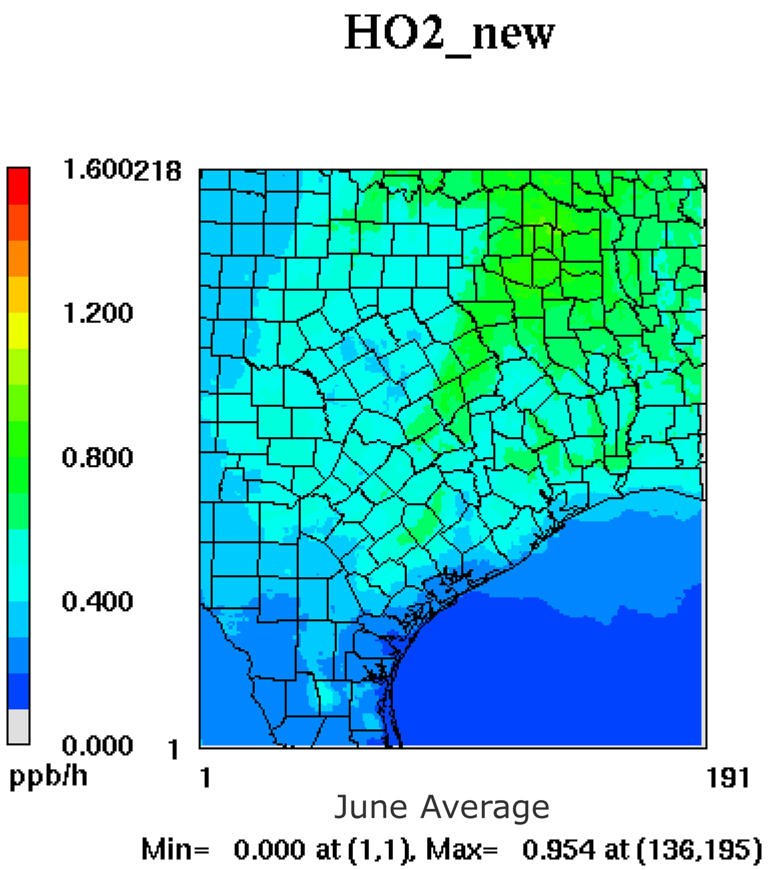
June Average

Min= 0.000 at (1,1), Max= 2.598 at (158,173)

HOx Radical Production

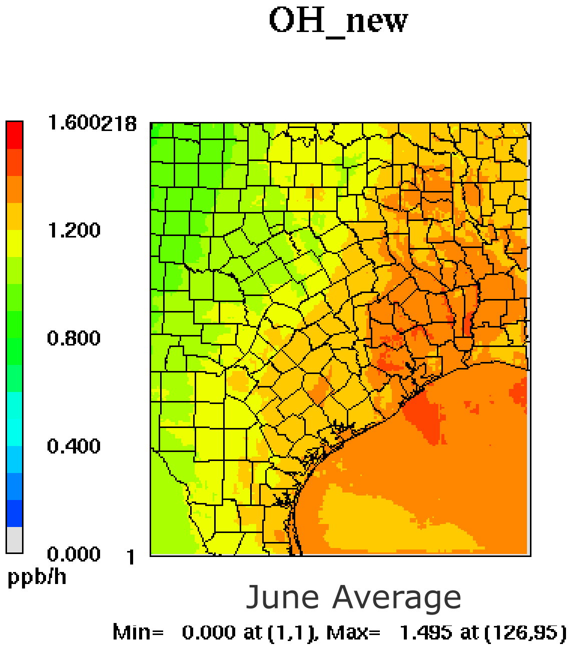
New HOx mainly results from photolysis, but O_3 + alkene reactions are an exception



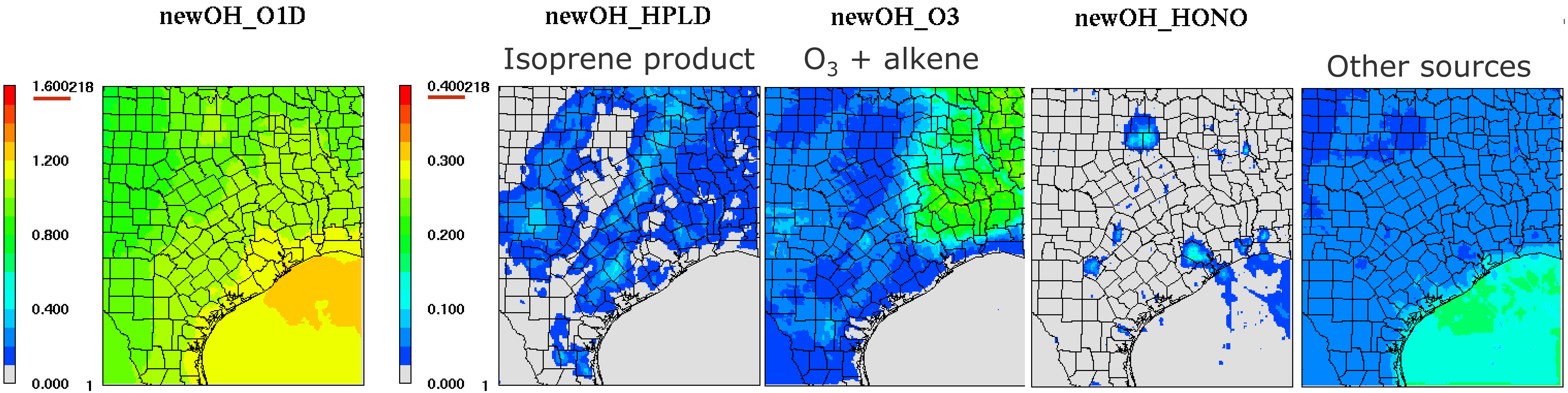


New OH production

Ozone photolysis to O(^{1}D) dominates by far. O_3 + alkene reactions larger with high BVOC emission. HONO restricted mainly to urban areas. HPLD (from isoprene at low NOx) has widespread importance. Over the Gulf, the “other sources” includes iodine chemistry.



June Average



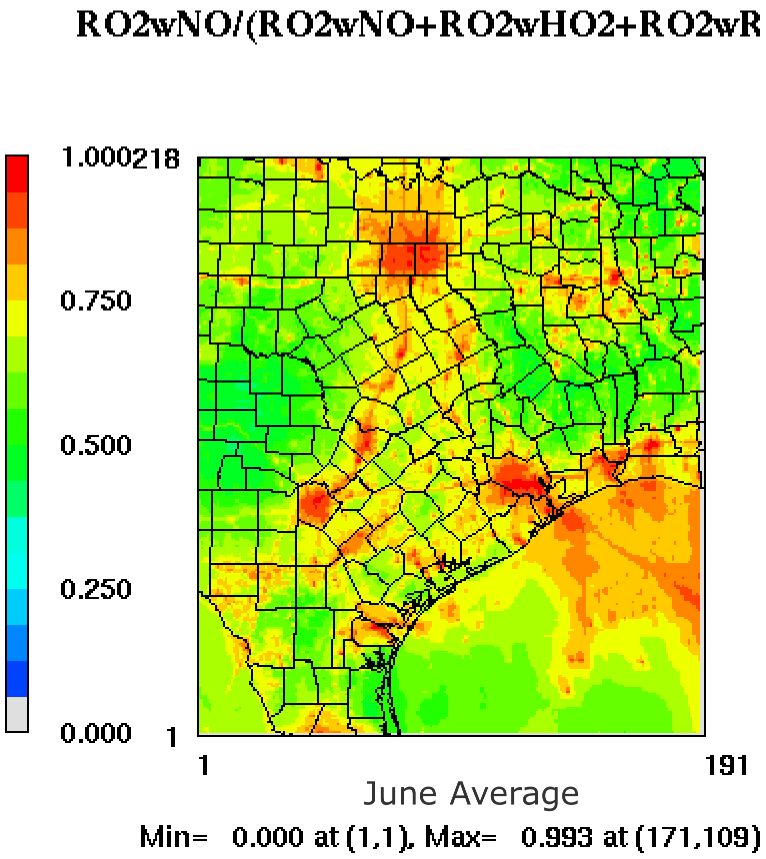
Isoprene product

O_3 + alkene

Other sources

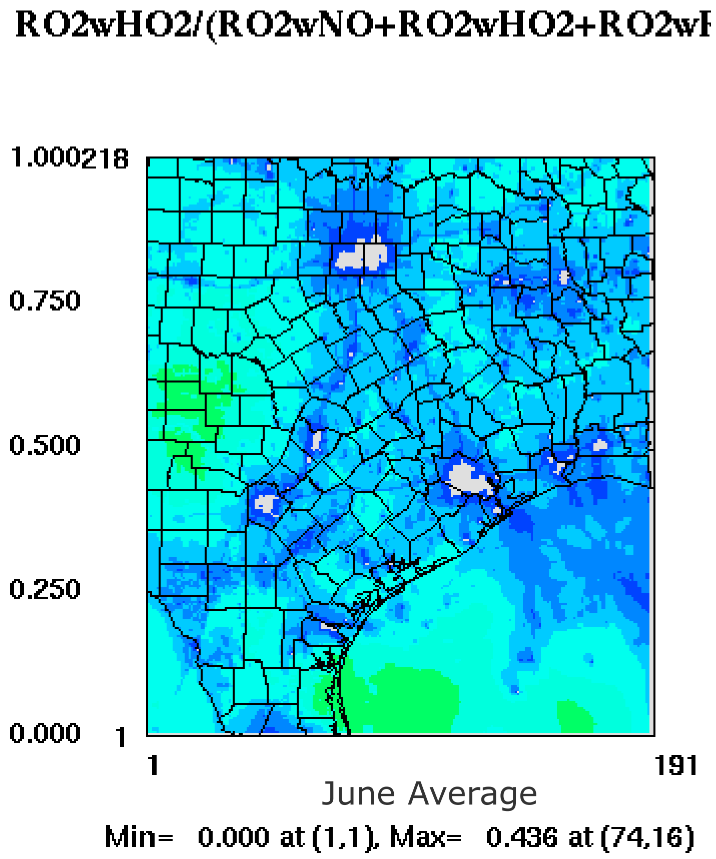
Fate of RO_2 radicals

Three branches available to RO_2: reaction with NO, HO_2, RO_2


RO2wNO/(RO2wNO+RO2wHO2+RO2wR

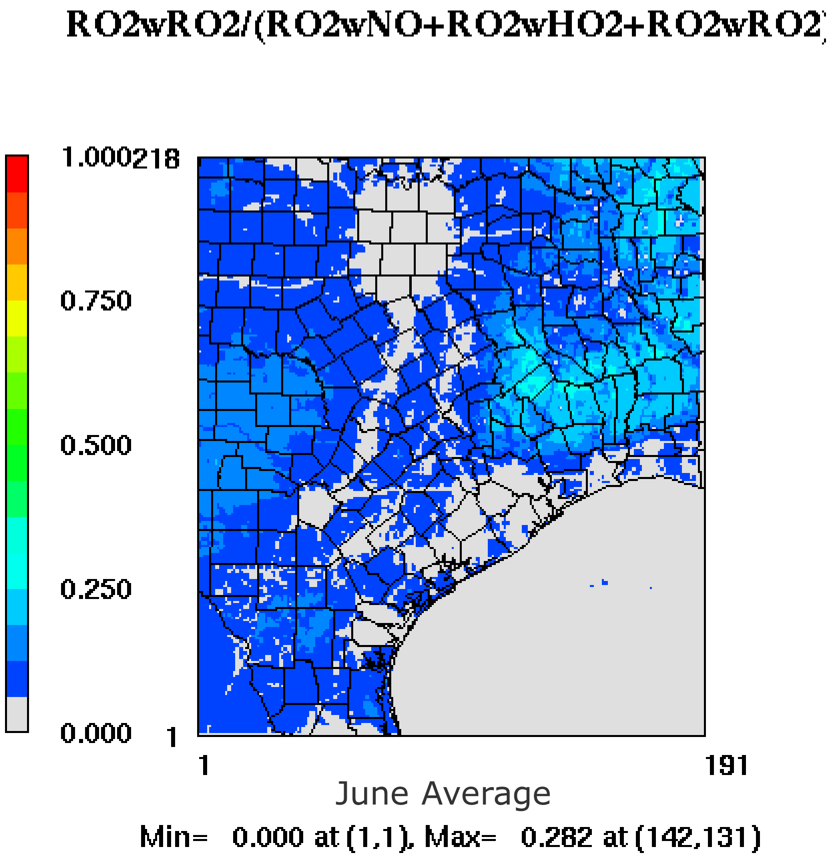
June Average

Min= 0.000 at (1,1), Max= 0.993 at (171,109)


RO2wHO2/(RO2wNO+RO2wHO2+RO2w

June Average

Min= 0.000 at (1,1), Max= 0.436 at (74,16)


RO2wRO2/(RO2wNO+RO2wHO2+RO2wRO2

June Average

Min= 0.000 at (1,1), Max= 0.282 at (142,131)