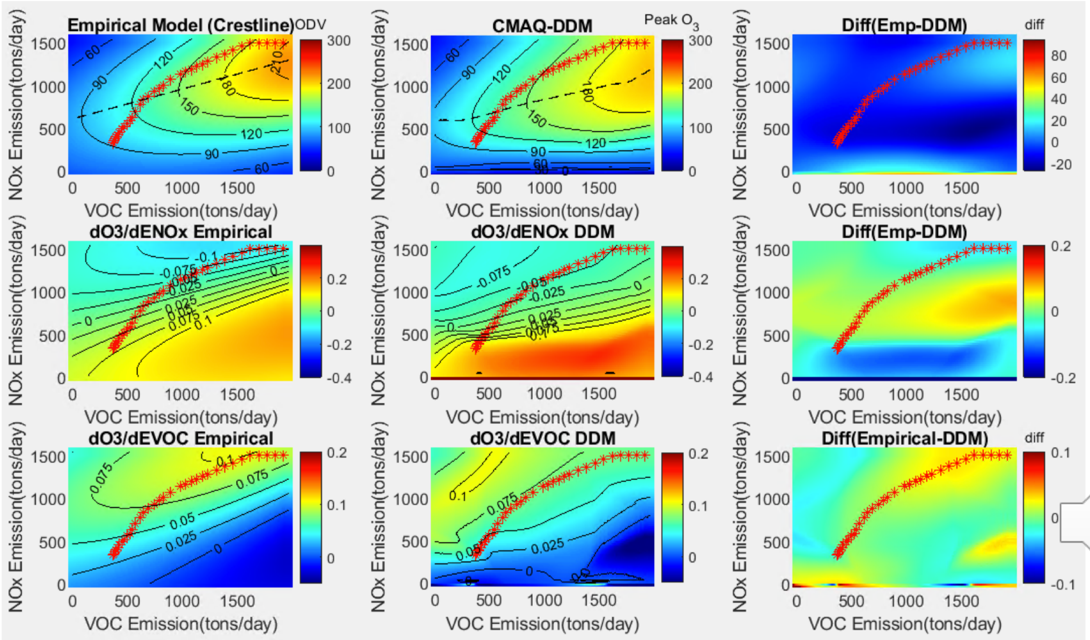
Development of Ozone-NOx-VOC Emissions Isopleth using CMAQ-HDDM and Inverse Distance Weighted Method for Southern California and the Comparison with Empirically-based Method

Yu (Alex) Qian, Zongrun Li, Yongtao Hu, Peter Vasilakos, Talat Odman, Jennifer Kaiser, and Armistead (Ted) Russell

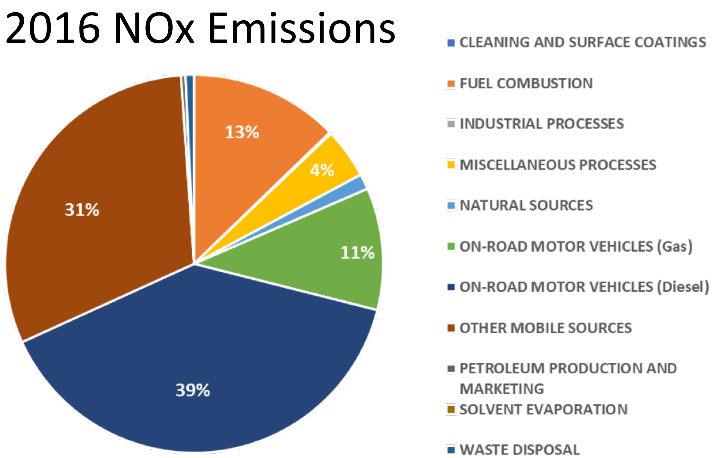
Georgia Institute of Technology

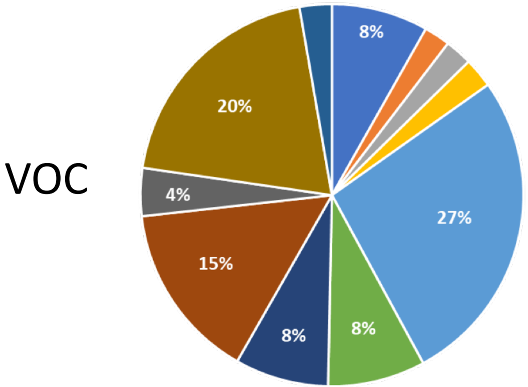
CMAS 2021



A little history

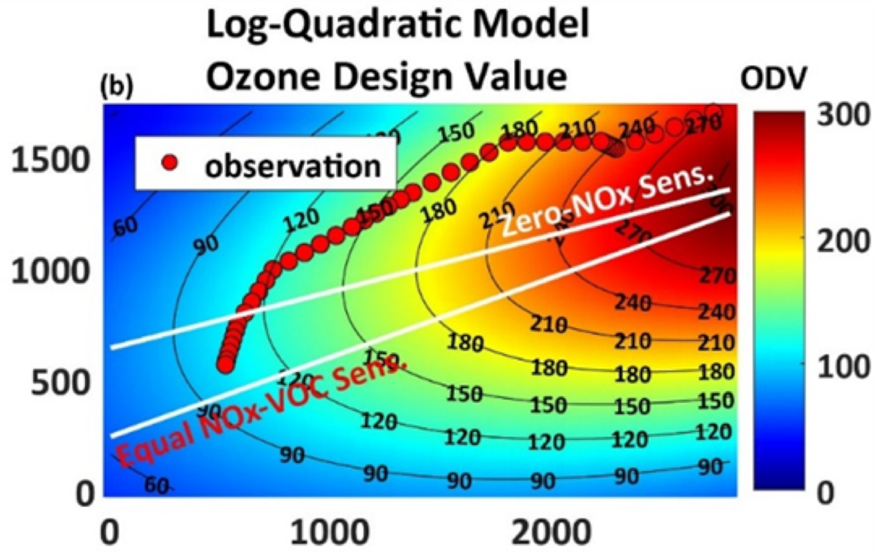
- Ozone in Los Angeles getting up to 490 ppb (1-hr, 1980)
  - Dramatic improvements since then
    - 2019: 137 ppb
- CARB and SCAQMD continue(d) to apply increasingly stringent air pollution controls
  - (Estimated) emissions
    - 1980: ~1600 tons/day of NOx, 2700 tpd VOC
    - 2016: ~400 tpd NOx, ~400 tons/day of VOC)
  - As part of their air quality management planning, SCAQMD applied urban scale air quality model to demonstrate attainment in early 2000s. (standard, then, was 0.12 ppm 1-hr; now 0.070 ppm 8-hr)
    - Still at it: results suggest that another ~60-80% reduction required (down to 80 t/d NOx and 290 t/d VOC)
    - This has major implications for specific sources



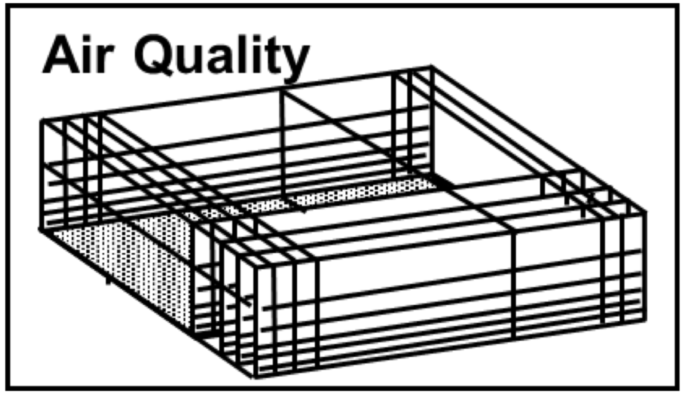


Background and Motivation

- The relationship between ozone concentrations and precursor emissions are key to identifying effective air quality strategies and related regulations.
- Empirical modeling
  - Develop ozone isopleths based on observed ozone and estimated NOx and VOC emissions in the SoCAB
    - Simple regression (nothing fancy: life is a Taylor series, let's just hope the first five terms dominate in this case)
      - Apply to ozone design value, sites individually and all sites together
- Chemical transport modeling
  - Apply CMAQ-HDDM to 1985, 2001, 2011, 2016 and 2028, plus additional edge points
    - Use simulated ozone levels and their sensitivities to develop isopleths



$$\text{Log}_{10}(\text{ODV}\left(E_{NO_x}, E_{VOC}\right) = f\left(E_{NO_x}, E_{VOC}\right)$$



Objectives

- Understand and **quantify trends** in air quality and the **response to emissions controls** in the South Coast Air Basin (SoCAB) of California using empirical analyses.
  - ➢ Develop empirically-derived pollutant isopleths
    - ➢ Earlier work: Qian et al. 2019 ES&T Letters
- Assess the ability of CMAQ (as a state-of-the-science chemical transport model) to capture **observed changes** in ozone over the last 40+ years over the SoCAB.
  - ➢ Develop CMAQ-derived pollutant isopleths
- **Compare** (quantitatively) empirically- and CMAQ-derived emissions sensitivities.
  - ➢ Identify potential reasons for differences
- Forecast **future pollutant concentrations** and their sensitivities to emissions

Initial Work - Basin-wide Empirical Model Developed

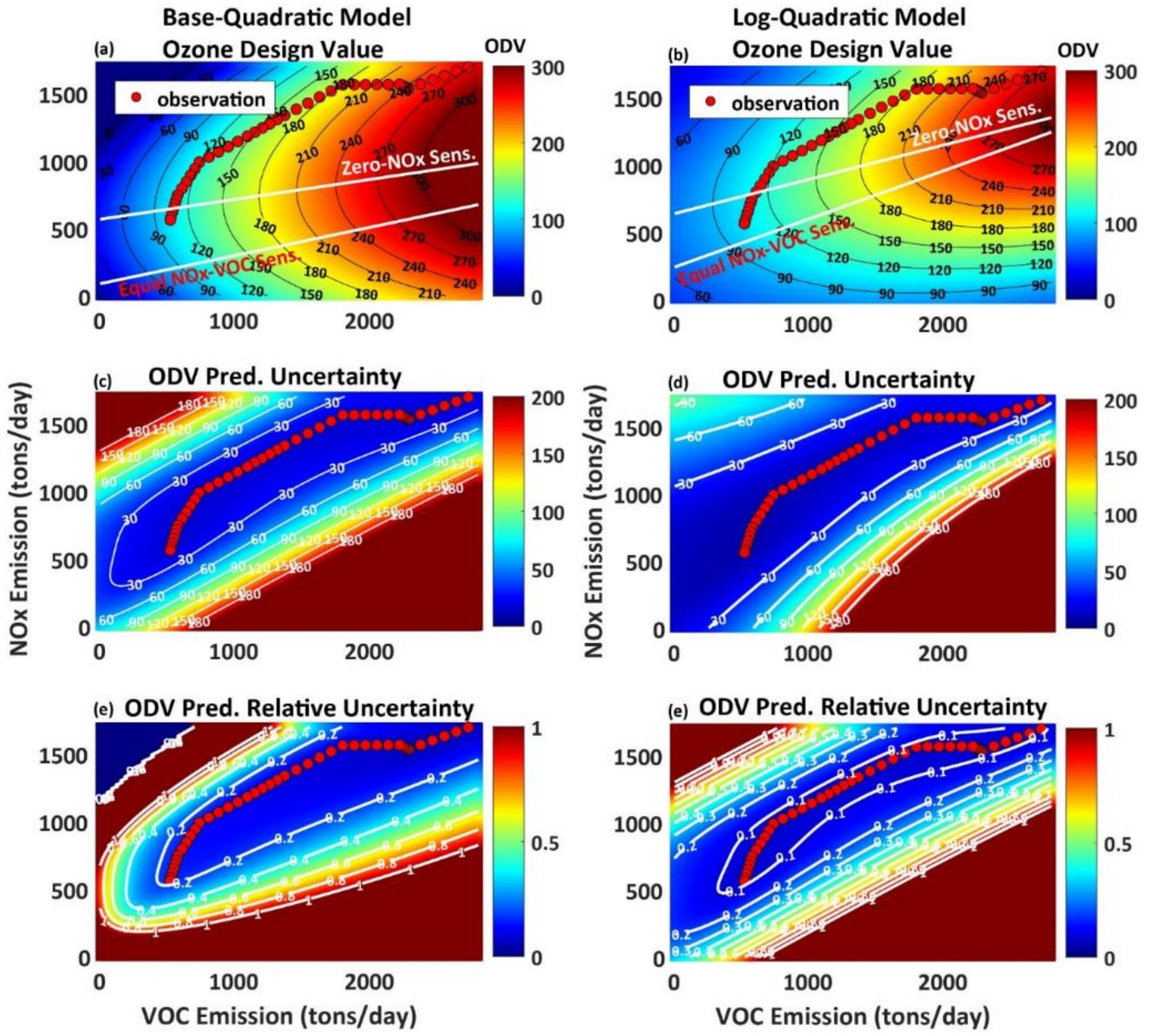
- **Study Domain:** South Coast Air Basin (SoCAB), California
  - Has the highest ozone levels in the US.
    - Driver for emissions controls
  - Long record available
- **Data Sources:**
  - Time Period: 1975 to 2019
  - Basin-wide (without consideration of individual site info.)
  - Observationally-based, yearly ozone design values (ODVs)
    - ODV: average of the fourth highest annual maximum daily average eight-hour ozone concentration (ppb) of three consecutive years.
  - Estimated yearly emission data for NOx and VOC (tons per day).
- **Empirical Isopleth Development Using Regression Modeling**
  - Between yearly ODV and emissions of NOx and VOC (based on Taylor series)

$$\mathrm{ODV}\left(E_{NO_x}, E_{VOC}\right) \text{ or } \mathbf{Log_{10}}\left(\mathrm{ODV}\left(E_{NO_x}, E_{VOC}\right)\right) = f(E_{NO_x}, E_{VOC} \ldots)$$

$$\cong \beta + \alpha_{NO_x} * E_{NO_x} + \alpha_{VOC} * E_{VOC} + \alpha_{NO_x-VOC} * (E_{NO_x} * E_{VOC}) + \alpha_{NO_x^2} * E_{NO_x}^2 + \alpha_{VOC^2} * E_{VOC}^2$$

Results

- **ODV – Emission Relationships**
  - 1^{st} order sensitivities are positive.
  - 2^{nd} order sensitivities are negative.
  - 2^{nd} order sensitives to VOC is smaller
  - Zero-NOx Sens.
    - Further NOx emissions reductions are beneficial.
  - Reconstruct the reasonable isopleth.
- **Model Performance**
  - Good performance with $R^2 > 0.98$.
  - Log-quadratic model performs better.
  - Assessed uncertainties
    - Emissions
- **Low uncertainties** along **"emissions trajectory"**
  - Low uncertainty in sensitivities
  - 56 ppb with 20 ppb uncertainty at (0,0)

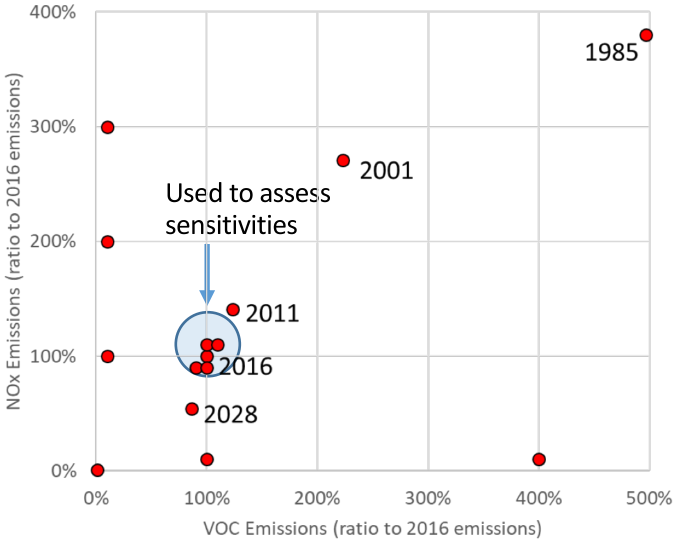


Coef.	Intercept	α_NOx	α_VOC	α_NOx*VOC	α_NOx^2	α_VOC^2	R^2
Base	1.03	0.1347	0.1101	3.5E-05	-1.2E-04	-1.1E-05	0.98
Log	1.72	0.0004	0.0002	1.8E-07	-3.5E-07	-6.9E-08	0.99

Chemical Transport Modeling

- CMAQ-HDDM
  - CMAQ: Community, Multiscale Air Quality Model
    - Regional scale CTM widely used for scientific and regulatory model
      - Used by EPA and the SCAQMD for policy analyses
  - HDDM: High order, decoupled, direct method for providing model sensitivities
    - Simultaneously solves differential equations to get sensitivities of model species to model input.
- Apply CMAQ-HDDM to South Coast Air Basin (SoCAB)
  - 1985, 2011, 2016 and 2028
    - Using two 2016 episode meteorology capturing high periods in 2016
      - June 2 - 4, 2016 and July 20 - 29, 2016 (13 days)
    - 4 km nested in 12 km
    - Using emissions inventory for each individual year
    - **Gridded inventories do not exist for 1985**
      - **Reconstruct using county-specific inventories, VMT and census data**

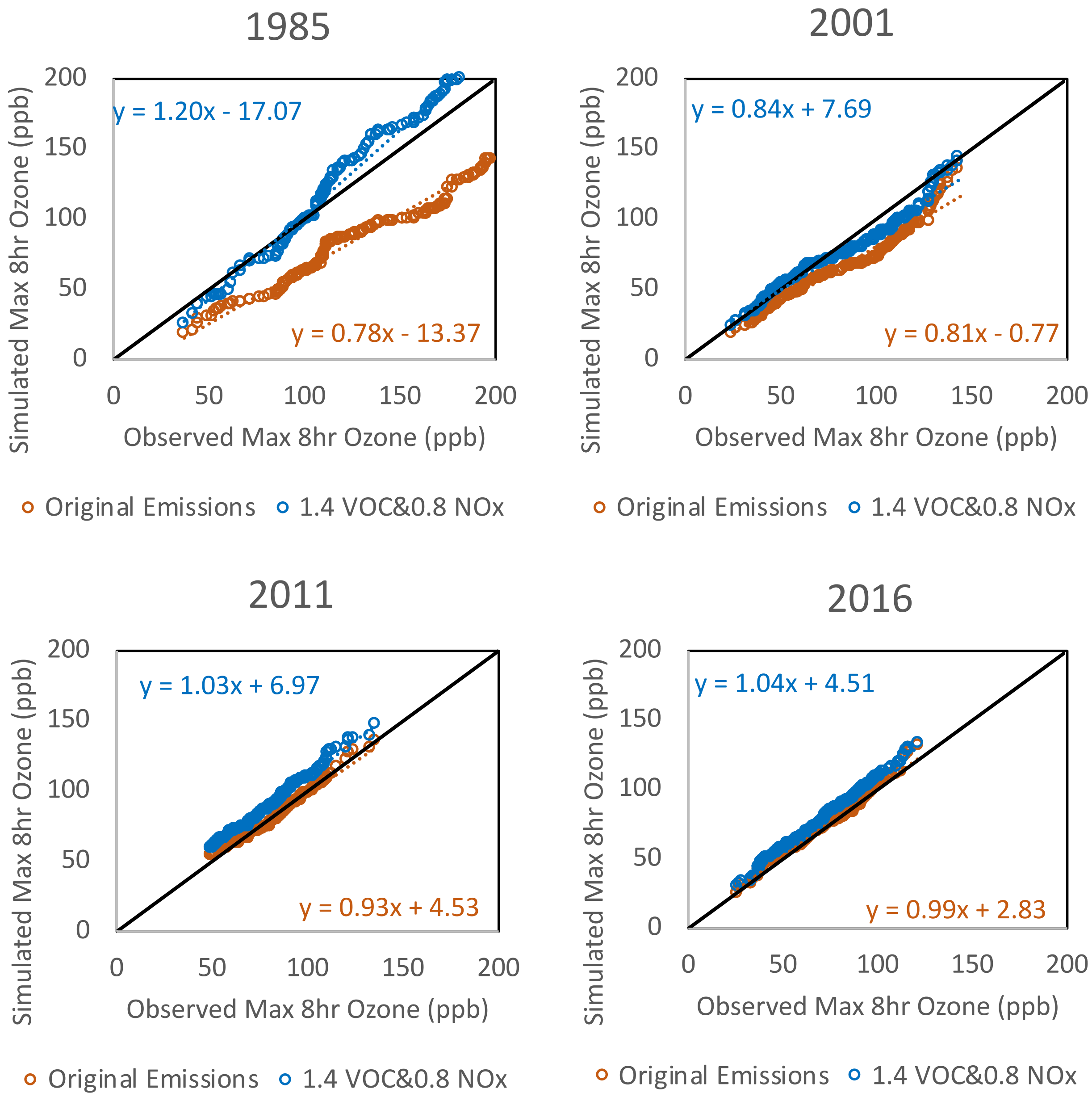
Simulations Conducted



Scale references ratio to 2016 emissions

Evaluation of CMAQ Simulation Results

- Comparison between observed and simulated MDA8 Ozone
  - Rank-ordered comparison to match peak levels
    - 2016 periods chosen based on high ozone
  - With and without the adjustment on emissions (based on sensitivity test)
    - 40% higher VOC emissions
    - 20% lower NOx emissions
- Model performance in 2016 was good
- Increasing bias going back in time
- Suggests potential emissions biases
  - Used modeled sensitivities to estimate potential emissions inventory biases to get better model fit
    - Constant bias and time varying biases
      - Bias suggests +40% VOC, -20% NOx (Evolves over time)



Development of Isopleth Based on CMAQ-HDDM

- 6 HDDM parameters being produced and used:
  - C: Maximum Daily Averaged 8-hour ozone concentrations
  - S_V : first-order sensitivity to anthropogenic VOCs emissions
  - S_N : first-order sensitivity to anthropogenic NO_x emissions
  - S_{VN} : cross second-order sens. to anthropogenic NO_x and VOCs emissions
  - S_{VV} : second-order sens. to anthropogenic VOCs emissions
  - S_{NN} : second-order sens. to anthropogenic NO_x emissions

$$\mathbf{C}(\boldsymbol{V_0}+\boldsymbol{\Delta}\mathbf{V},\boldsymbol{N_0}+\boldsymbol{\Delta N})$$
$$=\mathbf{C}(\boldsymbol{V_0},\boldsymbol{N_0})+(\boldsymbol{\Delta V})\boldsymbol{S_V}(\boldsymbol{V_0},\boldsymbol{N_0})+(\boldsymbol{\Delta N})\boldsymbol{S_N}(\boldsymbol{V_0},\boldsymbol{N_0})+\frac{(\boldsymbol{\Delta V})^2}{2}\boldsymbol{S_{VV}}(\boldsymbol{V_0},\boldsymbol{N_0})+(\boldsymbol{\Delta V})(\boldsymbol{\Delta N})\boldsymbol{S_{VN}}(\boldsymbol{V_0},\boldsymbol{N_0})+\frac{(\boldsymbol{\Delta N})^2}{2}\boldsymbol{S_{NN}}(\boldsymbol{V_0},\boldsymbol{N_0})$$
$$+\frac{(\Delta V)^3}{6}\boldsymbol{S_{VVV}}(\boldsymbol{V_0},\boldsymbol{N_0})+\frac{(\Delta V)^2(\Delta N)}{2}\boldsymbol{S_{VVN}}(\boldsymbol{V_0},\boldsymbol{N_0})+\frac{(\Delta V)(\Delta N)^2}{2}\boldsymbol{S_{VNN}}(\boldsymbol{V_0},\boldsymbol{N_0})+\frac{(\Delta N)^3}{6}\boldsymbol{S_{NNN}}(\boldsymbol{V_0},\boldsymbol{N_0})+\cdots$$

- **Isopleth for each emission scenario is developed by using the Taylor series.**

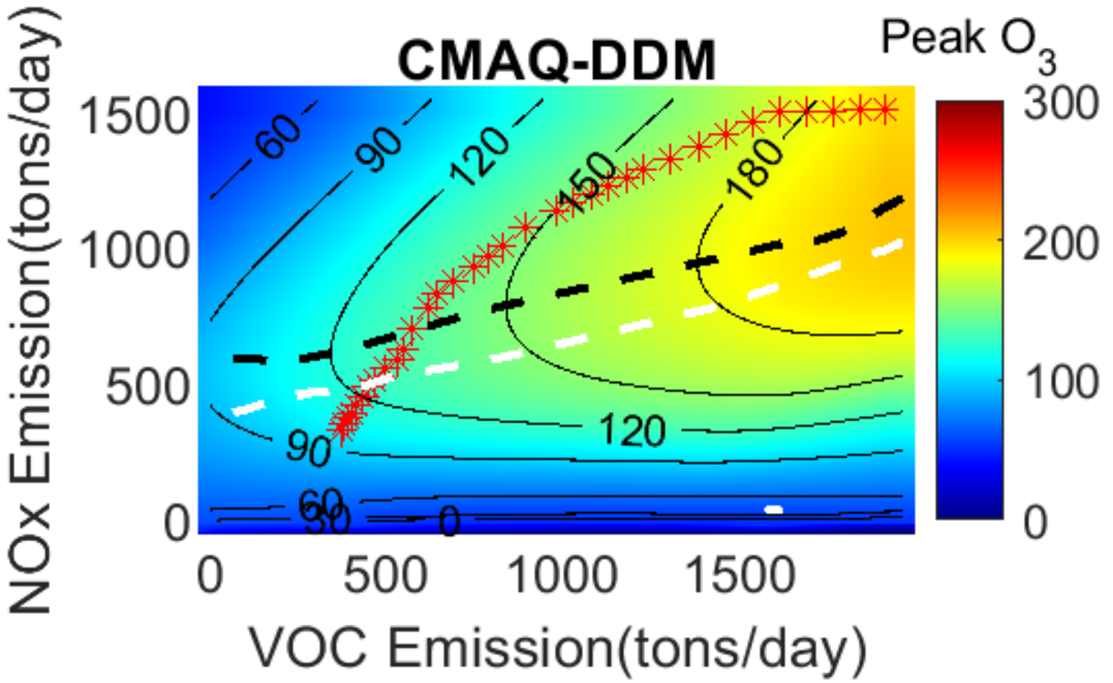
$$w_i=\frac{\frac{1}{\sqrt{d_i}}}{\sum_{i=1}^{n}\frac{1}{\sqrt{d_i}}}$$

- **Generate a combined isopleth by integrating all of the single isopleth**
  - Square-root Inverse distance weighted method

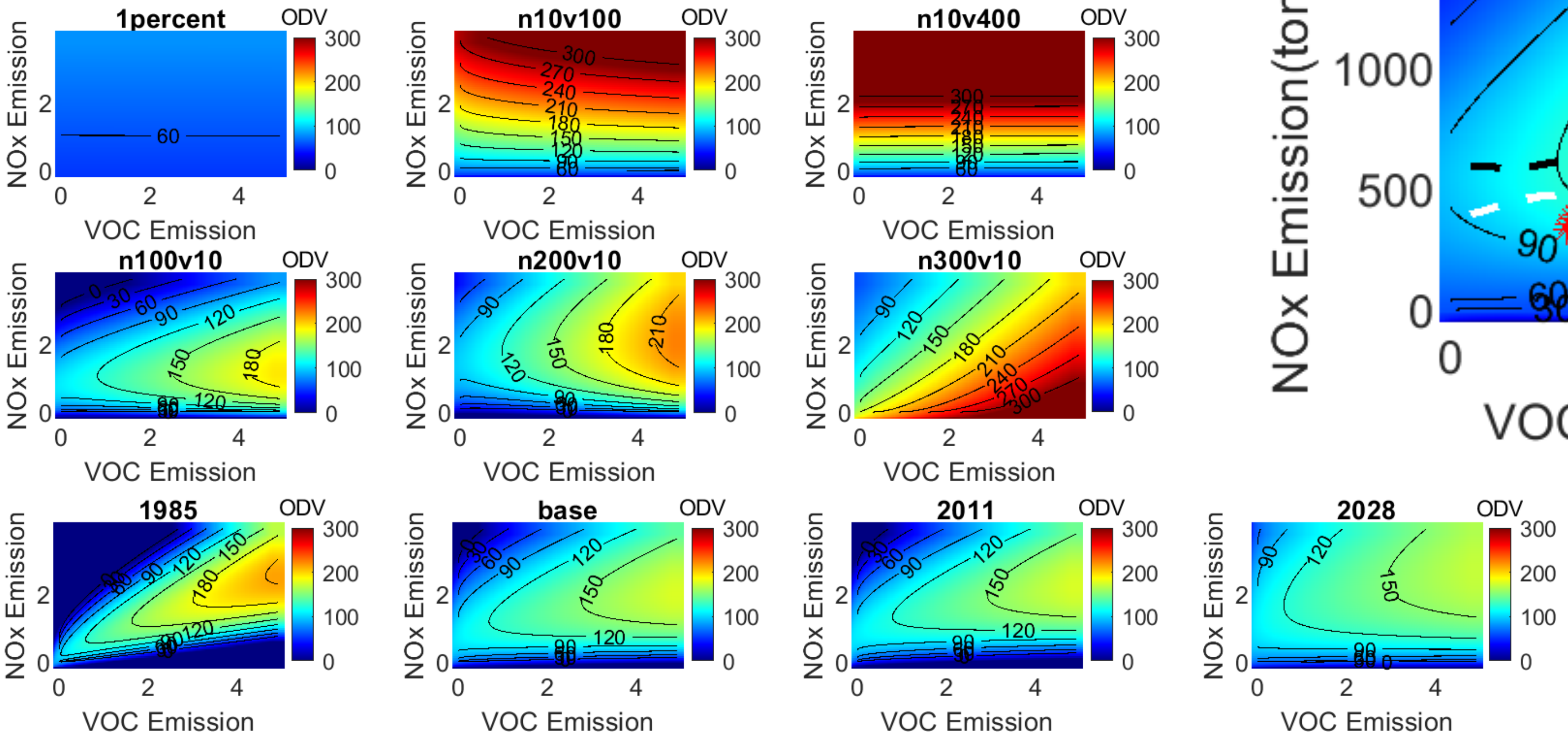
$$O_{3\ combine}=\sum_{i=1}^{n} w_i * O_{3\ i}$$

Square-root Inverse Distance Weighted Method (SRIDW): Isopleth construction (for Crestline)

Combined Isopleth using inverse distance weighted extrapolation.

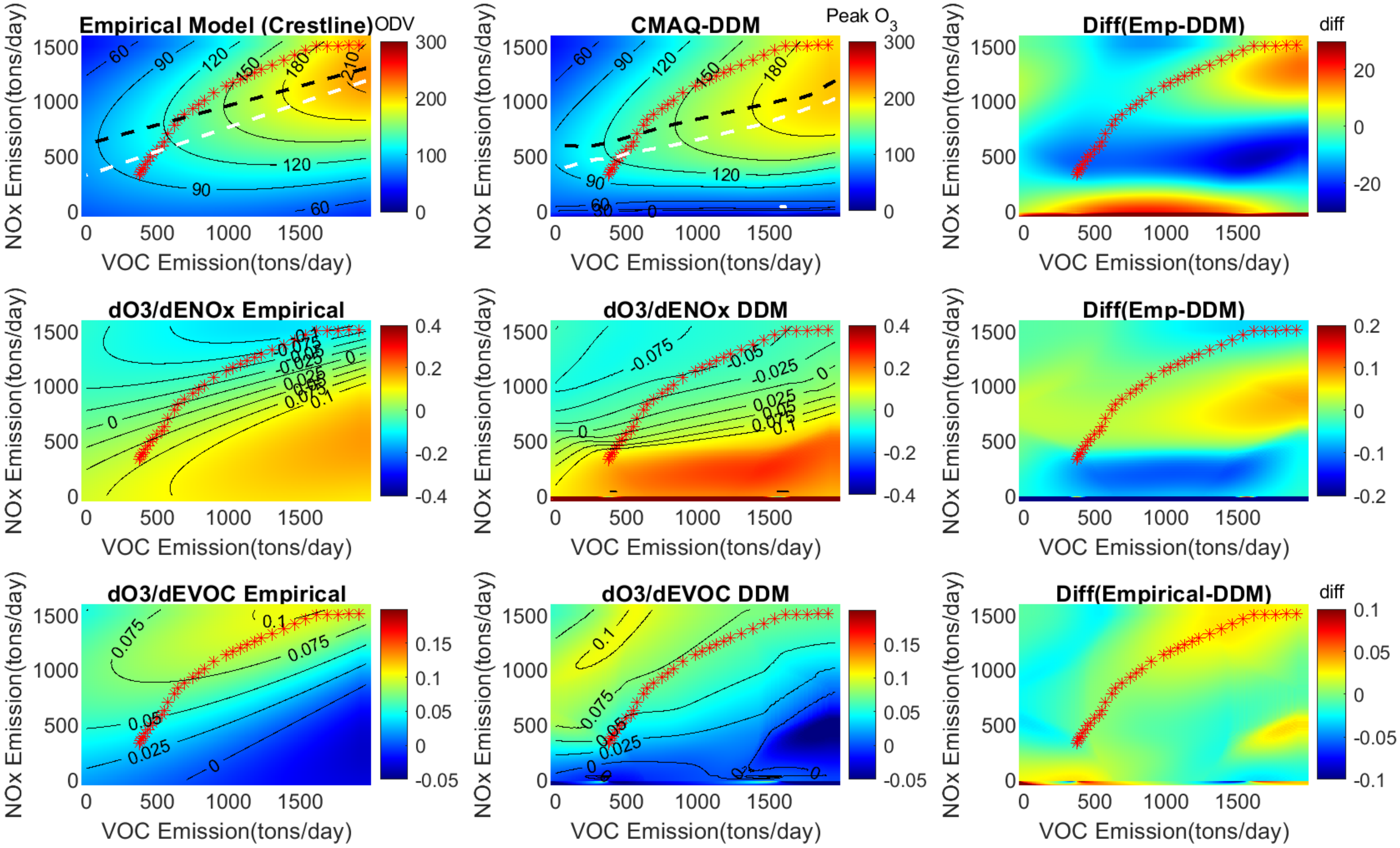


Isopleths generated by each simulation



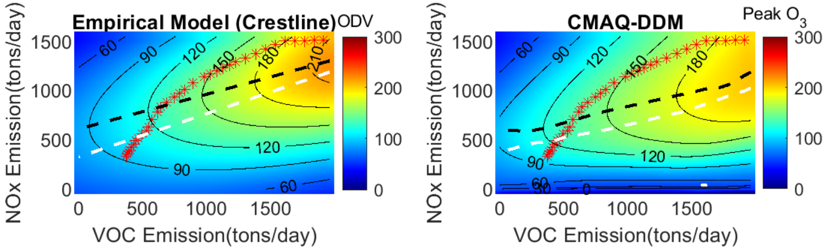
x,y axis are percentage emissions compared to 2016 base case

Comparison of Ozone and sensitivity Isopleths: Empirical v.s. CMAQ

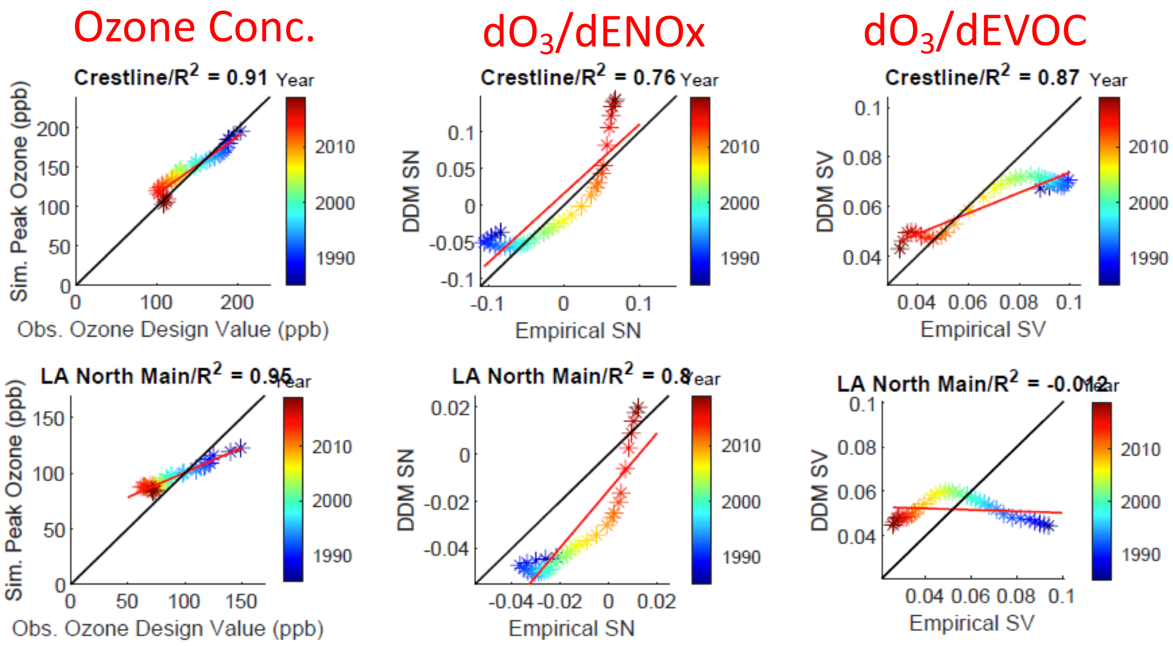


Emissions Trajectory Analysis

- **Comparison of historical ozone and sensitivity levels between simulated and observed (empirically estimated) from 1980s to 2019 along estimated emissions trajectory**
- **Use results along estimated emissions trajectory because**
  - Least uncertainty in empirical model results
  - Most regulatory relevant
  - Helps in comparing model results (observed to modeled ozone, etc.) in relationship to the sensitivities



- **Empirical vs. CMAQ ozone levels compare reasonably well around the basin**
- **Sensitivities show more significant differences**
  - VOC sensitivities
    - Modeled are flatter
  - NOx sensitivities
    - Relationship not at all constant
  - **Differences in sensitivities are what is of importance**



SN: first-order sensitivity to anthropogenic NO_x emissions (ppb/(tons/day));
SV: first-order sensitivity to anthropogenic VOCs emissions (ppb/(tons/day)).

Correlation between CMAQ and Spatial-Empirical model

R^2	O3	SN	SV
Azusa	0.92	0.69	0.13
Glendora	0.95	0.58	0.22
West LA	0.86	0.76	0.78
LA North Main	0.95	0.81	0.02
Reseda	0.69	0.73	0.17
Burbank	0.79	0.94	0.44
Pico Rivera	0.85	0.76	0.00
Pomona	0.78	0.72	0.49
Pasadena	0.92	0.69	0.00
Long Beach	0.82	0.64	0.07
LAX	0.60	0.24	0.51
Santa Clarita	0.79	0.73	0.16
Anaheim	0.81	0.84	0.00
Mission Viejo	0.75	0.77	0.20
La Habra	0.75	0.87	0.00
Banning	0.67	0.76	0.07
Perris	0.79	0.78	0.07
Riverside	0.86	0.82	0.34
Lake Elsinore	0.88	0.76	0.20
Crestline	0.91	0.76	0.87
Upland	0.86	0.73	0.48
Fontana	0.87	0.73	0.49
Redlands	0.87	0.79	0.62
San Bernardino	0.83	0.80	0.47
Overall	0.82	0.74	0.28

Summary

- Observations provide a viable method to develop ozone isopleths in LA
  - **Strengths**
    - This provides the actual response to (estimated) emissions, very good fit, low uncertainty along the trajectory of observations, can be used to assess model responses.
  - **Weaknesses**
    - Uses estimated emissions (+/-), less certain when projecting (but not bad).
- CTM-based isopleths developed using concentration+sensitivity method
  - **Compared to observations**
    - Ozone compares well, sensitivities less well
      - Sensitivities are really important for air quality planning (and science)
    - Biases in ozone, ozone sensitititivities, NOx and CO suggest a ~40% low bias in estimated VOC emissions, 20% high bias in estimated NOx emissions
      - Volatile consumer products might be part of this.
    - Is the main issue emissions estimates? Lot more to be investigated.
- Boundary Conditions (BCs) Impact on Simulation (Not shown today)
  - For 2016 case, simulated ozone concentrations and sensitivities agreed well by using static and hemispheric BCs.
  - BCs would bring more uncertainty when emissions are getting lower, especially when estimating background ozone levels.

Thanks!

Questions?