WASHINGTON STATE UNIVERSITY

Application of Direct Decoupled Method (DDM) for the TCOPS field study period

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Sensitivity Methods

How the model results change if we change a parameter?

Brute Force Method:
Run the model for the base case and after changing the parameter. Quickly become inefficient as a large number of sensitivity coefficients need to be computed

Decoupled Direct Method(DDM):
Sensitivity equations are derived directly from model equations and are solved decoupled from concentrations.

Decoupled Direct Method (DDM)

DDM-3D calculates sensitivity coefficients by applying the same numerical algorithms and operator splitting used to calculate concentrations

atmospheric diffusion equation:

$$\frac{\partial C_i}{\partial t} = -\nabla(\mathbf{u}C_i) + \nabla(\mathbf{K}\nabla C_i) + R_i + E_i$$

First-order sensitivity coefficients :

$$\mathbf{S}_j^{(1)} = \frac{\partial \mathbf{C}}{\partial \epsilon_j}, \quad \frac{\partial S_{ij}^{(I)}}{\partial t} = -\nabla(\mathbf{u}S_{ij}^{(I)}) + \nabla(\mathbf{K}\nabla S_{ij}^{(I)}) + E_i + \mathbf{J}_i\mathbf{S}_j^{(I)}$$

second-order sensitivity coefficients:

$$\mathbf{S}_{j,k}^{(2)} = \frac{\partial^2 \mathbf{C}}{\partial \epsilon_j \partial \epsilon_k}, \quad \frac{\partial S_{ij}^{(2)}}{\partial t} = -\nabla(\mathbf{u}S_{ij}^{(2)}) + \nabla(\mathbf{K}\nabla S_{ij}^{(2)}) + \mathbf{J}_i\mathbf{S}_j^{(2)} + \mathbf{J}^*_{S^{(I)}}\mathbf{S}_j^{(I)}$$

U: the three-dimensional wind field
K: the turbulent diffusivity tensor
Ri and Ei: the chemical reaction rate and emission rate of species i
Ji: the ith row vector in the Jacobian matrix
J (Jik = ∂Ri/∂Ck): photochemical interactions between species
Ei: the unperturbed emission rate of the sensitivity parameter.

- For *j* equal *k*, the $\mathbf{S}^{(2)}$ represents the local curvature of the concentration-parameter relationship.
- For *j* not equal *k*, $\mathbf{S}^{(2)}$ represents a "cross-sensitivity" interaction between the sensitivities to two different parameters, *pj* and *pk*.

Sensitivities Magnitude and Sign

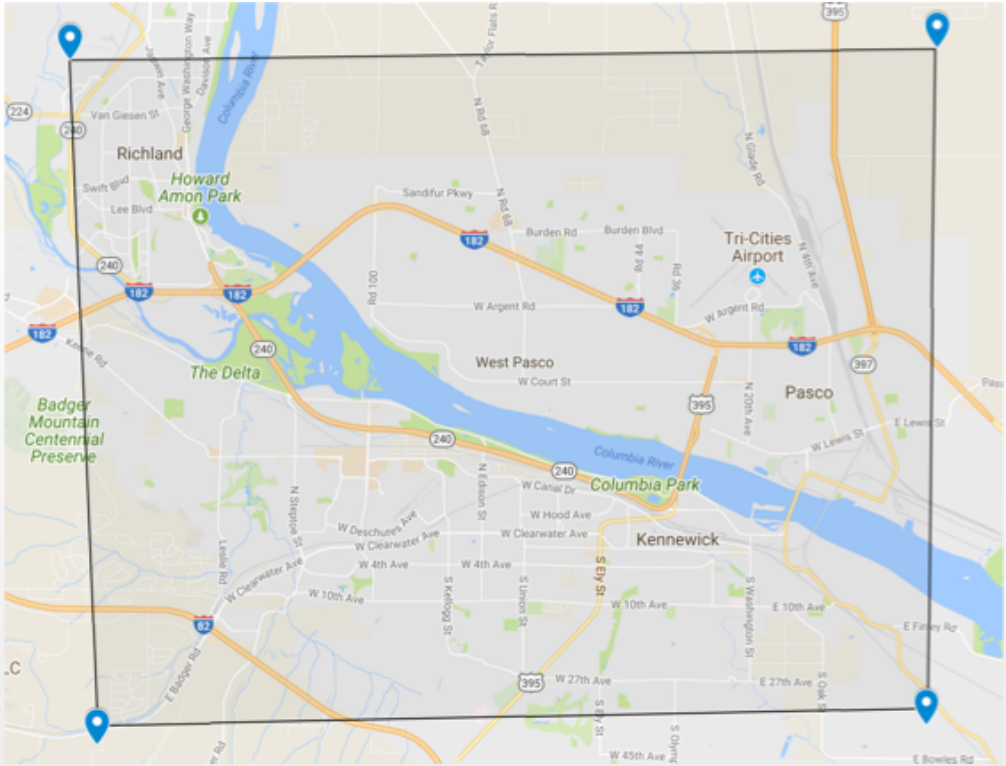
- **First-order sensitivity coefficients** represent the responsiveness of ozone to infinitesimal perturbations, generally accepted as being accurate for up to 30% change in input parameter.
- To project to larger perturbations away from a base case we use **second-order sensitivity coefficients**

- **Positive first-order sensitivity: Increasing** the parameter will **increase** ozone concentration and decreasing the parameter will decrease ozone concentration

- **Negative first-order sensitivity**: **Increasing** the parameter will **decrease** ozone concentrations and decreasing the parameter will increase ozone concentration

- **Negative second-order sensitivity:** reflecting the **concave** response of ozone to the parameter. slope of concentration-emission response curve decreases with increasing emissions.
- **Positive second-order sensitivity:** reflecting the **convex** response of ozone to the parameter. slope of concentration-emission response curve increases with decreasing emissions.
- Greatest nonlinearity occurs where chemical regime changes and hence spatial gradients in first-order sensitivity are large.

Tri-cities Box



DDM was run for this box of 20 grid cells for:

- all sources, and
- mobile sources

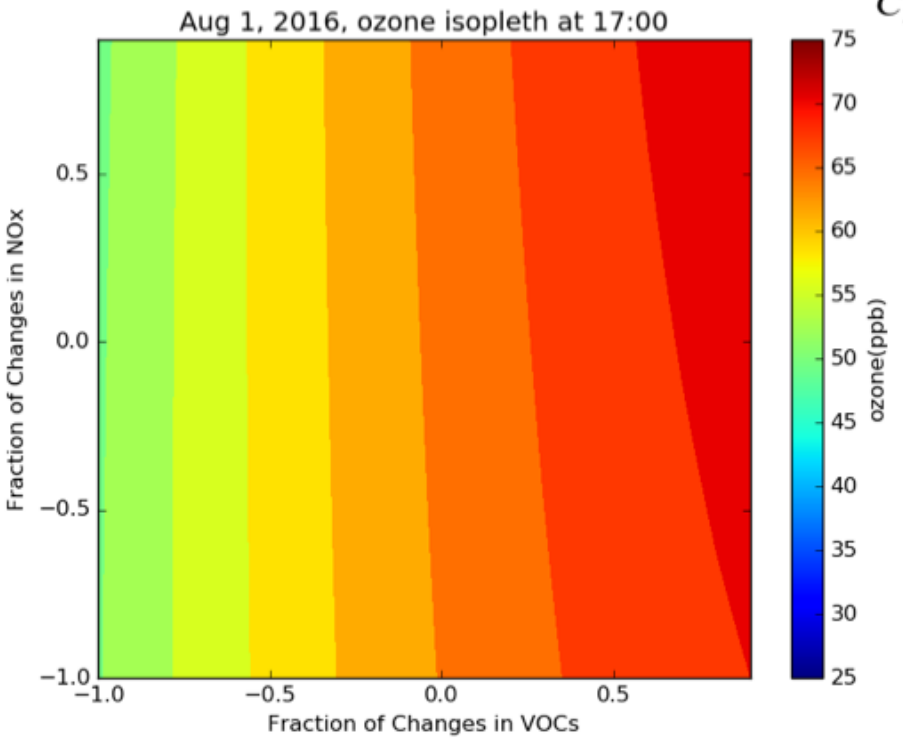
Taylor Expansion

- An important application of the sensitivity coefficients is projection from base case scenario.
- Tylor expansion is the main way of such projection:

$$C_{\varepsilon_j,\varepsilon_k} \approx C_{base} + \varepsilon_j S_j^{(1)} + \frac{1}{2}\varepsilon_j^2 S_j^{(2)} + \varepsilon_k S_k^{(1)} + \frac{1}{2}\varepsilon_k^2 S_k^{(2)} + \varepsilon_j \varepsilon_k S_{j,k}^{(2)}$$

Cbase: ozone concentration in base case
Sj(1): first-order O3_VOC sensitivity coefficient
Sj(2): second-order O3_VOC sensitivity coefficient
Sk(1): first-order O3_NOx sensitivity coefficient
Sk(2): second-order O3_NOx sensitivity coefficient
Sjk(2): cross-sensitivity term to account for interactions between the impacts of changing the two parameters

Ozone Isopleths



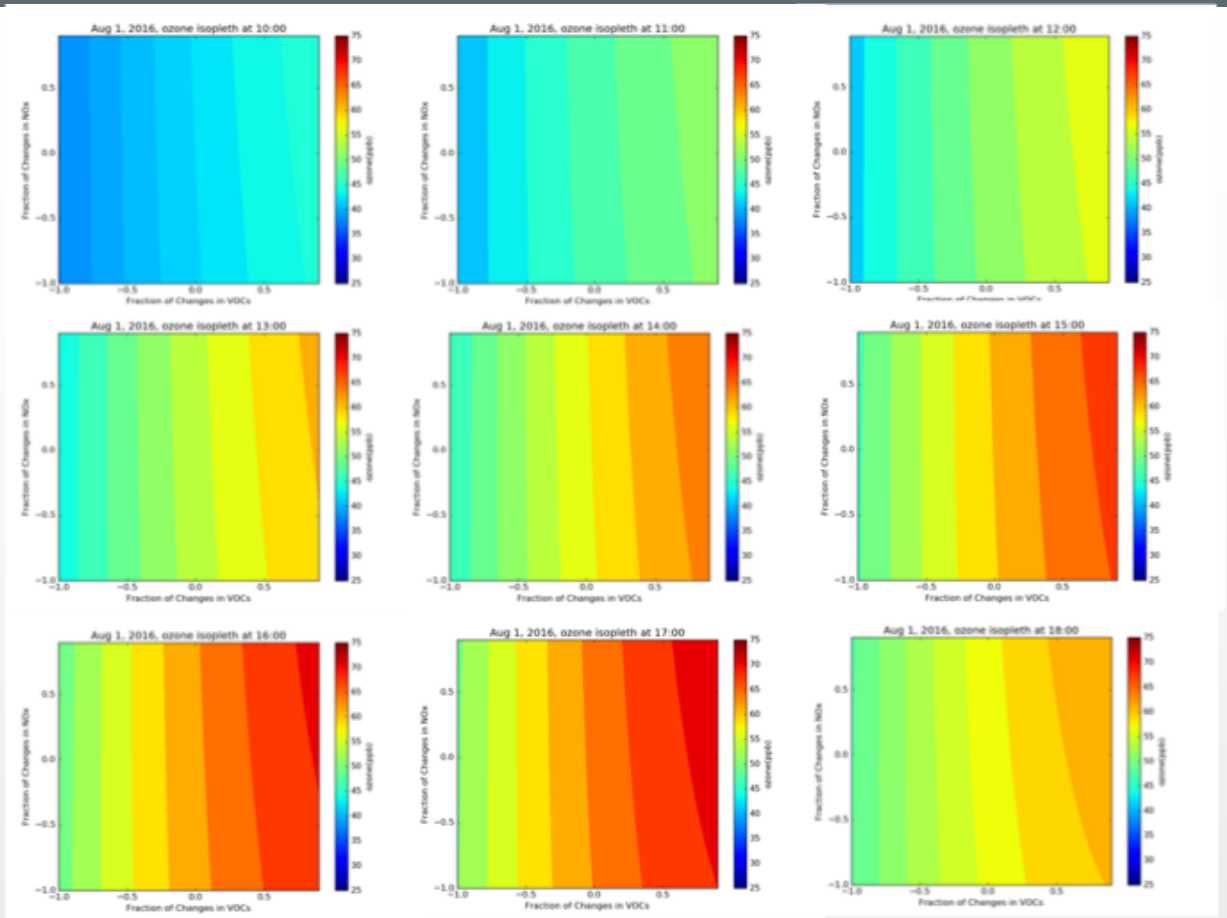
$$C_{\varepsilon_j,\varepsilon_k} \approx C_{base} + \varepsilon_j S_j^{(1)} + \tfrac{1}{2}\varepsilon_j^2 S_j^{(2)} + \varepsilon_k S_k^{(1)} + \tfrac{1}{2}\varepsilon_k^2 S_k^{(2)} + \varepsilon_j \varepsilon_k S_{j,k}^{(2)}$$

$-1<\varepsilon_j<1$
$-1<\varepsilon_k<1$

$\varepsilon_j=\varepsilon_k=0$	base case
$\varepsilon_j=1$	double VOCs
$\varepsilon_k=1$	double Nox
$\varepsilon_j=-1$	no VOCs
$\varepsilon_k=-1$	no NOx

ozone isopleths at the location of the peak ozone at 17:00, 1 August 2016

Ozone Isopleths



Hourly ozone isopleths at the location of the peak ozone on 1 August 2016

Zero-Out Source Contribution(ZOC)

- The zero-out source contribution (ZOC) of an emitter:
  The magnitude of the reduction in concentrations that would occur if that source did not exist
- Second-order approximations of ZOC can be computed by considering $\Delta\varepsilon j = -1$ in Taylor expansion equation

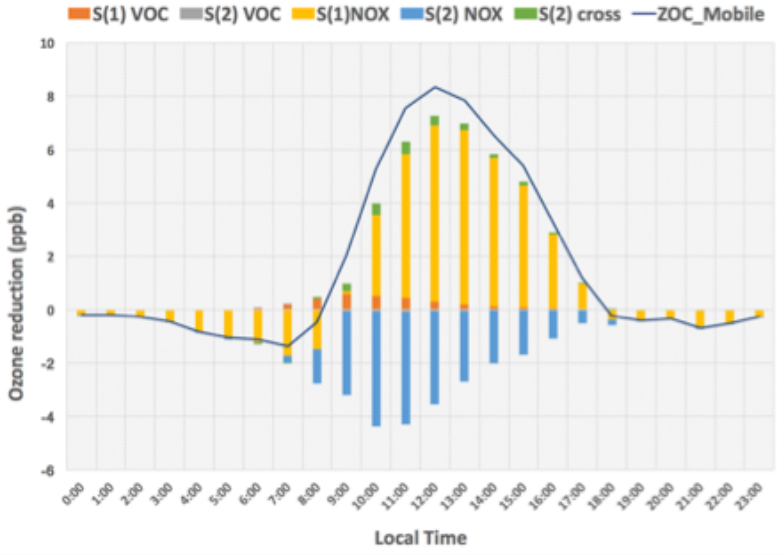
$$ZOC(P_j) \cong \mathbf{C}_0|_{p_j=P_j} - \mathbf{C}_j|_{\{p_j=0;i.e.,\Delta\epsilon_j=-1\}} \approx \mathbf{S}_j^{(1)} - \frac{1}{2}\mathbf{S}_{j,j}^{(2)}$$

$$ZOC(P_j + P_k) \approx (\mathbf{S}_j^{(1)} - \tfrac{1}{2}\mathbf{S}_{j,j}^{(2)}) + (\mathbf{S}_k^{(1)} - \tfrac{1}{2}\mathbf{S}_{k,k}^{(2)}) - \mathbf{S}_{j,k}^{(2)}$$

ZOC j ZOC k cross term

Mobile Source Contribution in Ozone Concentration in Tri-cities

July 29, 2016
Maximum ozone at 13:00 = 55.8ppb



Reduction in ozone concentrations due to zeroing out mobile sources at a hotspot inside the Tri-Cities box(line) and a decomposition of zero-out source contribution of mobile source NOx and VOC emissions

Benton and Franklin County Emission Inventory, on-road vehicles:

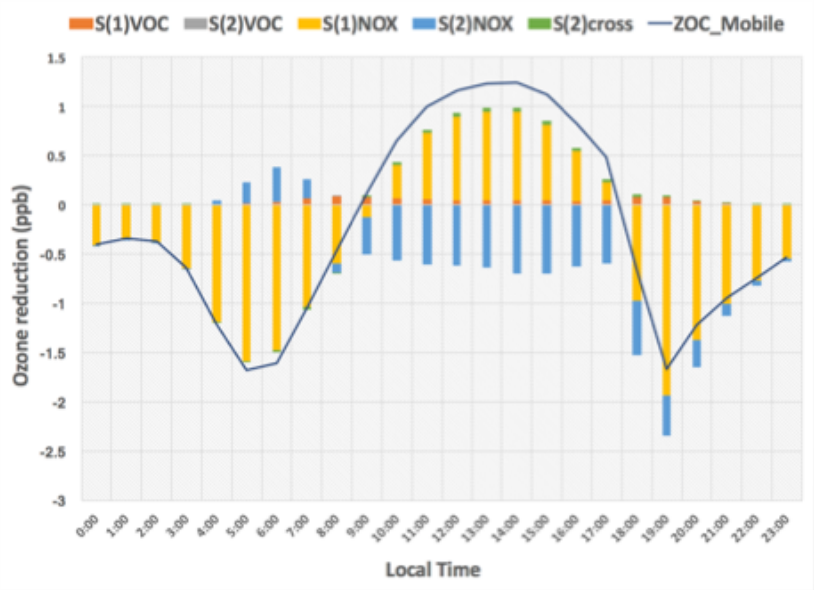
- 50% NOx
- 21% VOCs (excluding biogenic VOC)

- First-order VOC and NOx sensitivities are both positive, lowering their emissions can lower 8-hour ozone
- The response of Tri-Cities ozone to NO*x* is highly nonlinear, with daytime values reflecting a concave response. (This reflects that when NO*x* emissions are decreased, the slope of concentration-emission response curve increases.)
- The situation changes at night, when ozone can be titrated by NO*x* in the absence of sunlight and thus displays negative sensitivity.

Mobile Source Contribution in Ozone Concentration in Tri-cities

Spatially-average maximum ozone over Tri-cities happens at 16:00, and is equal to 39.4ppb

July 27- August 7, 2016
averaged over the box,



Episode-average reduction in ozone concentrations due to zeroing out mobile sources in Tri-Cities box(line) and a decomposition of zero-out source contribution of mobile source NOx and VOC emissions

Conclusions

- Based on DDM and Taylor expansion results, the ozone isopleths were created and showed that the Tri-cities region is highly VOC-limited.

- Eliminating mobile sources inside the 20-cell box can decrease ozone up to 8ppb on some hotspots. However, on average, it only decreases ozone up to 1.3ppb during the day time and will increase ozone up to 1.6ppb during dark time.

Suggestions for future experiments?