Non-linearity in Atmospheric Response: A Sensitivity Analysis Approach

Using

High-Order Decoupled Direct Method (HDDM)

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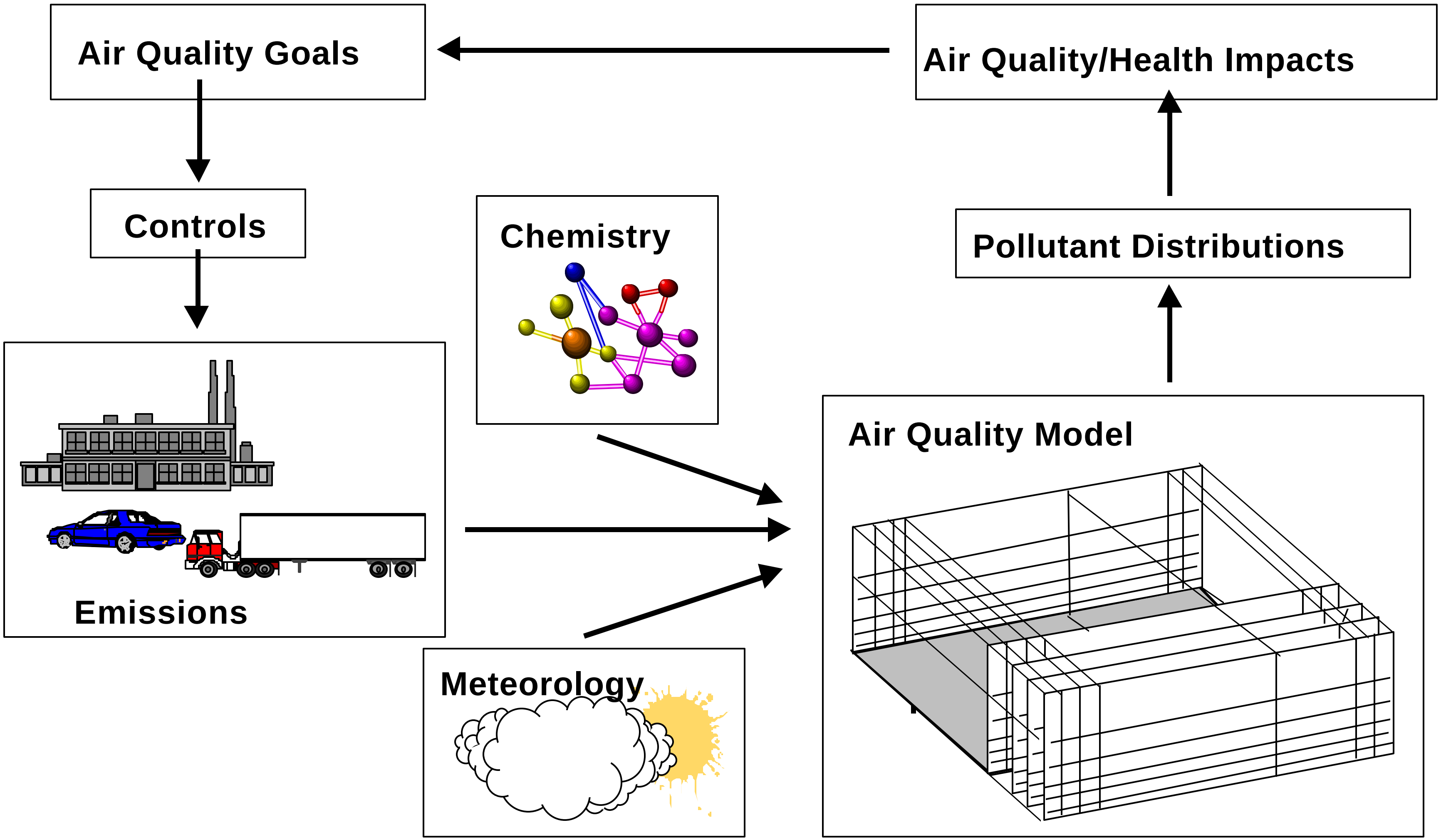
Air quality modeling



Air quality modeling (contd.)

- Air quality models are indispensable part of air quality management.
  - The main tool to evaluate the effectiveness of different control strategies.
- They possess more information than we generally use.
  - Local sensitivity coefficients, i.e. derivatives.
  - More dimensions to look at.
- First-order sensitivity analysis does not capture non-linearities.
  - Viewed as limitation.
- A high-order sensitivity analysis technique is developed.

The idea of sensitivity analysis

- How does the model respond to a change in an independent parameter?
  - Almost any modeling practice is, in essence, an effort to address this question directly or indirectly.
- In air quality modeling the sensitivity question is addressed directly to:
  - Quantify the atmospheric response to emissions control,
  - Find the uncertainty in the response,
  - Perform inverse modeling and data assimilation,
  - Etc.

Brute-force sensitivity analysis

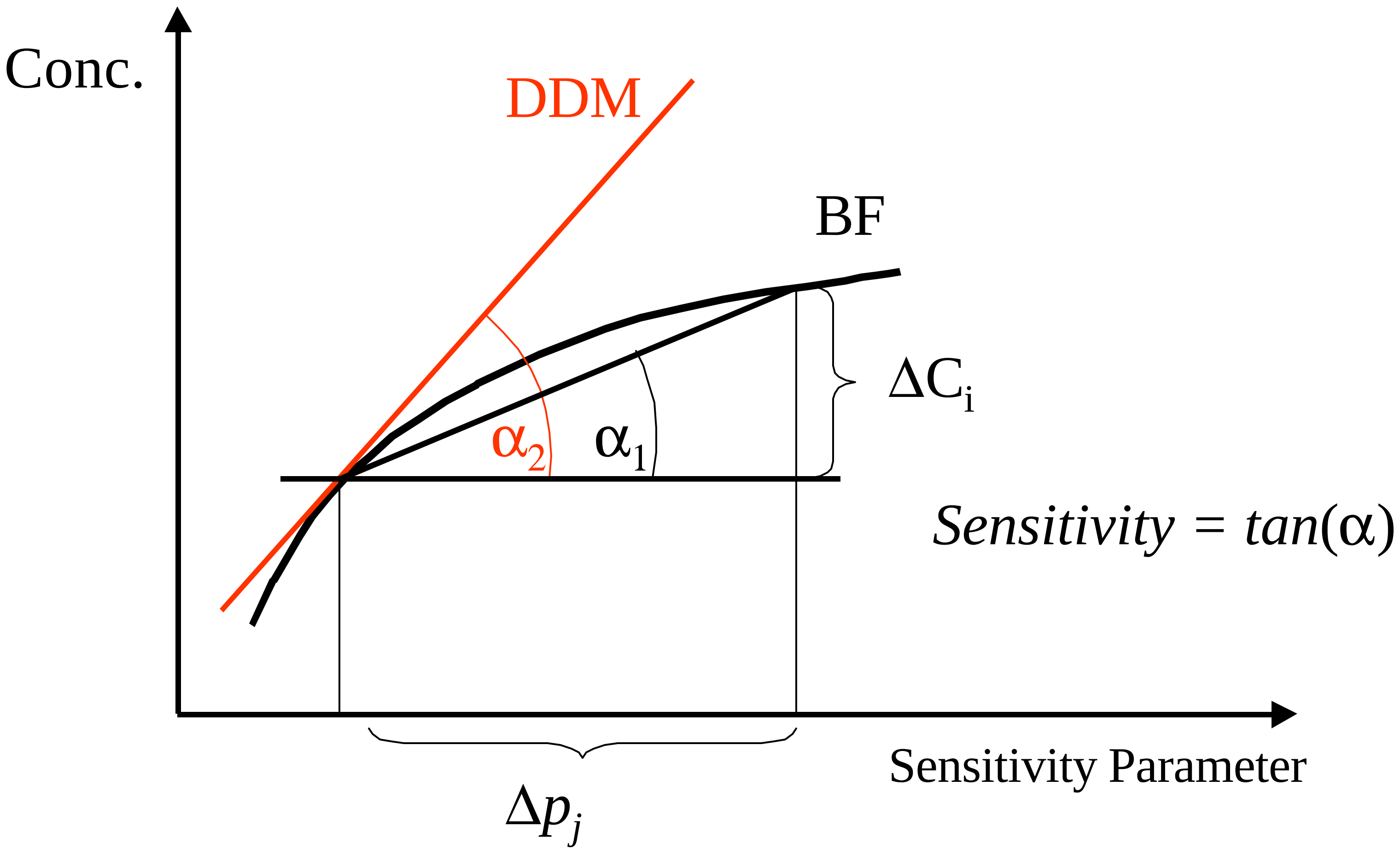
- Approximates the derivative as the sensitivity parameter approaches zero:

$$S_{ij} \cong \frac{\Delta C_i}{\Delta p_j}$$

- One-at-a-time perturbation of each sensitivity parameter.

- Pros and cons:
  - Easy to do,
  - One additional run for each additional sensitivity parameter,
  - Noisy for small perturbations,
  - Unrealistic for large perturbations.

Brute-force sensitivity analysis (contd.)



DDM sensitivity analysis

- Decoupled Direct Method (Dunker, 1981 and 1984): Sensitivity equations are derived and integrated decoupled from concentrations.
  - The only direct sensitivity technique that has been applied in 3-D air quality models [DDM-3D (Yang et al., 1997)],
  - Drawbacks of the brute-force sensitivity analysis avoided.

	Advection	Diffusion	Chemistry	Emissions
$\frac{\partial C_i}{\partial t} =$	$-\nabla(\mathbf{u}C_i)$	$+\nabla(\mathbf{K}\nabla C_i)$	$+R_i$	$+E_i$
$\frac{\partial S_{ij}}{\partial t} =$	$-\nabla(\mathbf{u}S_{ij})$	$+\nabla(\mathbf{K}\nabla S_{ij})$	$+\mathbf{JS}_j$	$+E_i$

DDM: computational efficiency

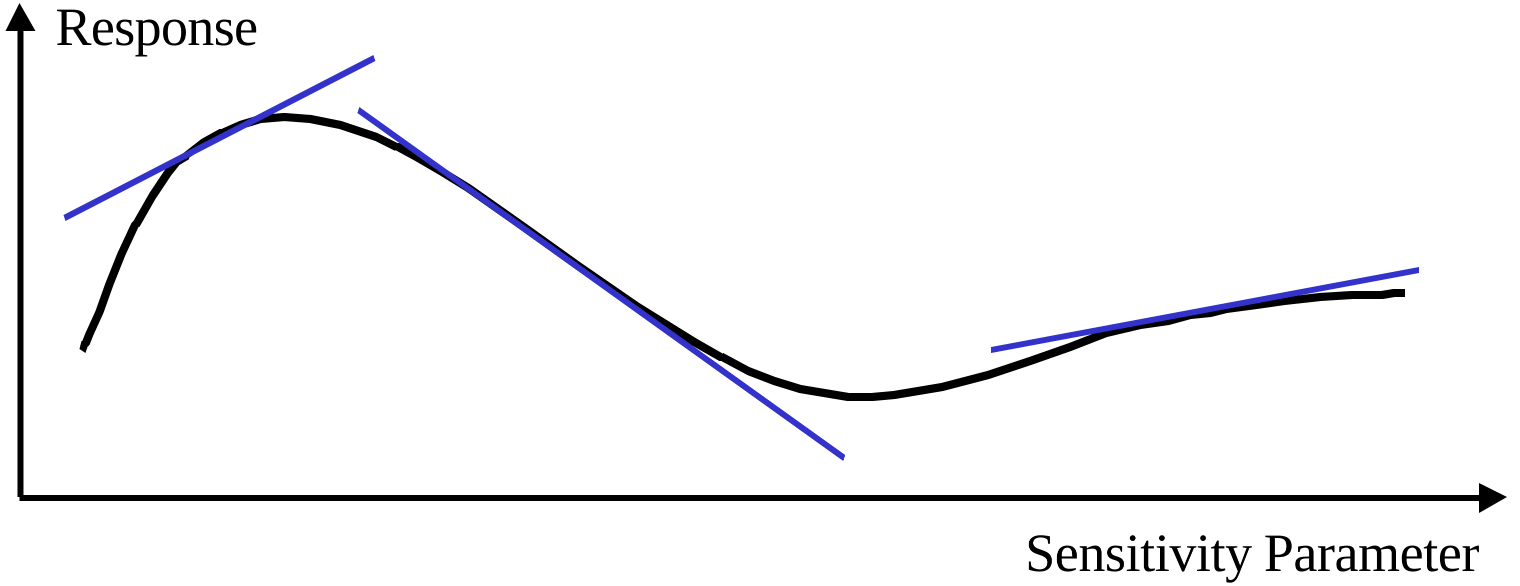
- DDM-3D is easy to implement because for the most part the same numerical routines are used to integrate the sensitivities.
- Chemistry discretization:

$$\mathbf{S}^{n+1} = \left(\mathbf{I} - \frac{\Delta t}{2} \mathbf{J_C} \right)^{-1} \left(\mathbf{I} + \frac{\Delta t}{2} \mathbf{J_C} \right) \mathbf{S}^{n}$$

  – Matrix inversion is done only once for each grid cell and time step.

How to address non-linearity?

First-order sensitivity coefficients (slopes) cannot describe the non-linearities in the response -- 2nd order (curvature) or higher-order coefficients are required.



Calculation of higher-order derivatives

High-order Decoupled Direct Method (HDDM):

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

$$\frac{\partial S_{ij}^{(2)}}{\partial t} = -\nabla(\mathbf{u}S_{ij}^{(2)}) + \nabla(\mathbf{K}\nabla S_{ij}^{(2)}) + \boxed{\mathbf{J}_i \mathbf{S}_j^{(2)} + \mathbf{J}^*_{S^{(1)}} \mathbf{S}_j^{(1)}}$$

$$\frac{\partial S_{ij}^{(m)}}{\partial t} = -\nabla(\mathbf{u}S_{ij}^{(m)}) + \nabla(\mathbf{K}\nabla S_{ij}^{(m)}) + \boxed{\mathbf{J}_i \mathbf{S}_j^{(m)} + \sum_{k=1}^{m-1} \binom{m-1}{k} \mathbf{J}^*_{S^{(k)}} \mathbf{S}_j^{(m-k)}}$$

Calculation of higher-order derivatives (contd.)

Chemistry discretization in HDDM:

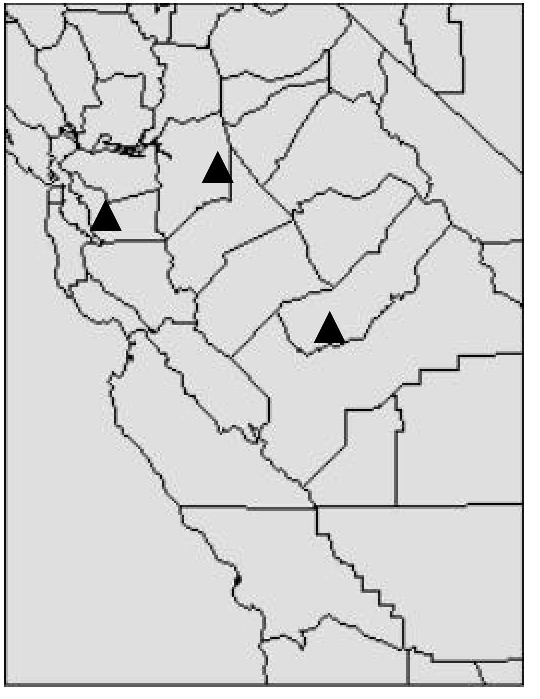
$$\mathbf{S}^{(1)^{n+1}} = \left(\mathbf{I} - \frac{\Delta t}{2}\mathbf{J_C}\right)^{-1}\left(\mathbf{I} + \frac{\Delta t}{2}\mathbf{J_C}\right)\mathbf{S}^{(1)^{n}}$$

$$\mathbf{S}^{(2)^{n+1}} = \left(\mathbf{I} - \frac{\Delta t}{2}\mathbf{J_C}\right)^{-1}\left[\left(\mathbf{I} + \frac{\Delta t}{2}\mathbf{J_C}\right)\mathbf{S}^{(2)^{n}} + \Delta t \mathbf{J}^{*}_{S^{(1)}}\mathbf{S}^{(1)}\right]$$

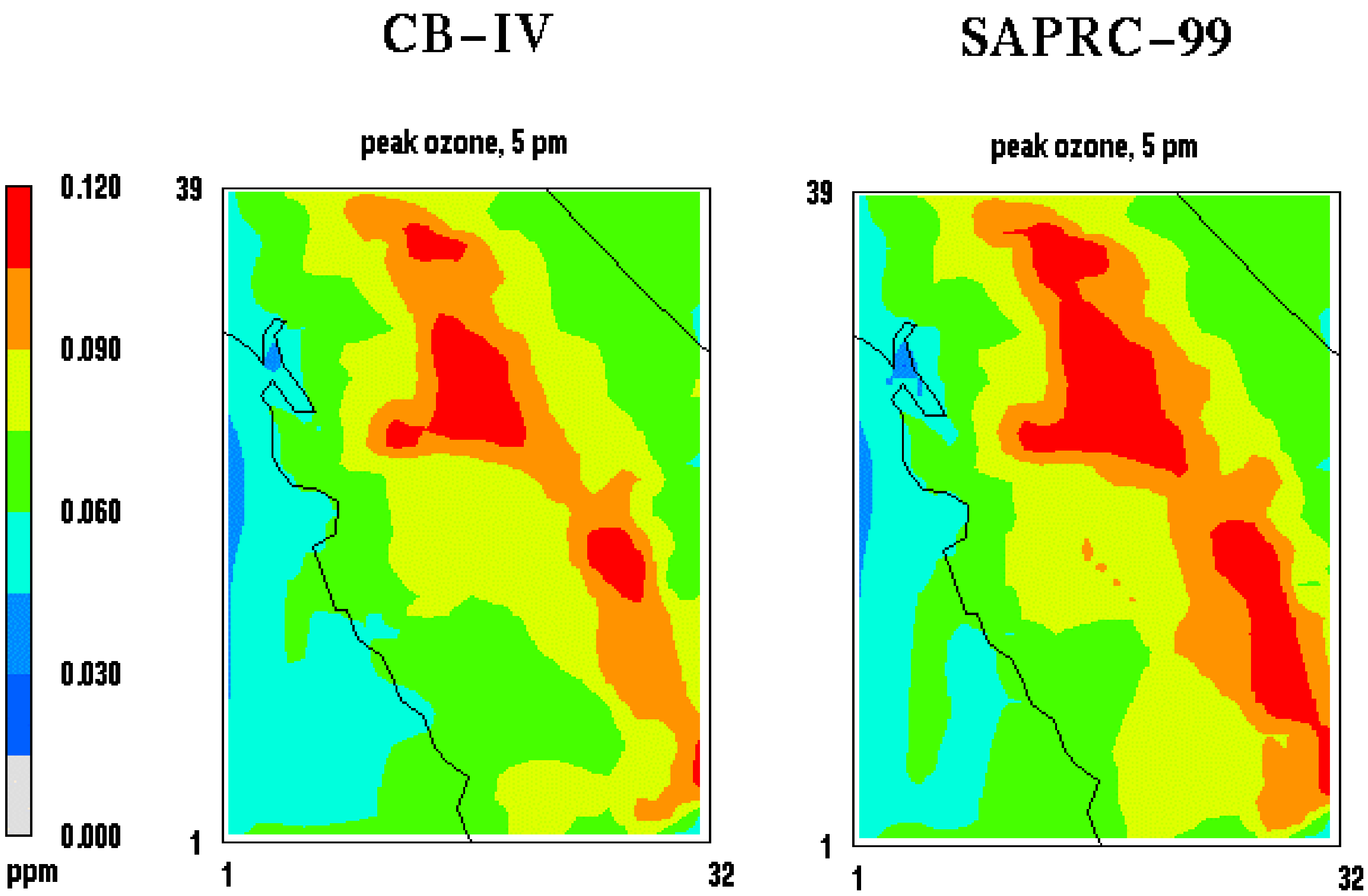
$$\mathbf{S}^{(m)^{n+1}} = \left(\mathbf{I} - \frac{\Delta t}{2}\mathbf{J_C}\right)^{-1}\left[\left(\mathbf{I} + \frac{\Delta t}{2}\mathbf{J_C}\right)\mathbf{S}^{(m)^{n}} + \Delta t \sum_{k=1}^{m-1}\binom{m-1}{k}\mathbf{J}^{*}_{S^{(k)}}\mathbf{S}^{(m-k)}\right]$$

Method test results

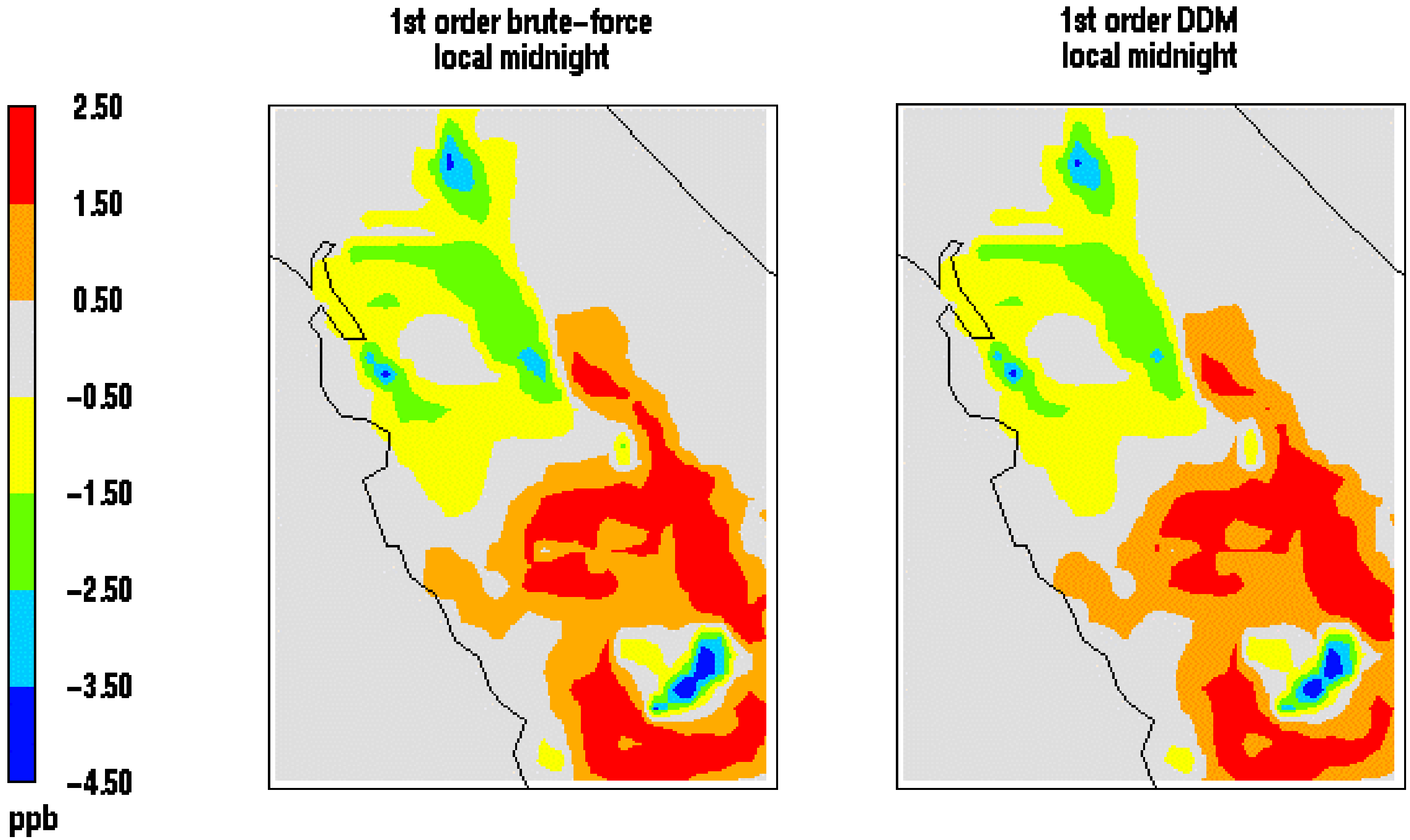
- Multiscale Air Quality SImulation Platform (MAQSIP)
  - Some modifications from the original model, inputs: e.g., BCs, mass conservation scheme,
  - Results shown for CB-IV, SAPRC-99 results to come soon.
- SARMAP modeling domain and episode (August 2-6, 1990).



Simulated ozone: SAPRC-99 vs. CB-IV

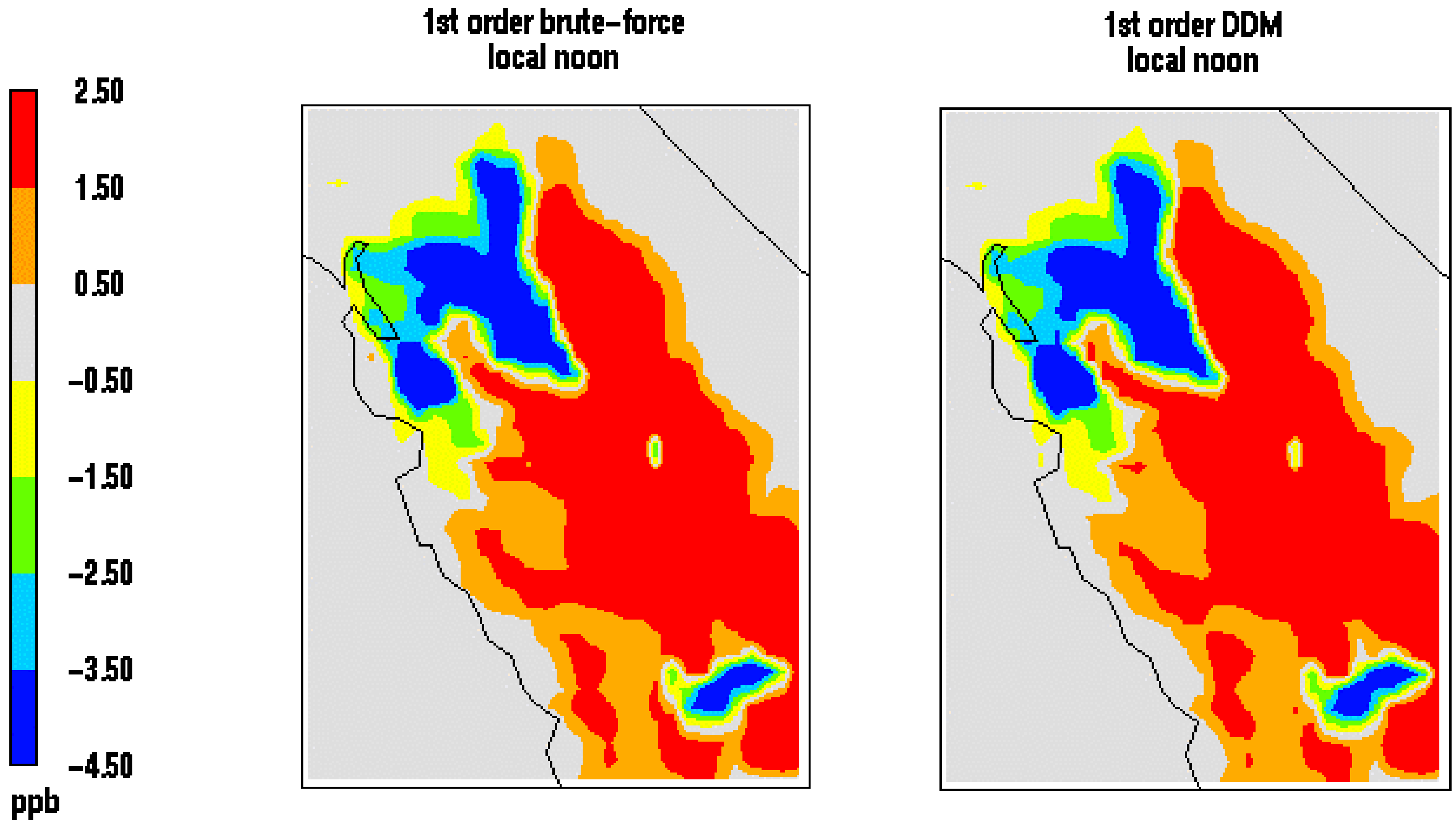


DDM vs. BF: 1st order derivative, midnight



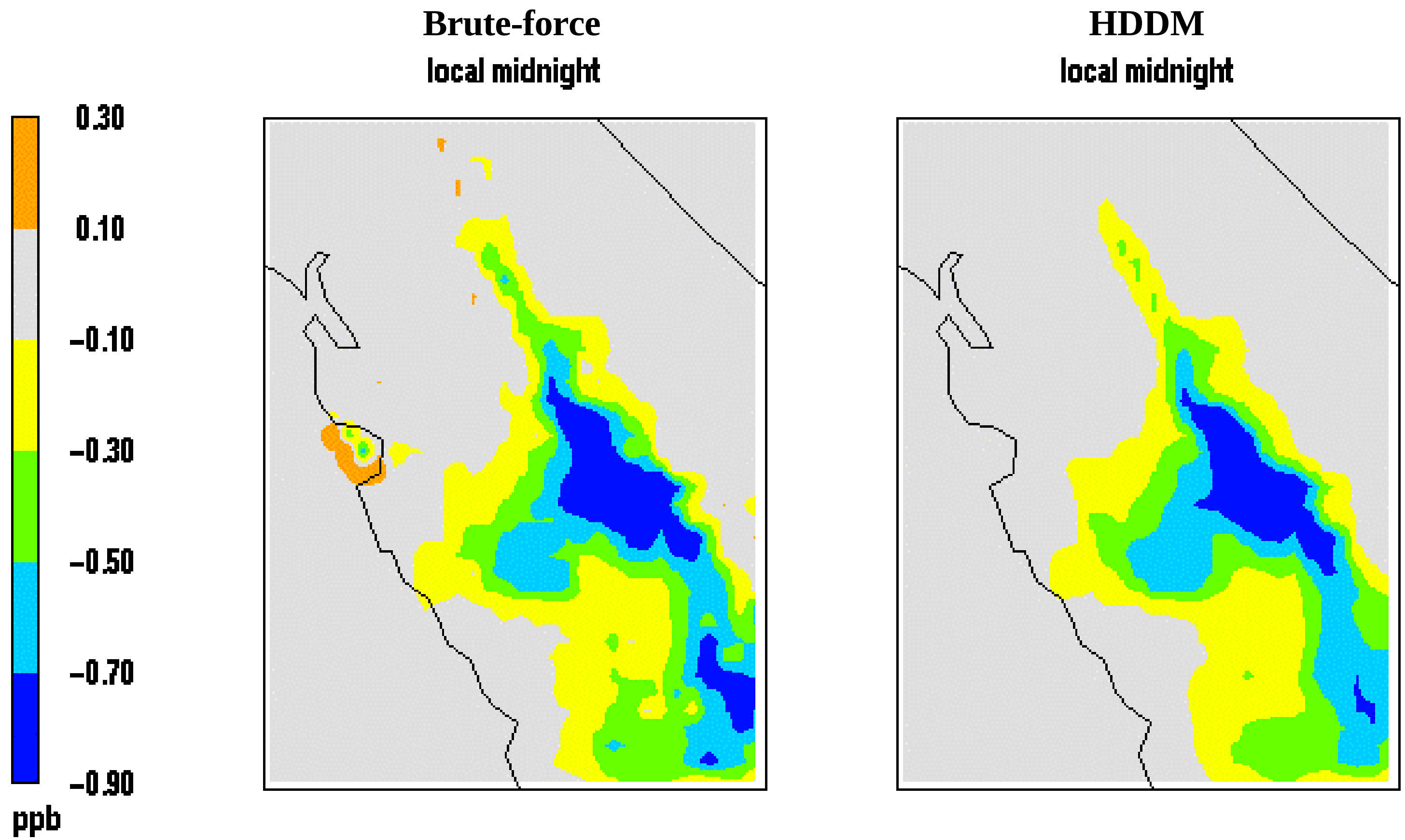
25% perturbation in domain-wide NO emissions

DDM vs. BF: 1st order derivative, noon



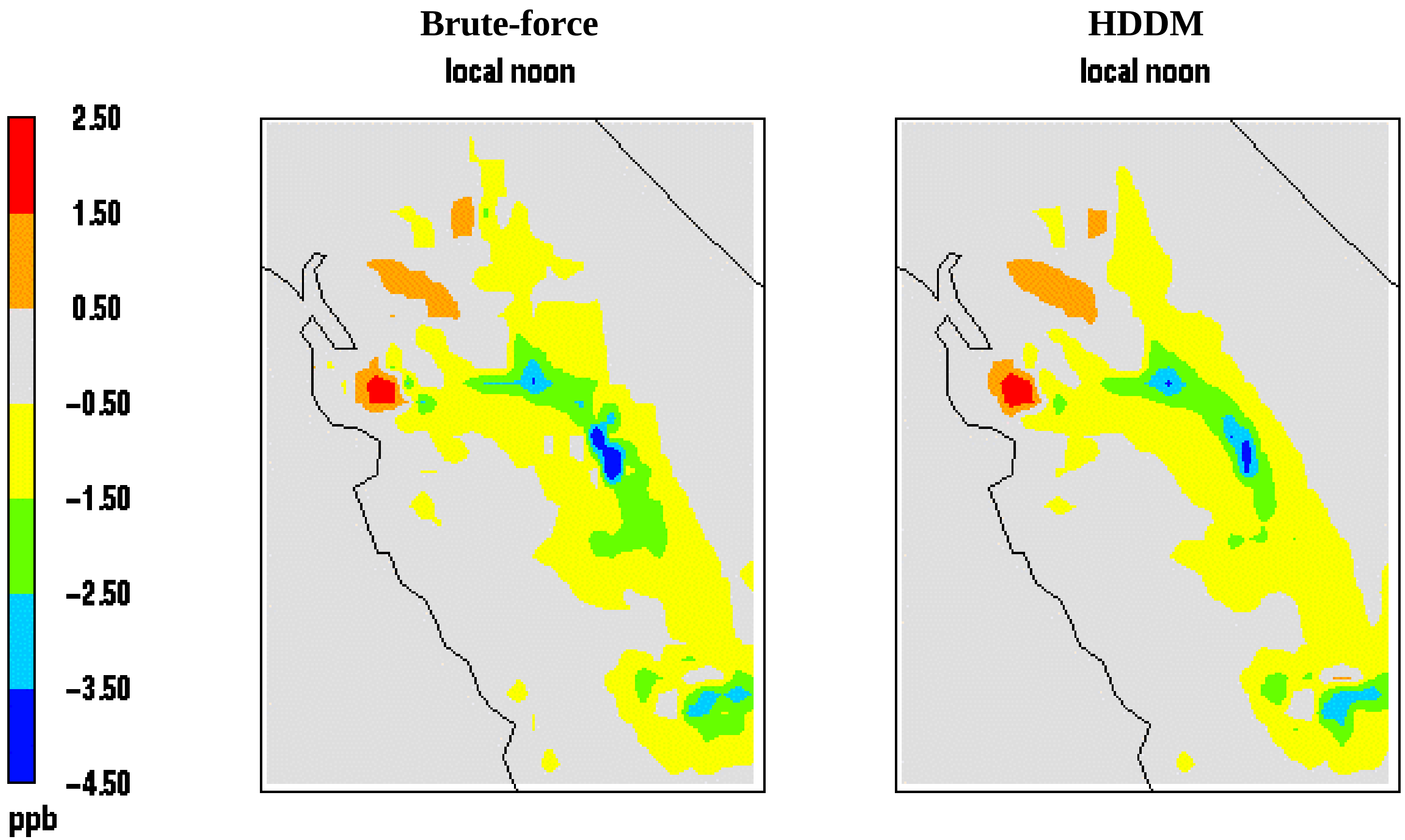
25% perturbation in domain-wide NO emissions

DDM vs. BF: 2nd order derivative, midnight



25% perturbation in domain-wide NO emissions

DDM vs. BF: 2nd order derivative, noon



25% perturbation in domain-wide NO emissions

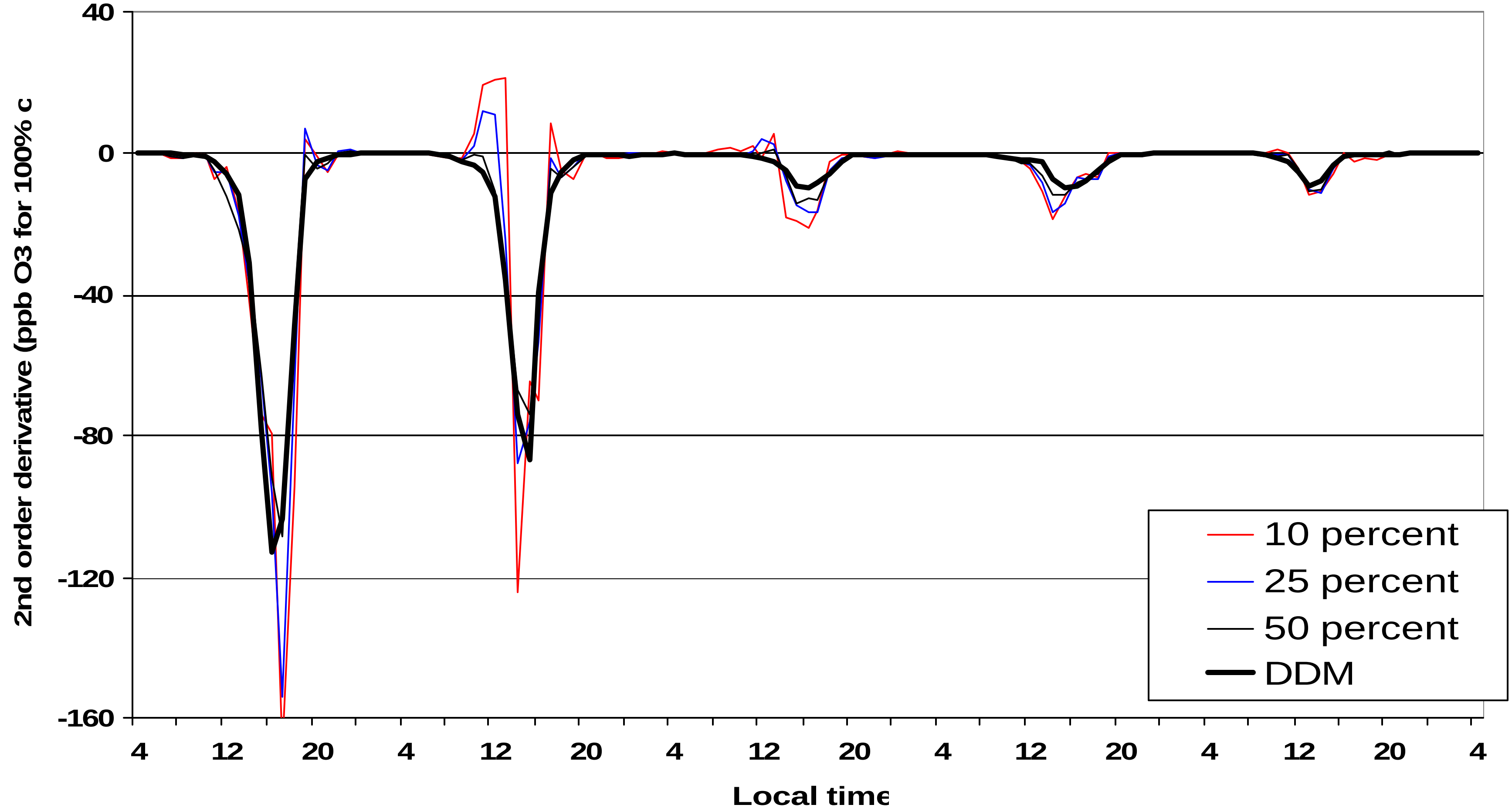
DDM vs. BF?

- Better agreement between BF and (H)DDM for the 1st order sensitivity coefficients than 2nd order:
  - 2nd order brute-force approximation is more susceptible to numerical noise.

$$S^{(1)} \cong \frac{C(p+\Delta p) - C(p-\Delta p)}{2\Delta p}$$

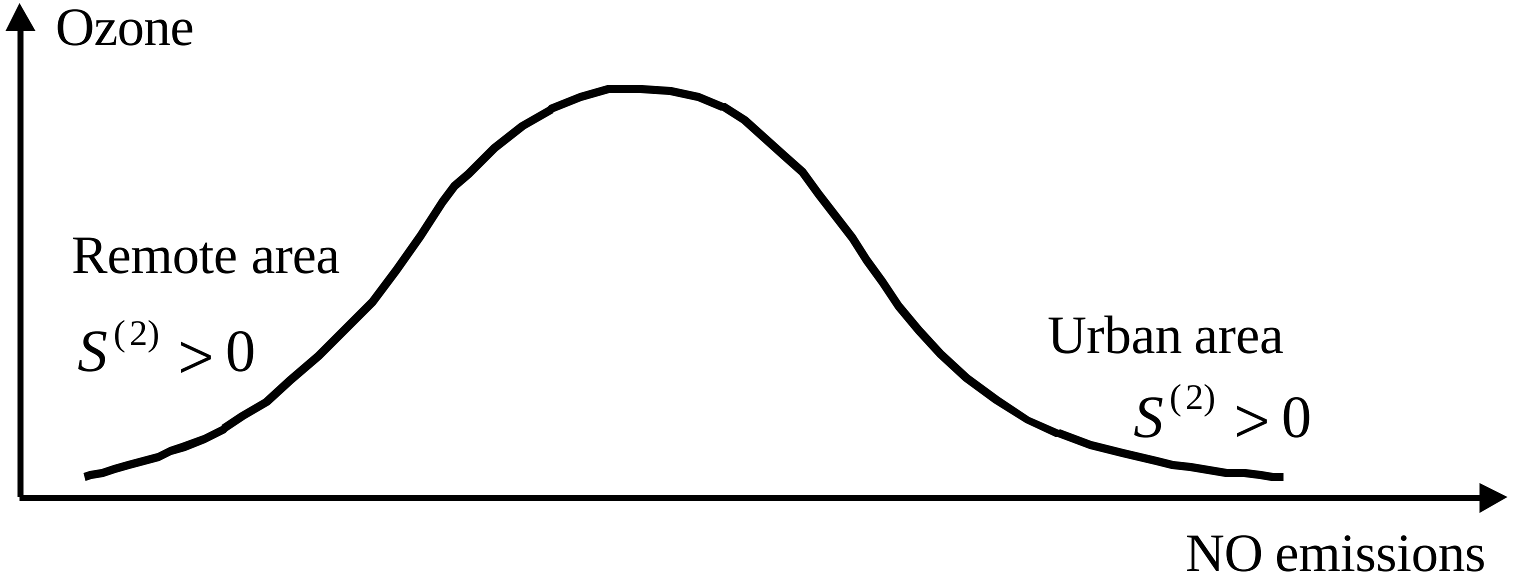
$$S^{(2)} \cong \frac{C(p+\Delta p) - 2C(p) + C(p-\Delta p)}{\Delta p^2}$$

DDM vs. BF 2nd order derivative: peak ozone

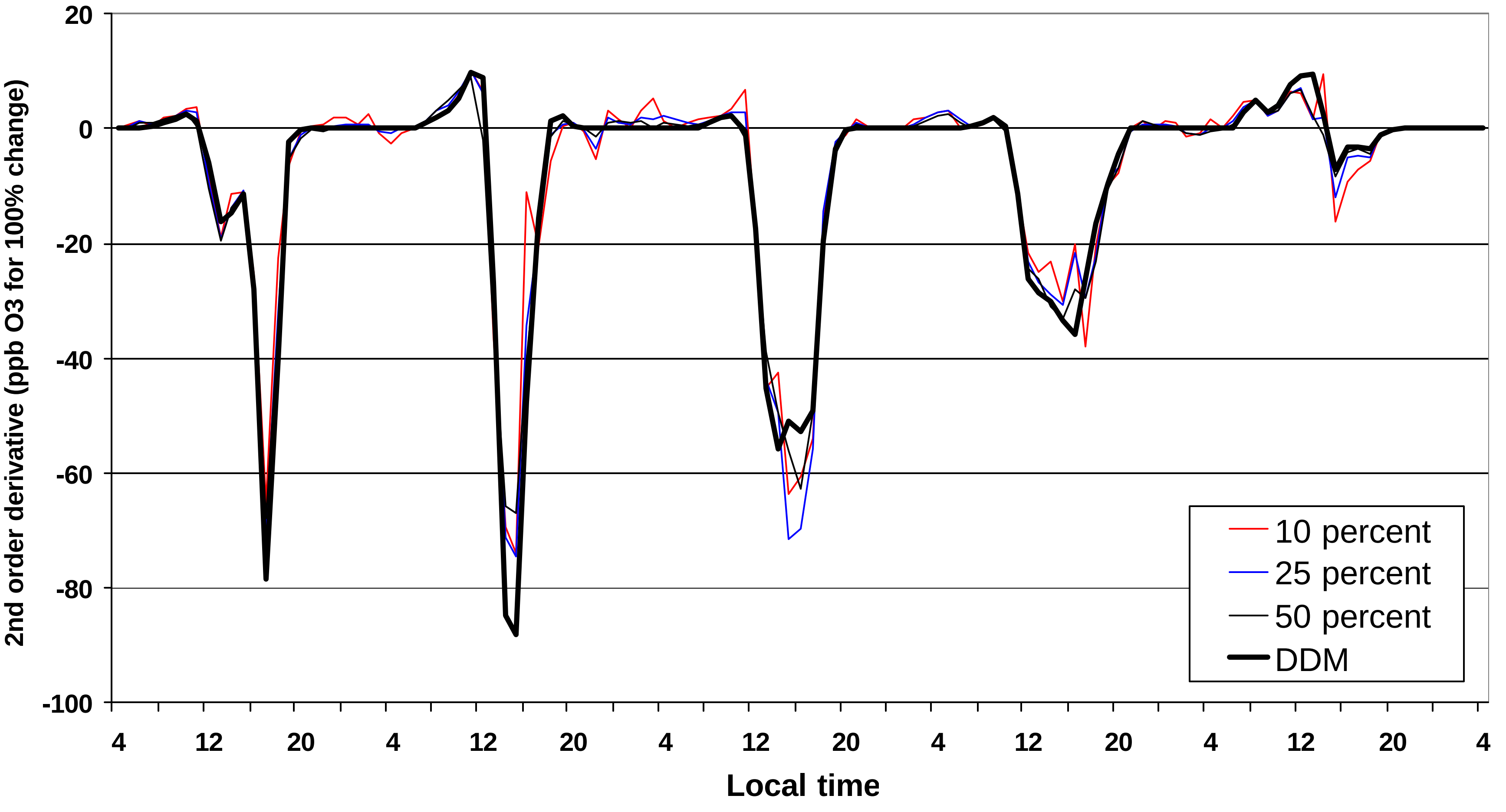


DDM vs. BF?

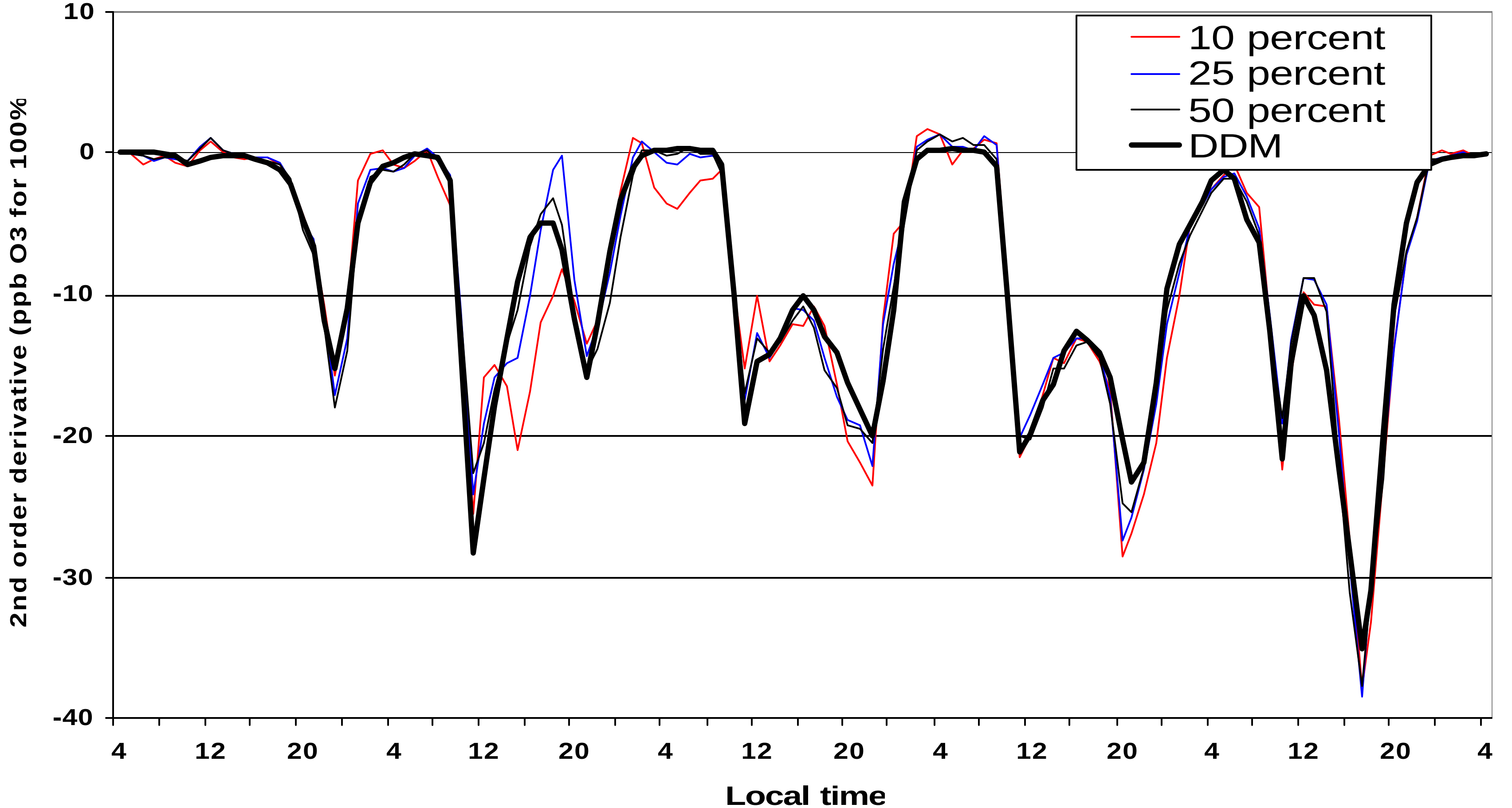
- A concave ozone response at high ozone concentration is unlikely.



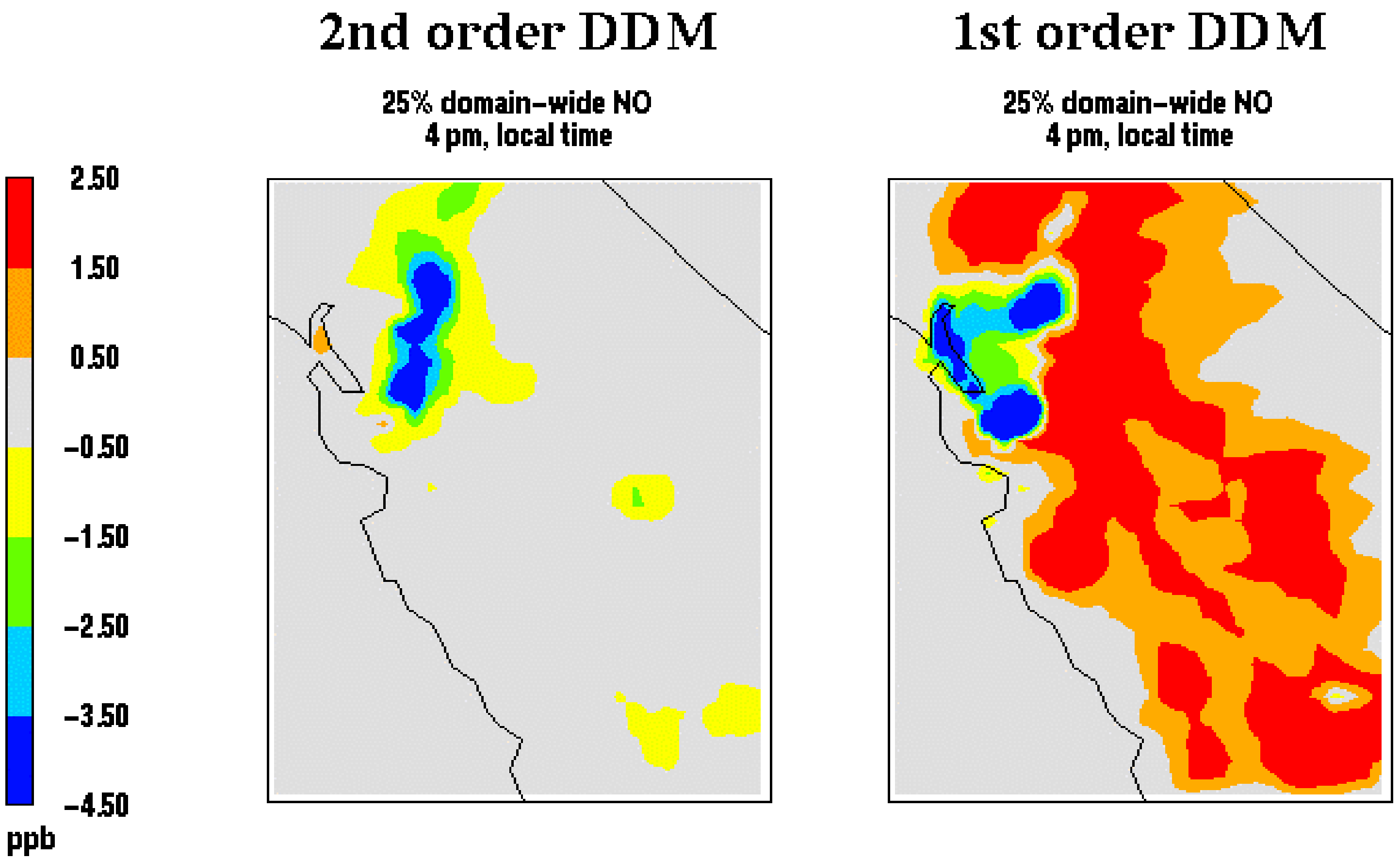
DDM vs. BF 2nd order derivative: VOC-limited



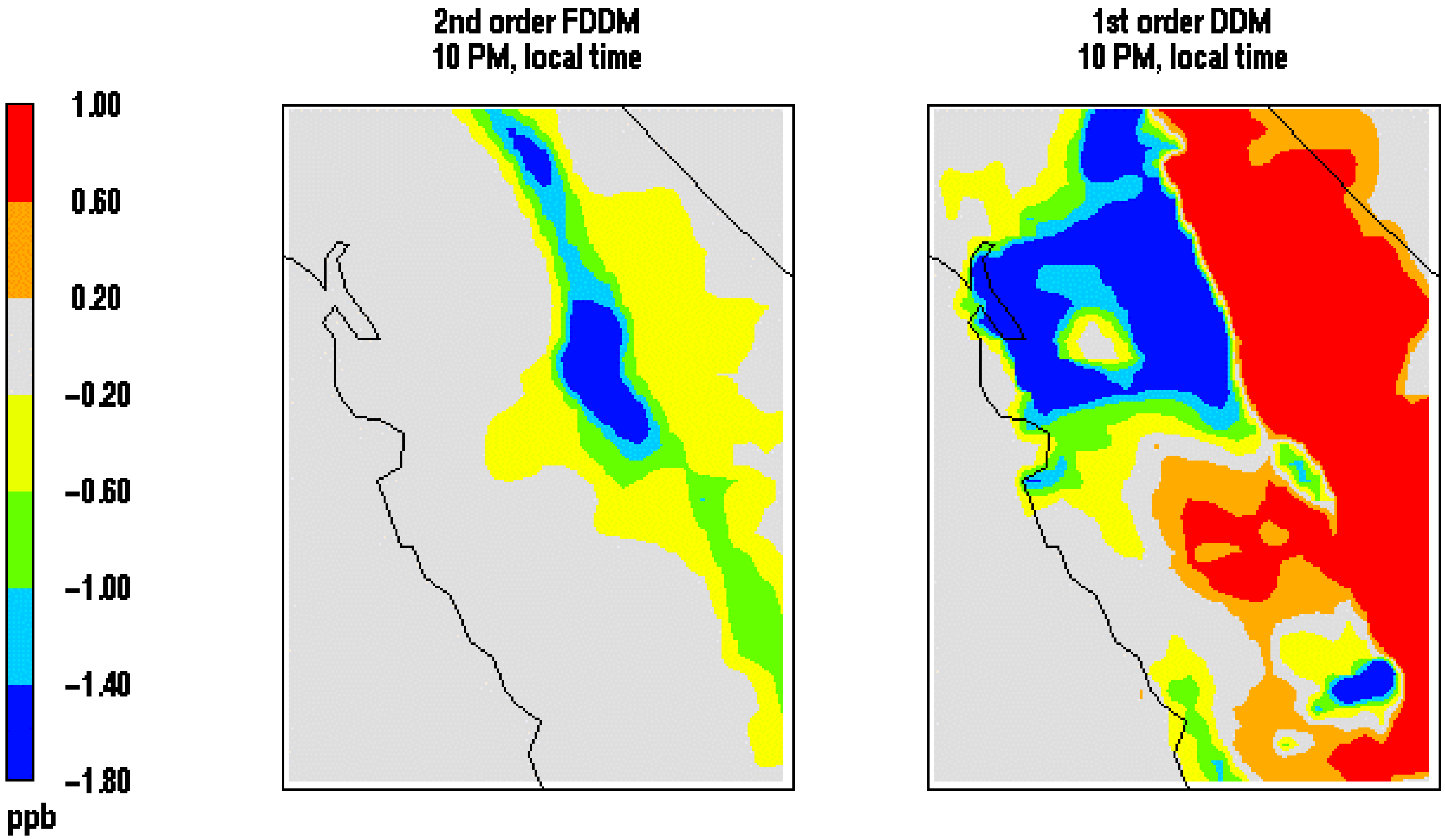
DDM vs. BF 2nd order derivative: NOx-limited



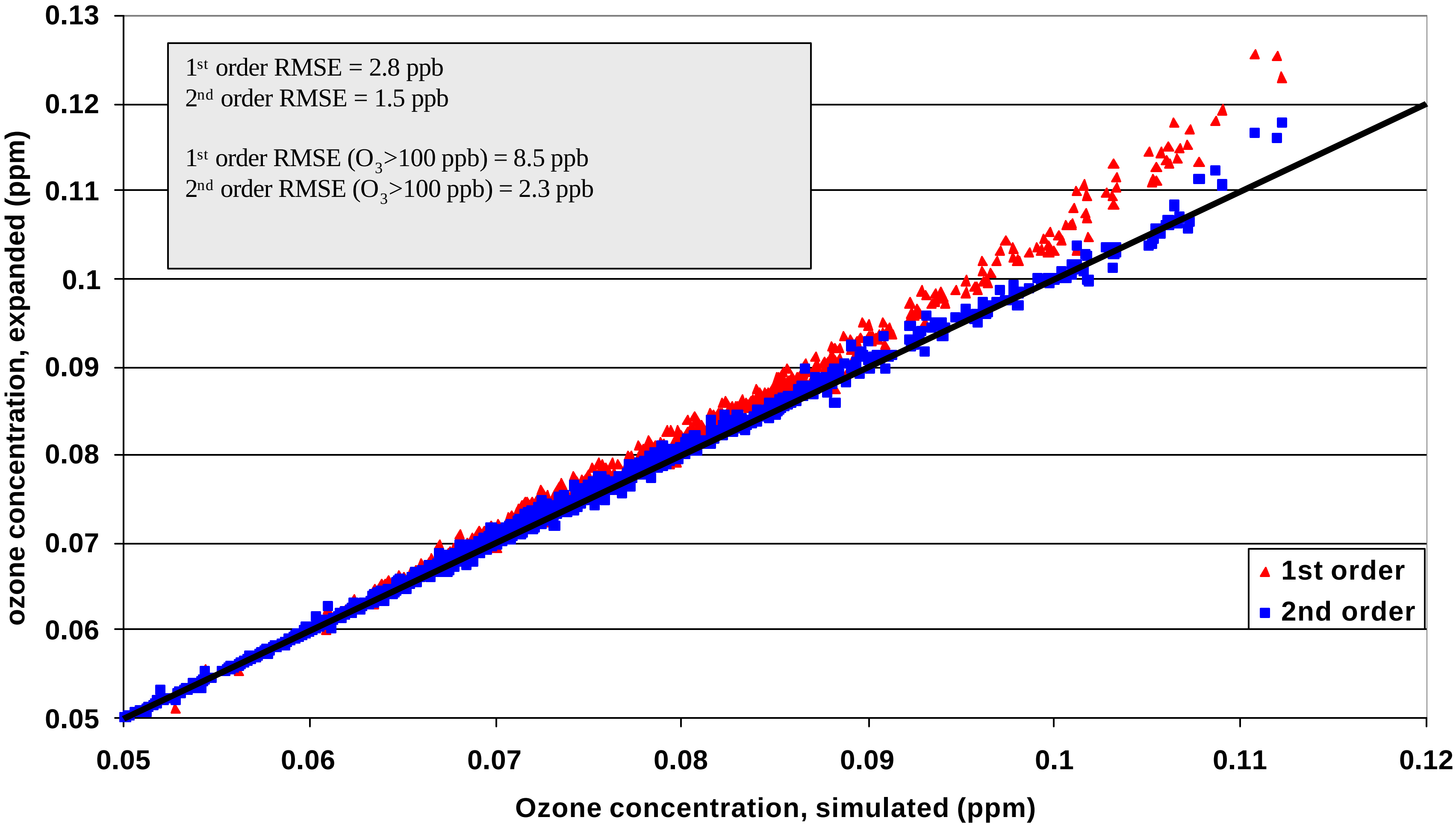
Non-linearity: where, why?



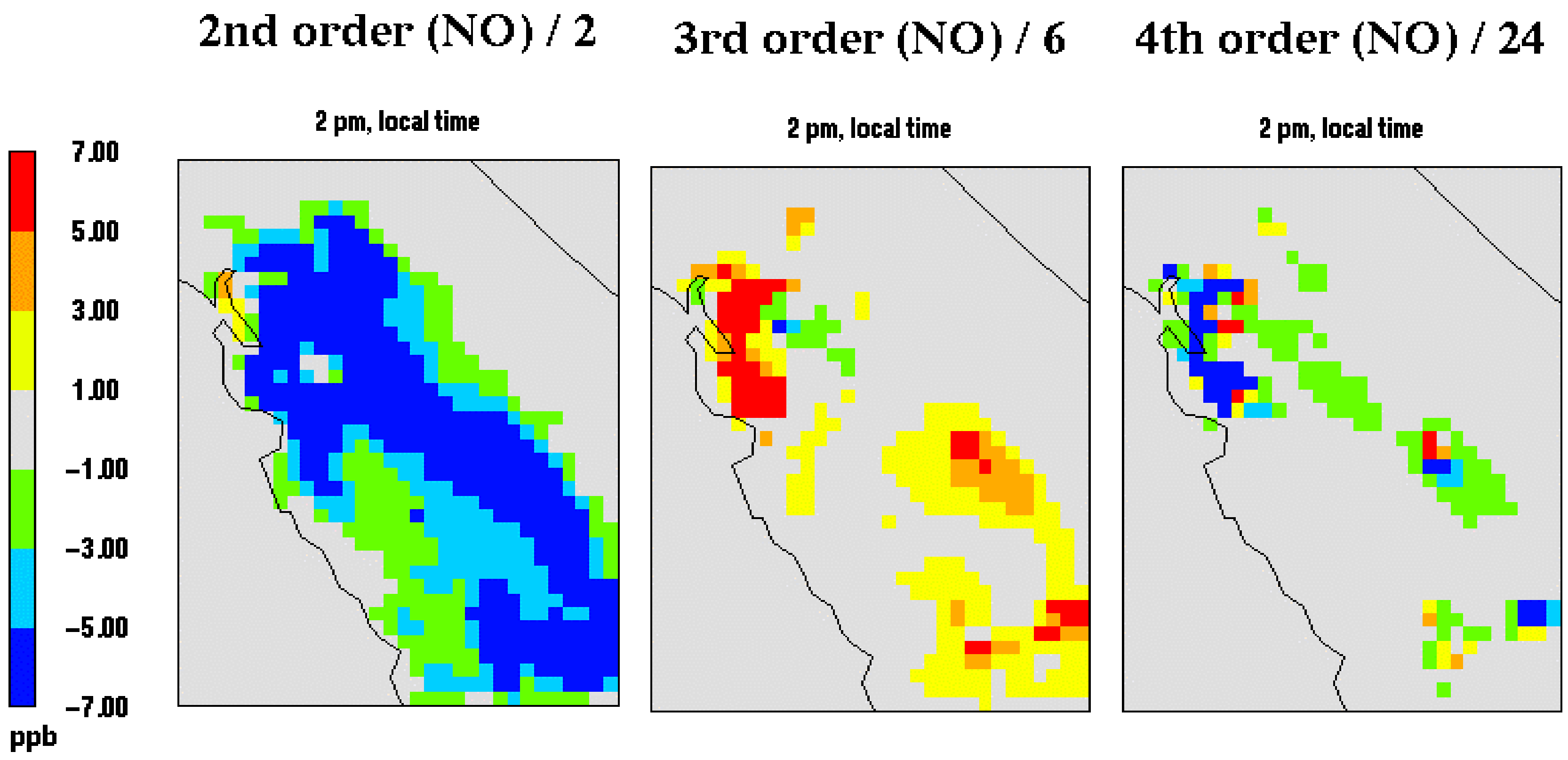
Non-linearity: where, why? (contd.)



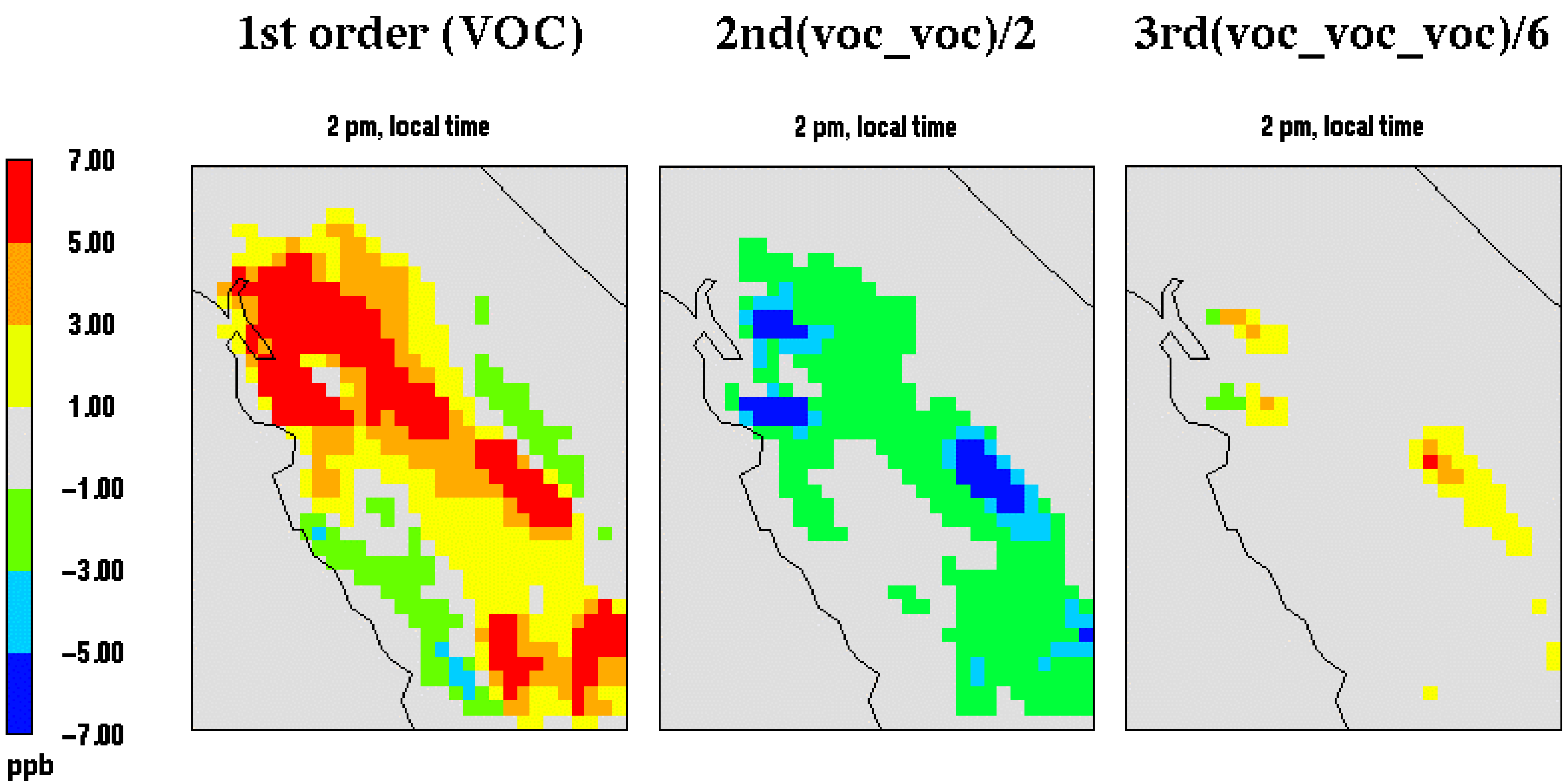
Non-linearity: where, why? (contd.)



Higher order derivatives (domain-wide NO)



Higher order derivatives (domain-wide VOC)



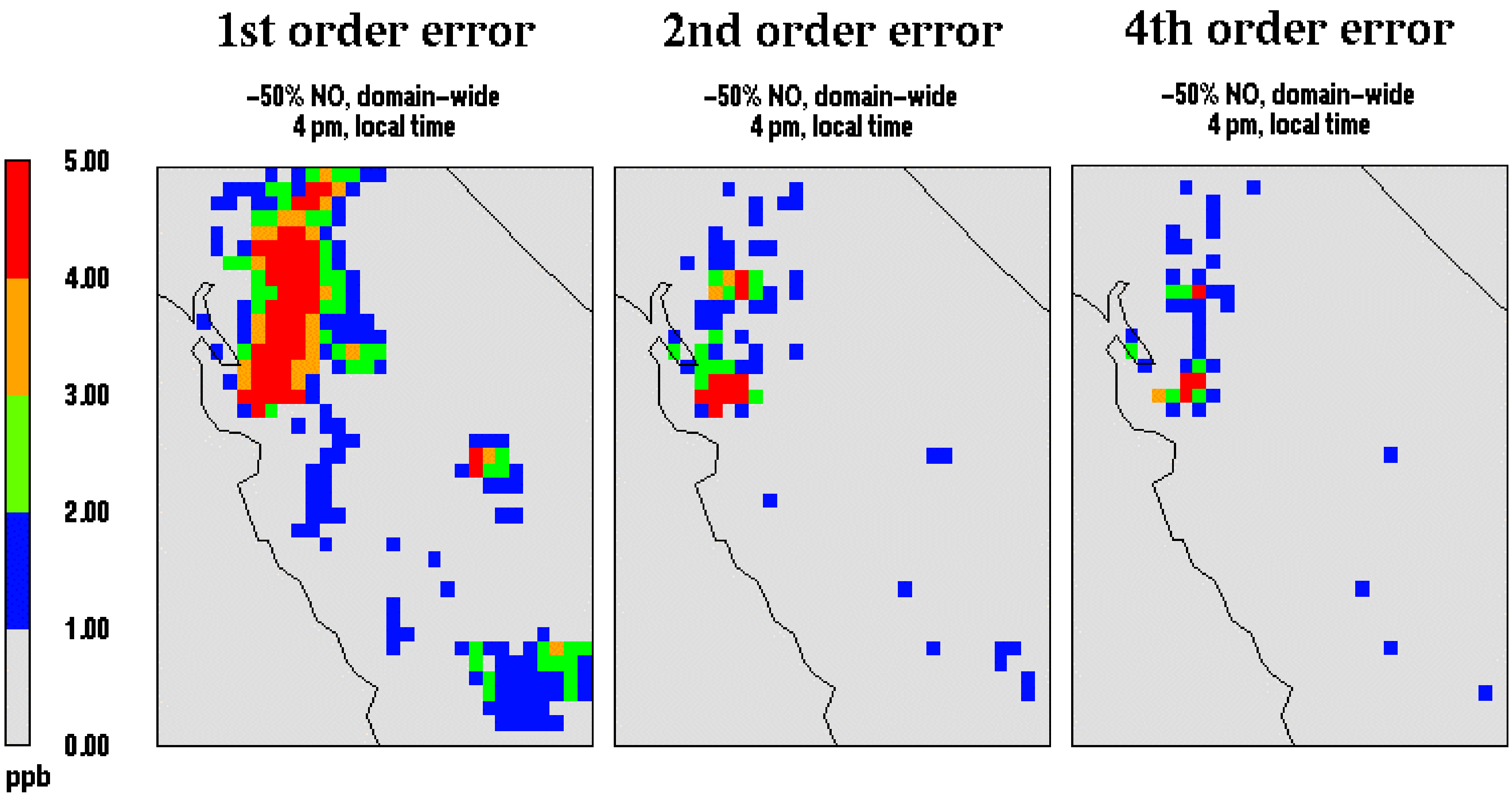
Taylor series expansion

- One way to test the accuracy of the calculated derivatives:

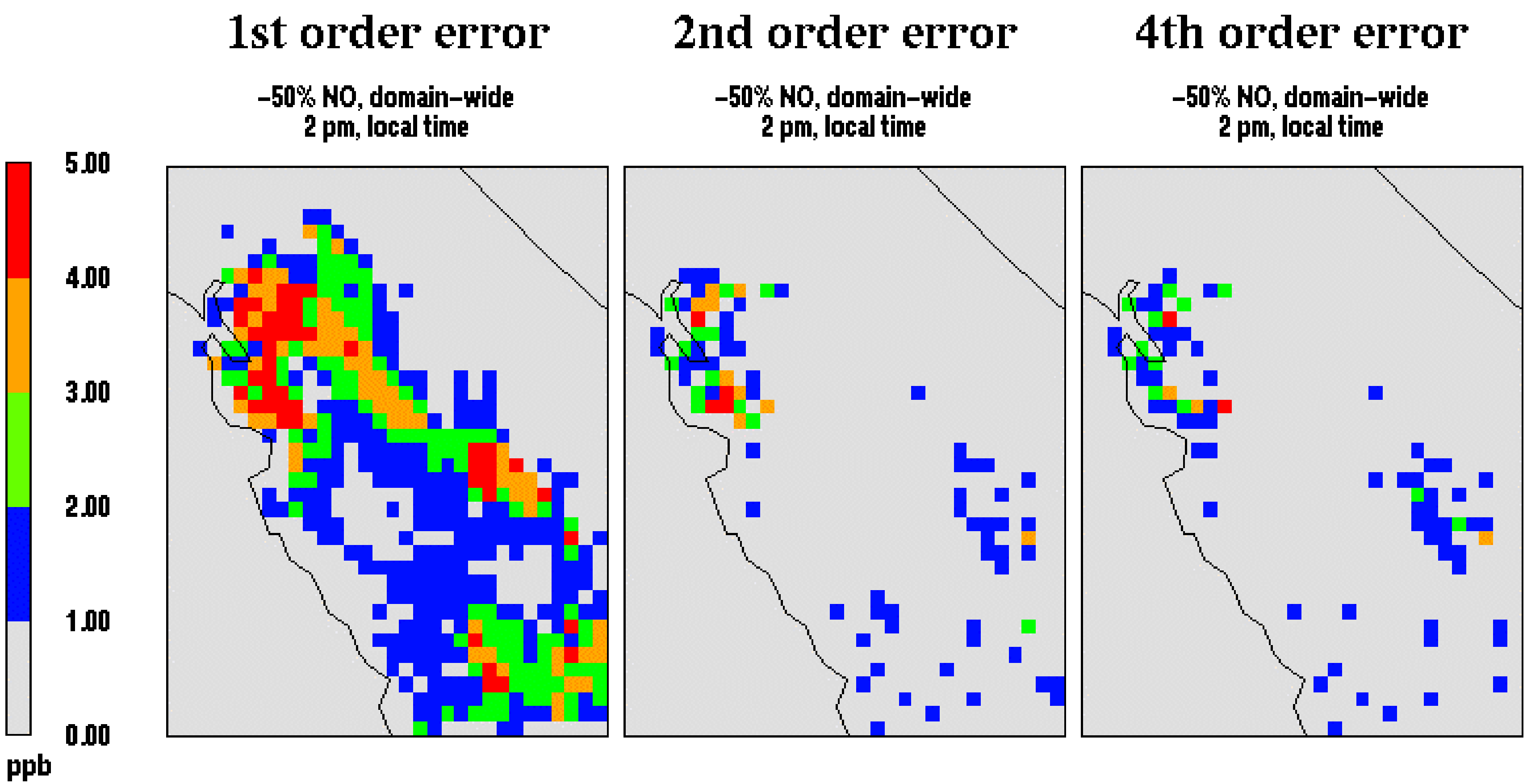
$$C(x_0+\Delta x) = C(x_0) + \Delta x S^{(1)}(x_0) + \frac{(\Delta x)^2}{2} S^{(2)}(x_0)$$
$$+ \frac{(\Delta x)^3}{6} S^{(3)}(x_0) + \frac{(\Delta x)^4}{24} S^{(4)}(x_0) + \dots$$

- More accurate Taylor expansion:
  - Higher-order terms,
  - Smaller perturbation.
- Not the most accurate scaling method (Kramer et al., 1984)

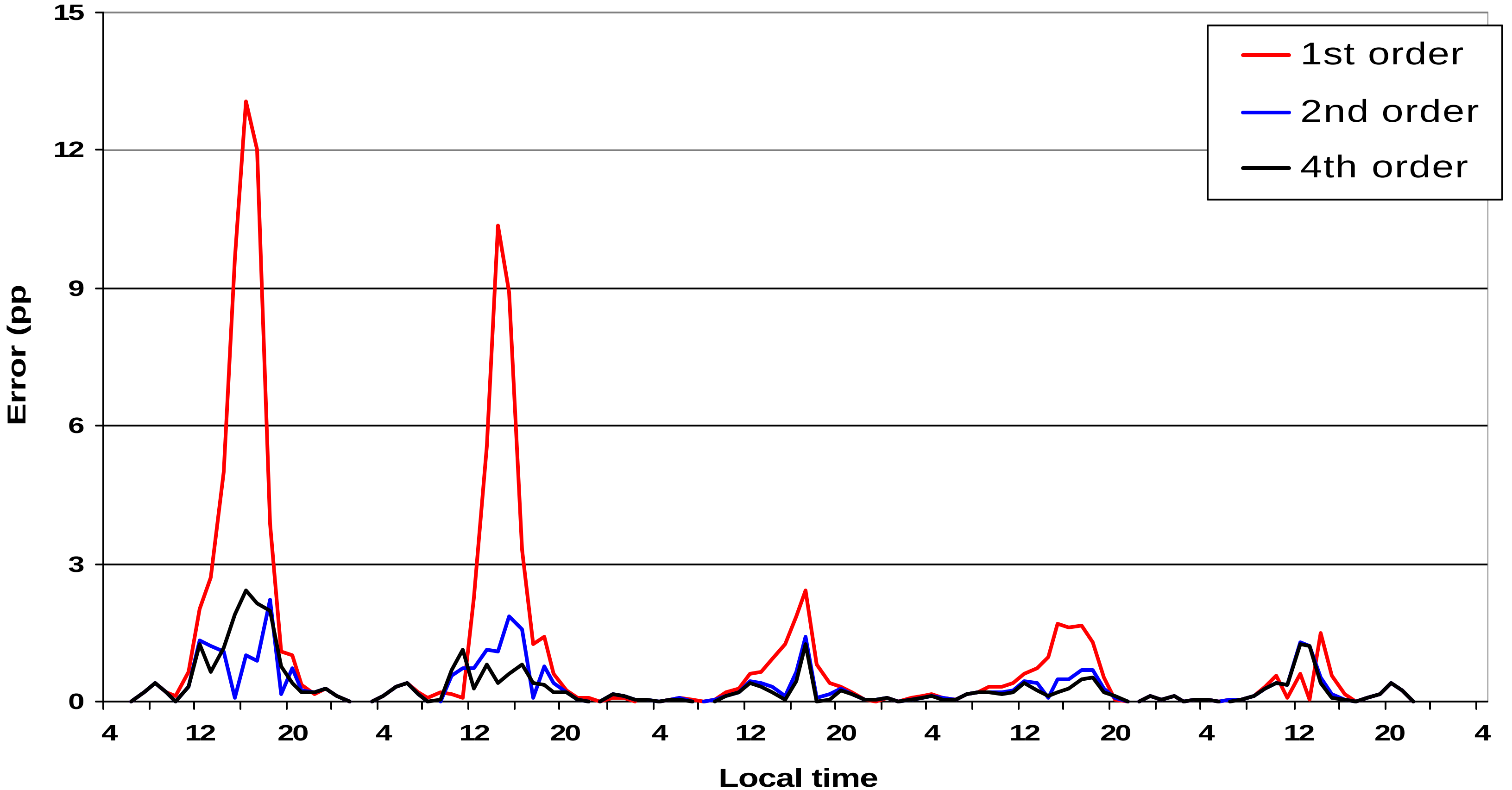
Taylor series expansion: 50% reduction in NO



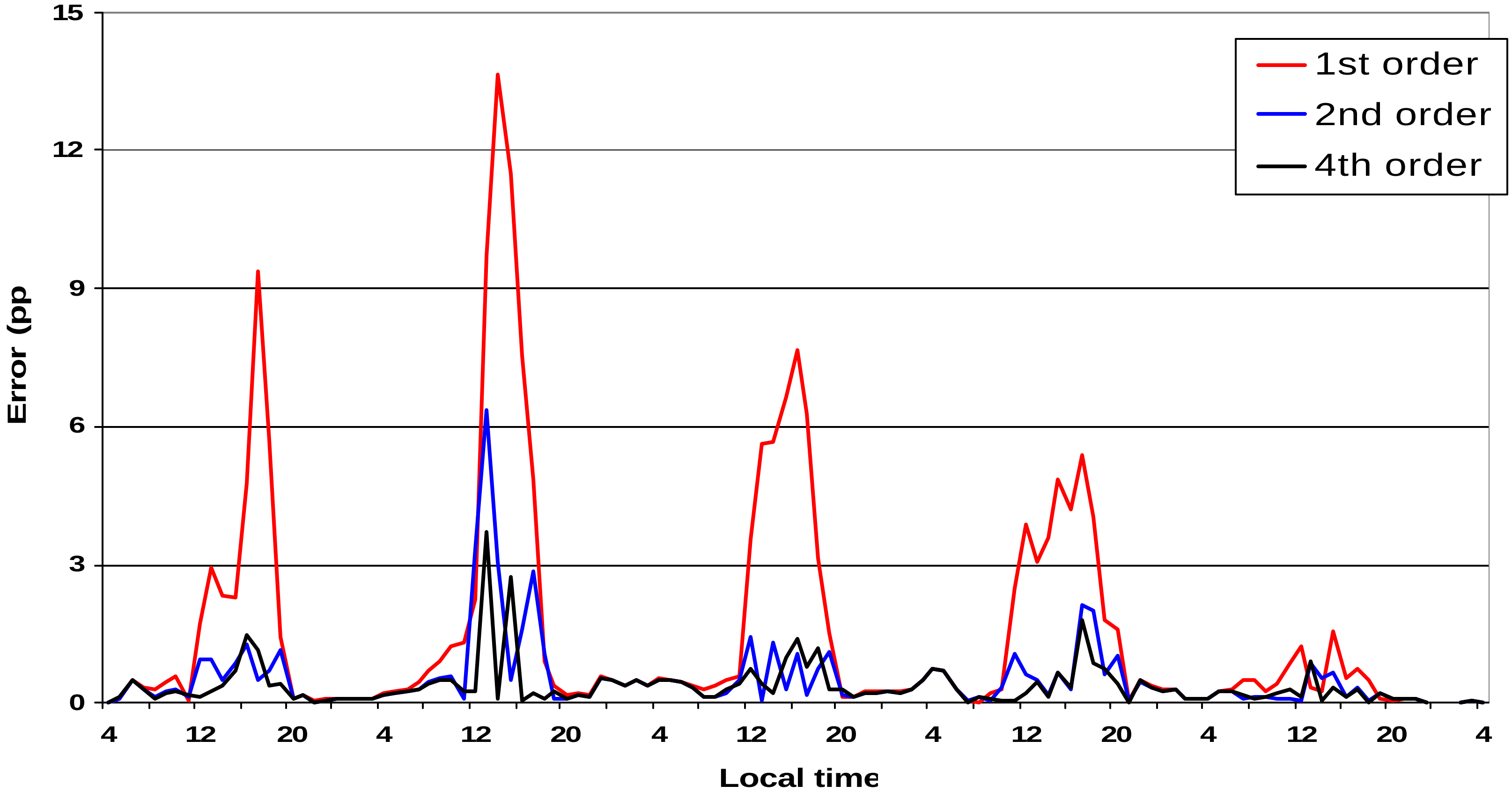
Taylor series expansion (contd.)



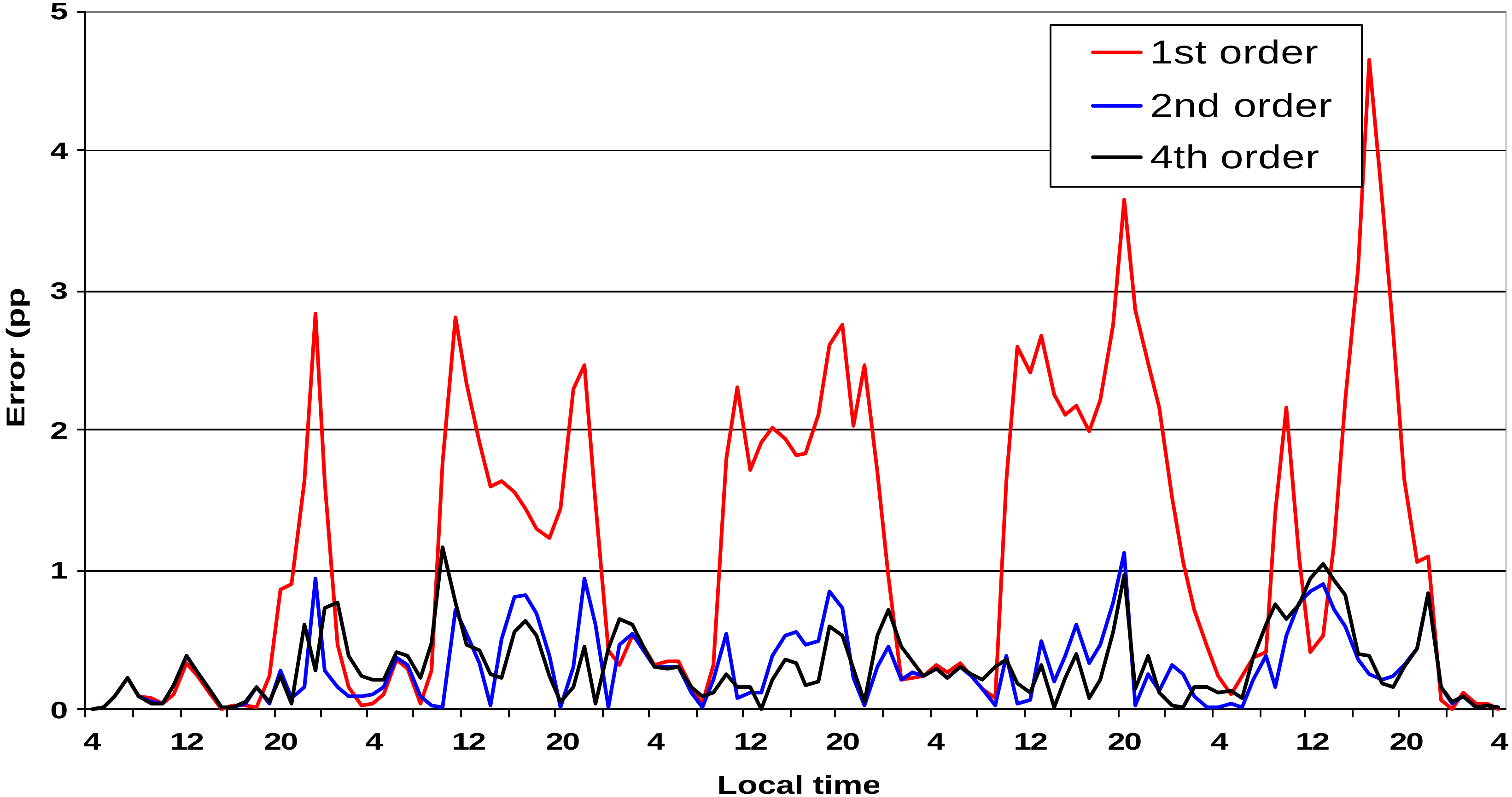
Taylor expansion: NO perturbation, location of the peak ozone



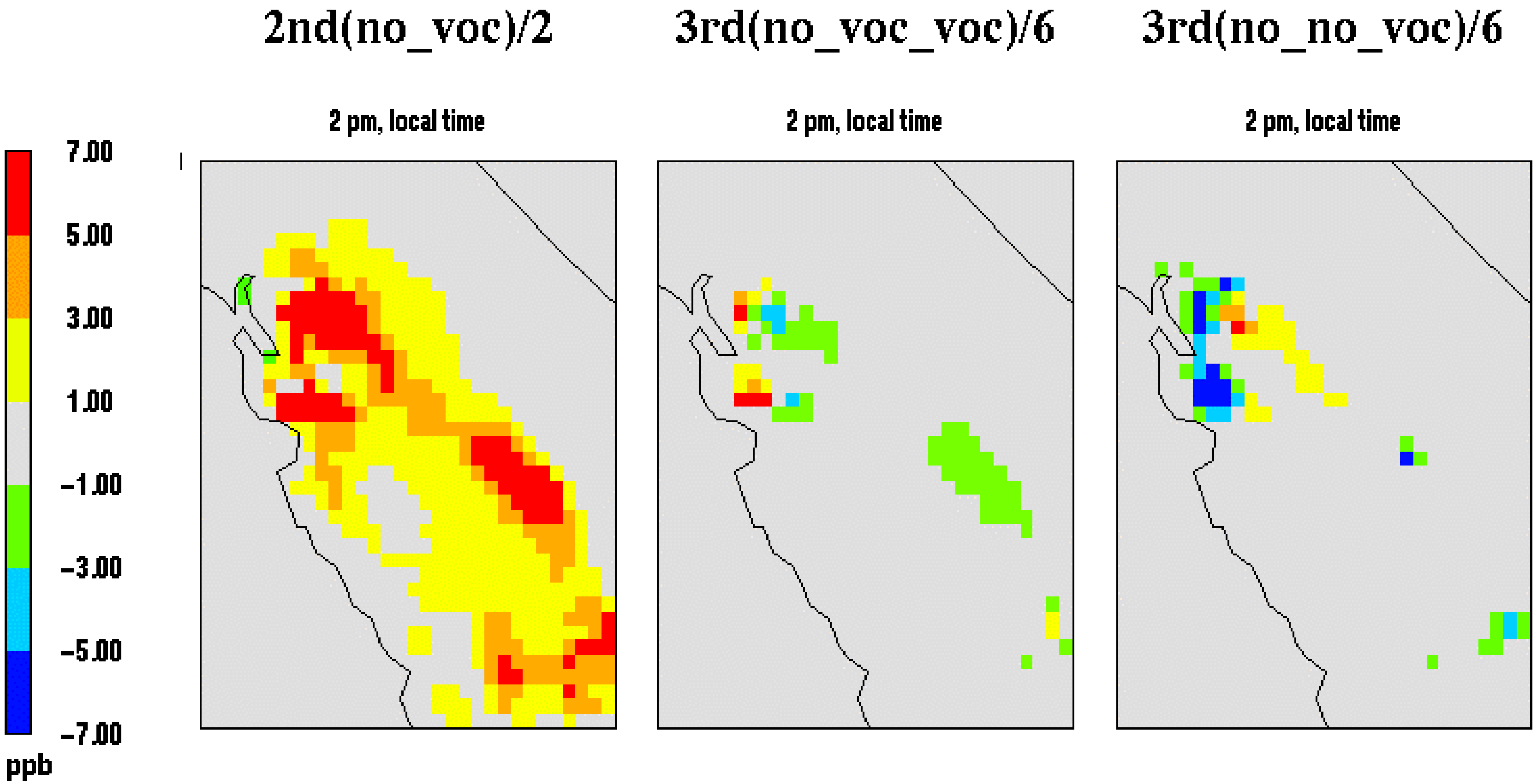
Taylor expansion: NO perturbation, VOC-limited



Taylor expansion: NO perturbation, NOx-limited



Cross-derivatives (domain-wide NO/VOC)



Taylor series expansion (contd.)

- For more than one sensitivity parameter:

$$C(x_0+\Delta x, y_0+\Delta y) = C(x_0, y_0) + \Delta x S_x^{(1)}(x_0, y_0) + \Delta y S_y^{(1)}(x_0, y_0)$$

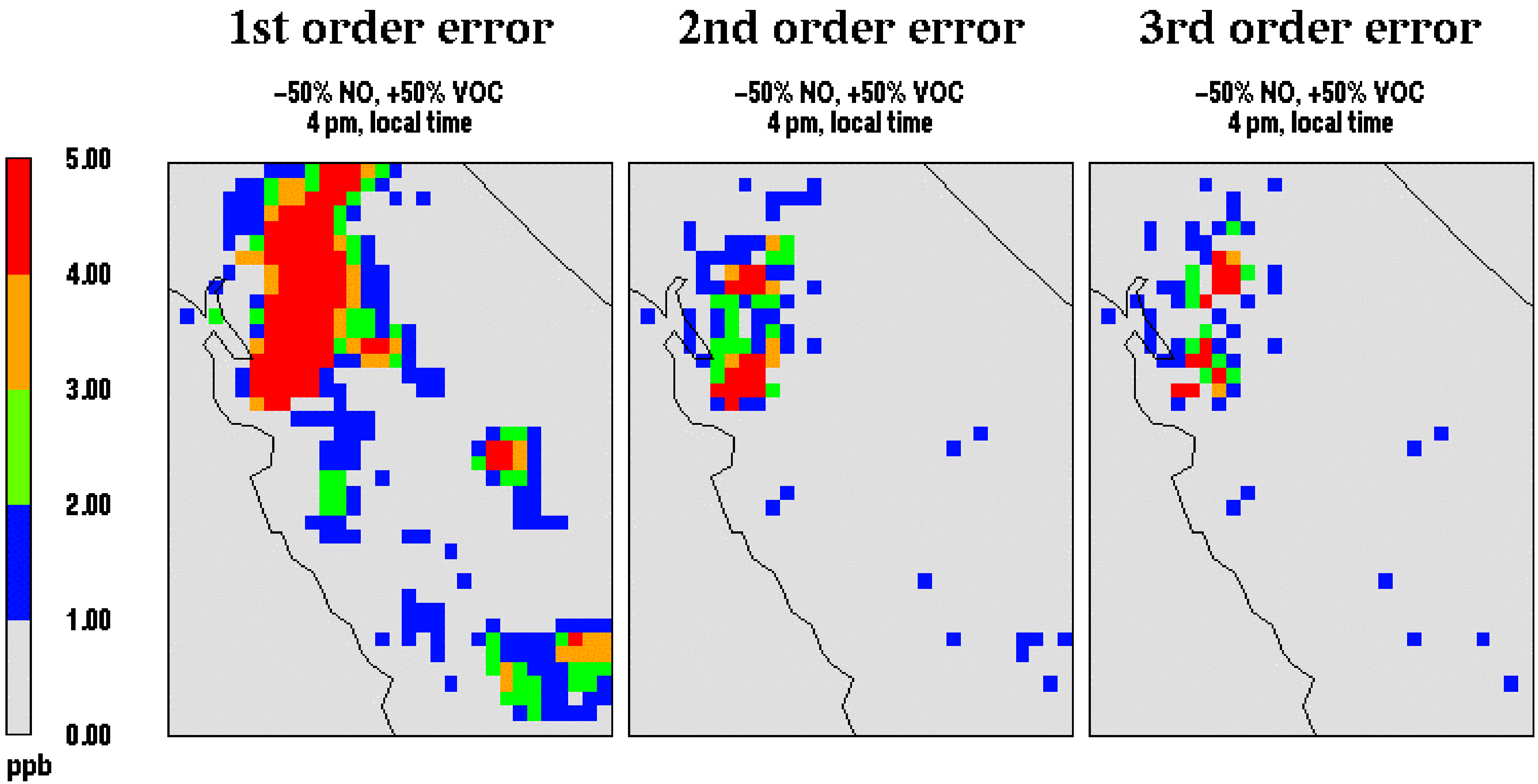
$$+\frac{(\Delta x)^2}{2} S_{xx}^{(2)}(x_0, y_0) + (\Delta x)(\Delta y) S_{xy}^{(2)}(x_0, y_0)$$

$$+\frac{(\Delta y)^2}{2} S_{yy}^{(2)}(x_0, y_0) + \frac{(\Delta x)^3}{6} S_{xxx}^{(3)}(x_0, y_0)$$

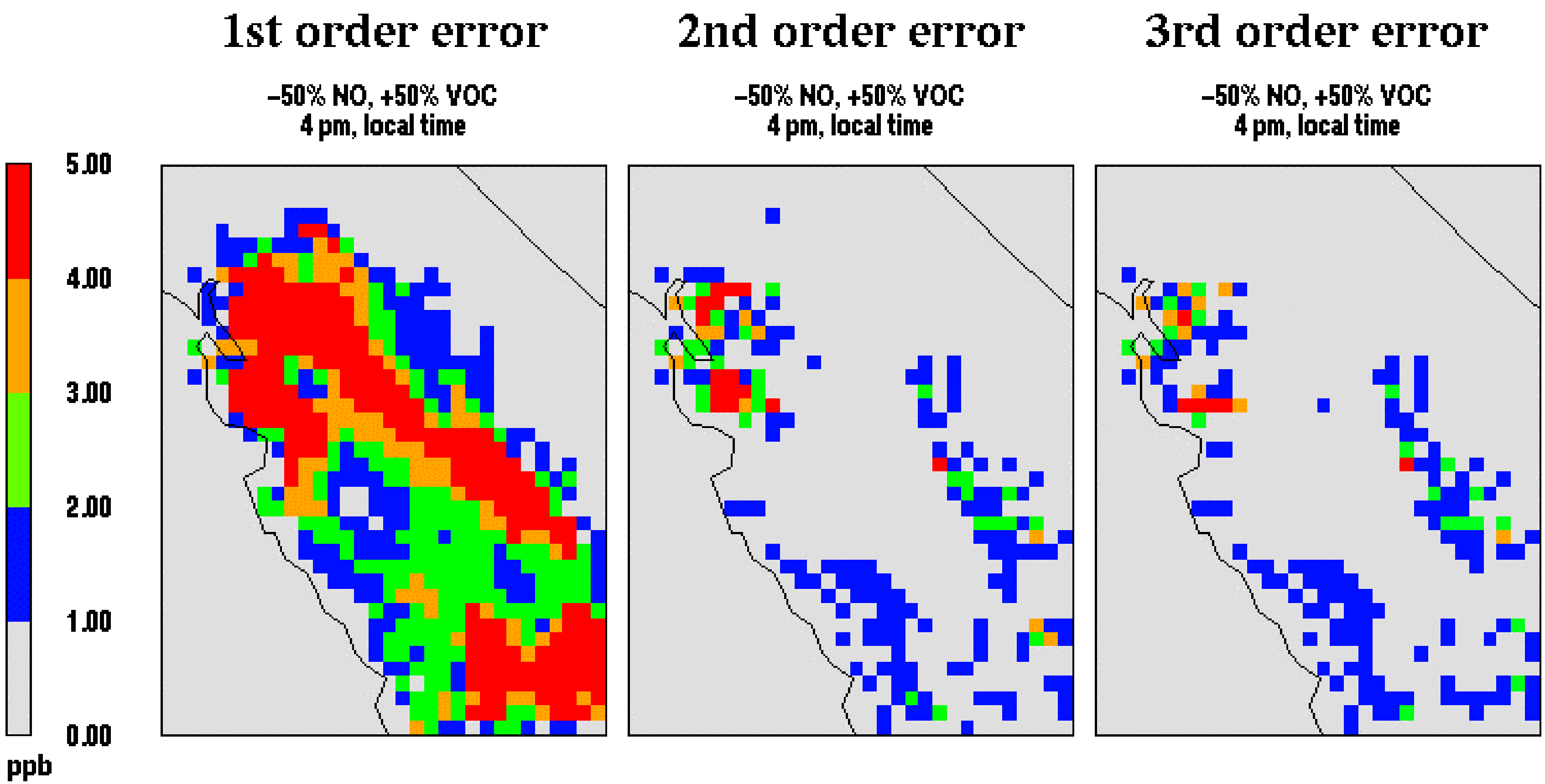
$$+\frac{(\Delta x)^2(\Delta y)}{2} S_{xxy}^{(3)}(x_0, y_0) + \frac{(\Delta x)(\Delta y)^2}{2} S_{xyy}^{(3)}(x_0, y_0)$$

$$+\frac{(\Delta y)^3}{6} S_{yyy}^{(3)}(x_0, y_0) + \ldots$$

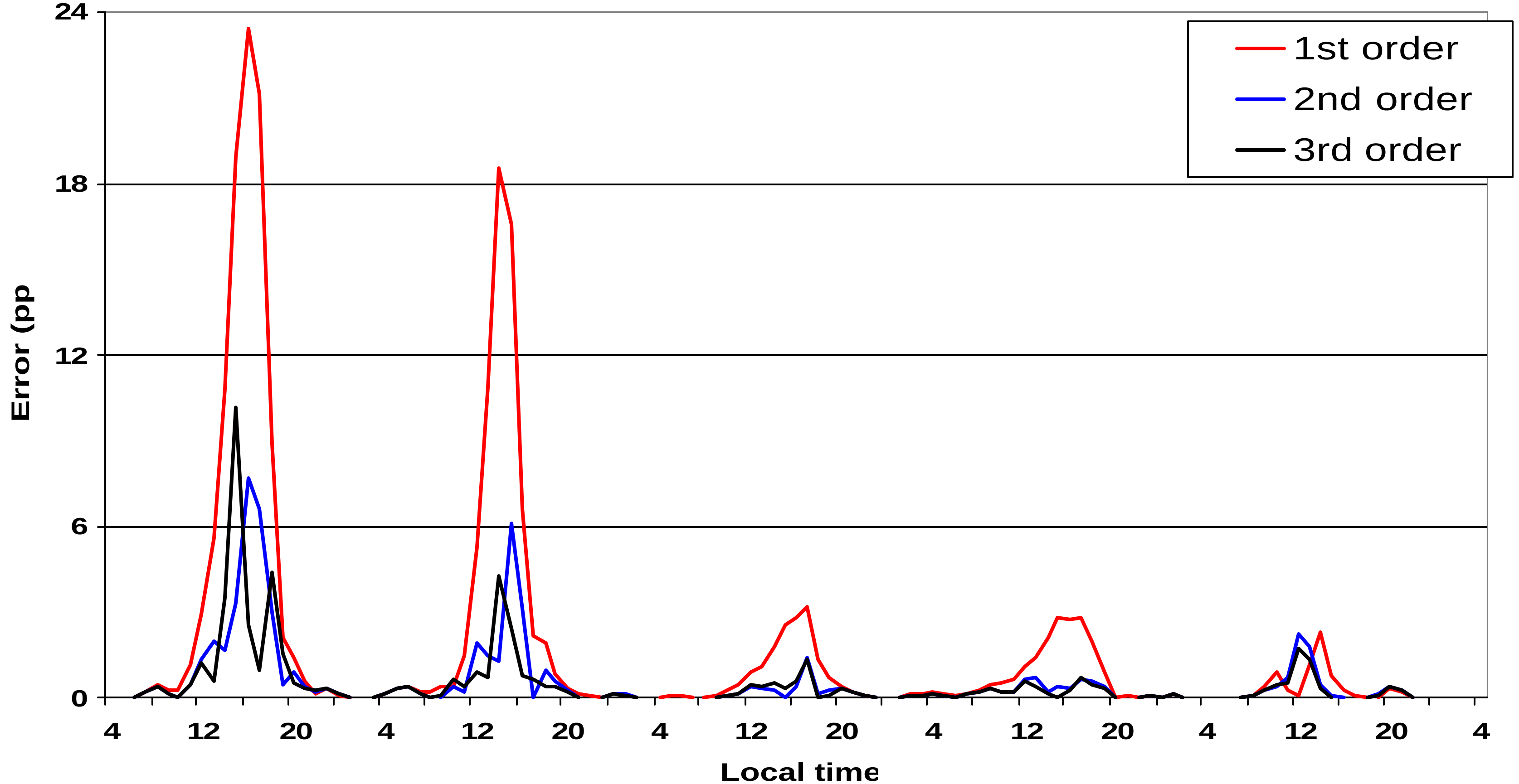
Taylor series expansion: combined perturbation



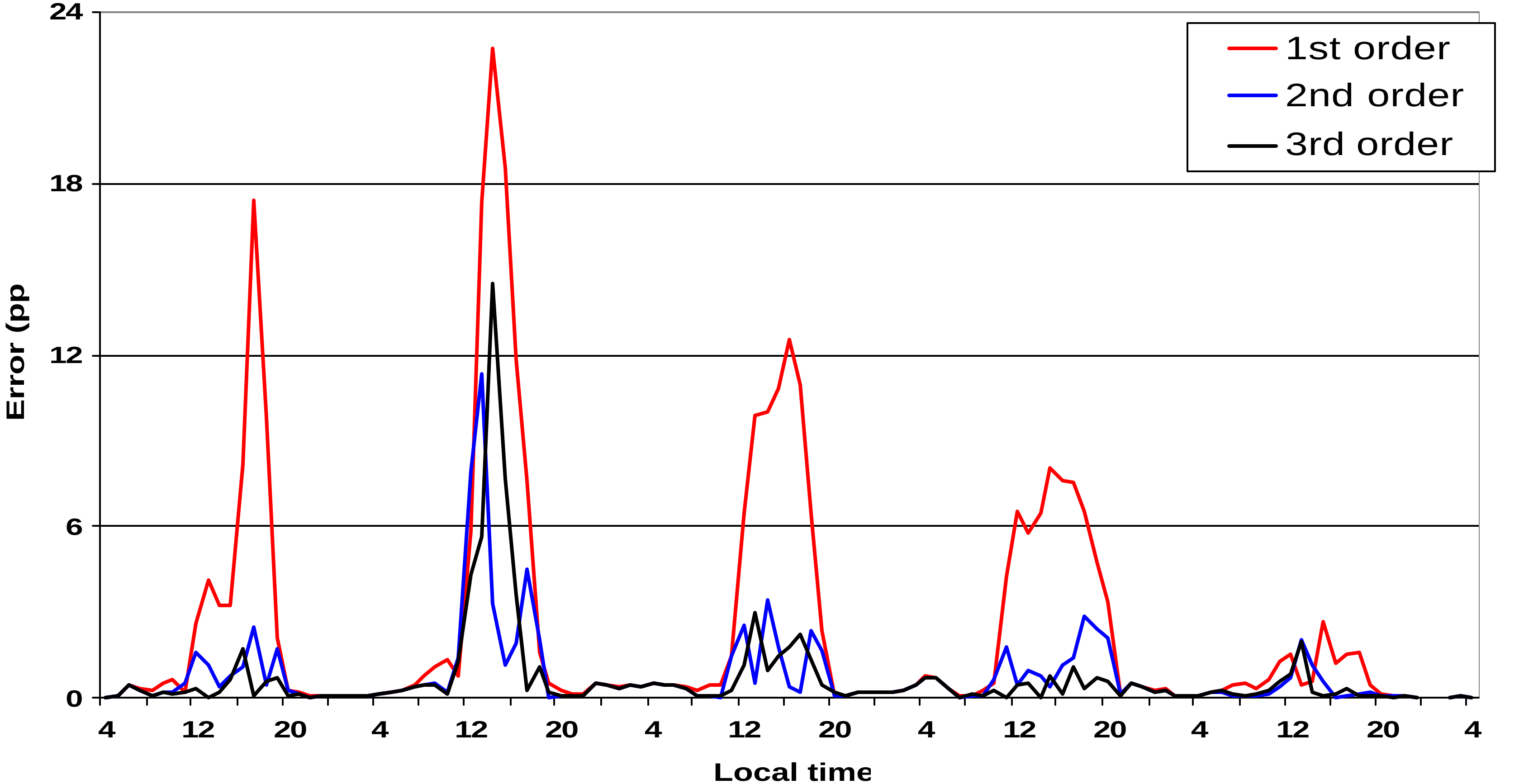
Taylor series expansion (contd.)



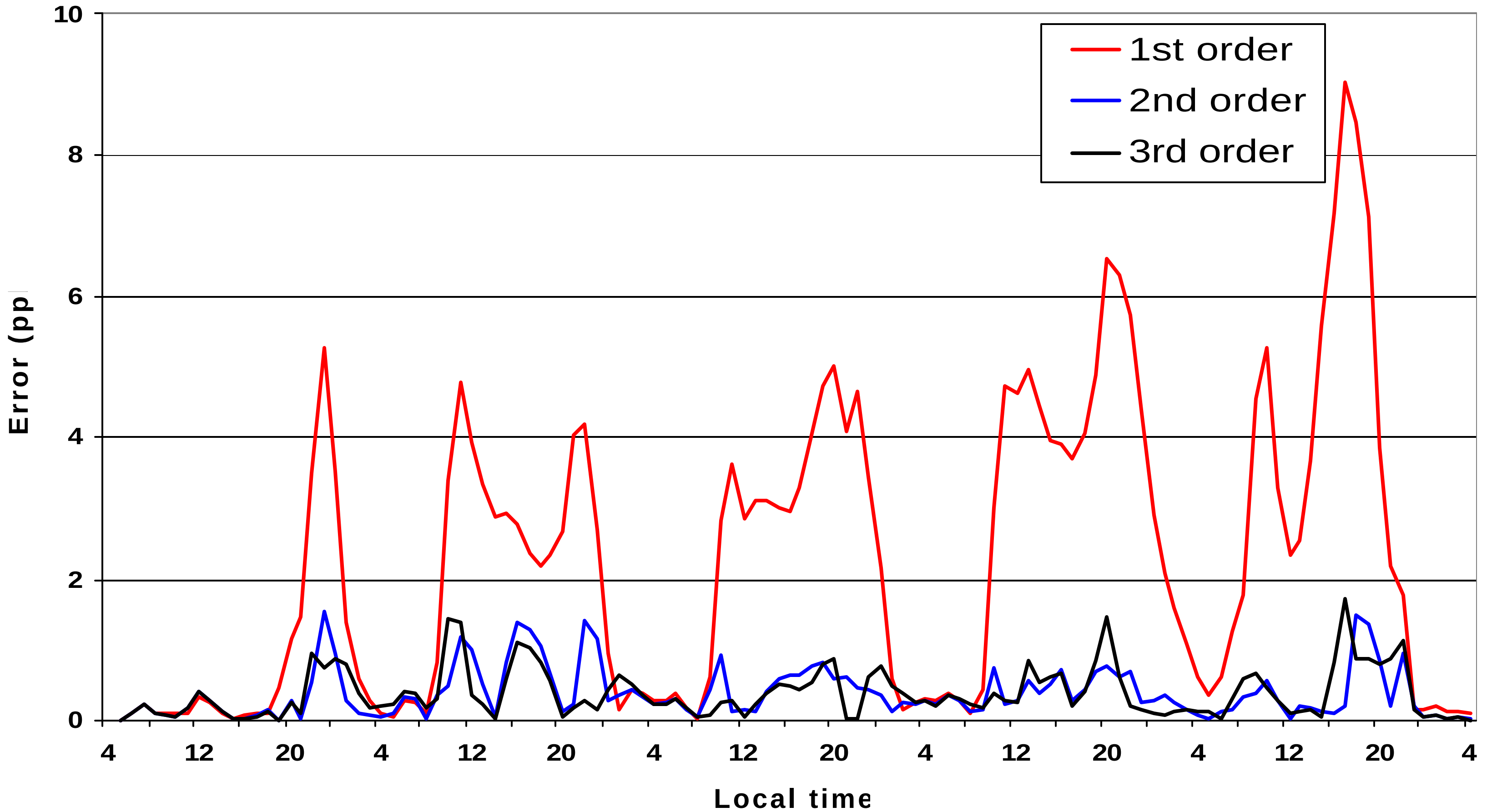
Taylor series expansion: NO/VOC perturbation, peak location



Taylor series expansion: NO/VOC perturbation, VOC-limited

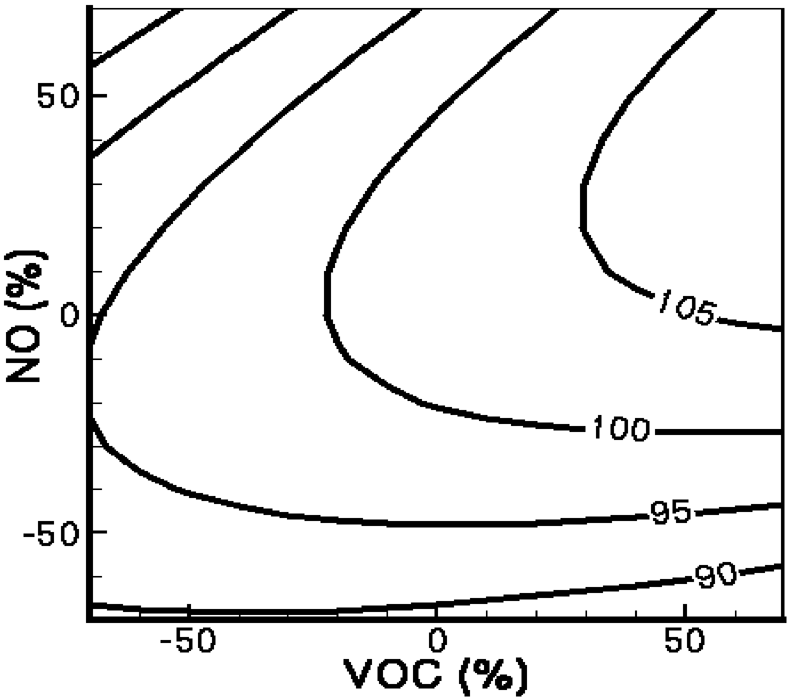


Taylor series expansion: NO/VOC perturbation, NOx-limited

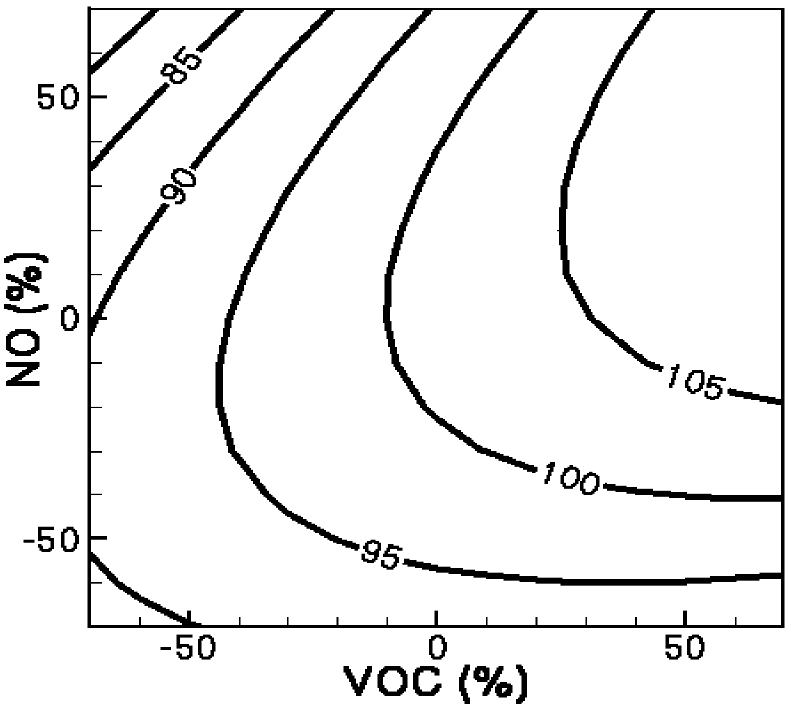


Ozone Isopleths (peak location), 2 PM

1st day

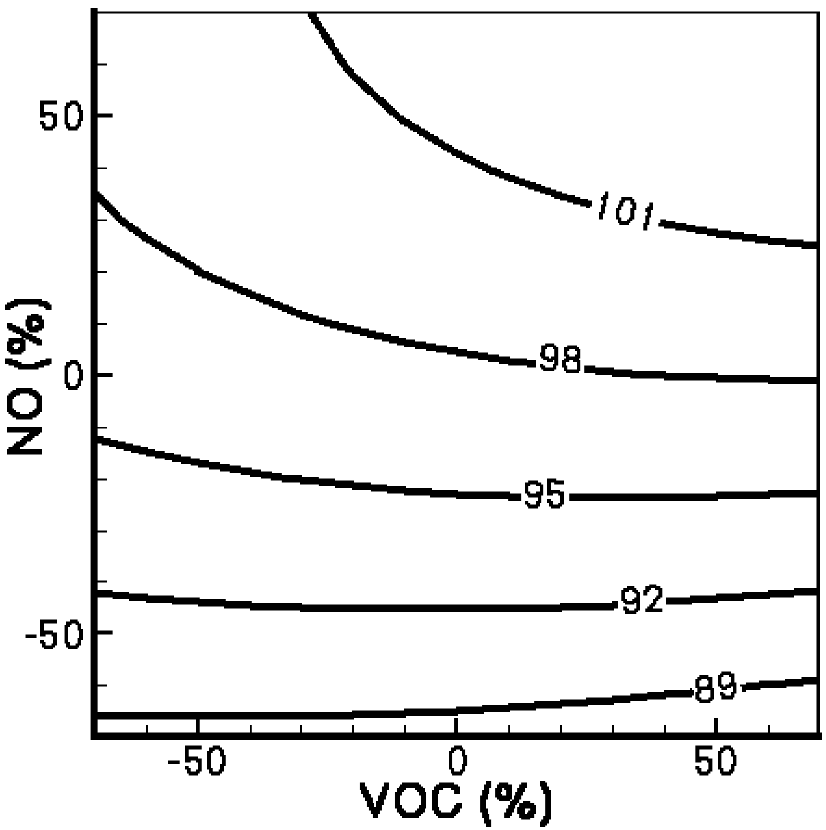


2nd day

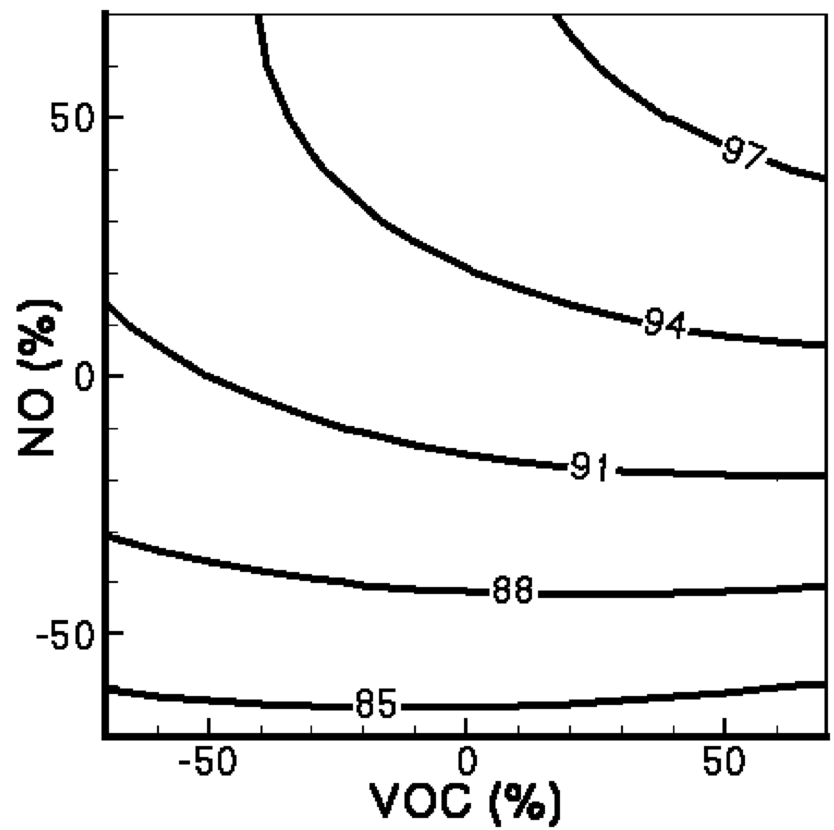


Ozone Isopleths (peak location), 2 PM (contd.)

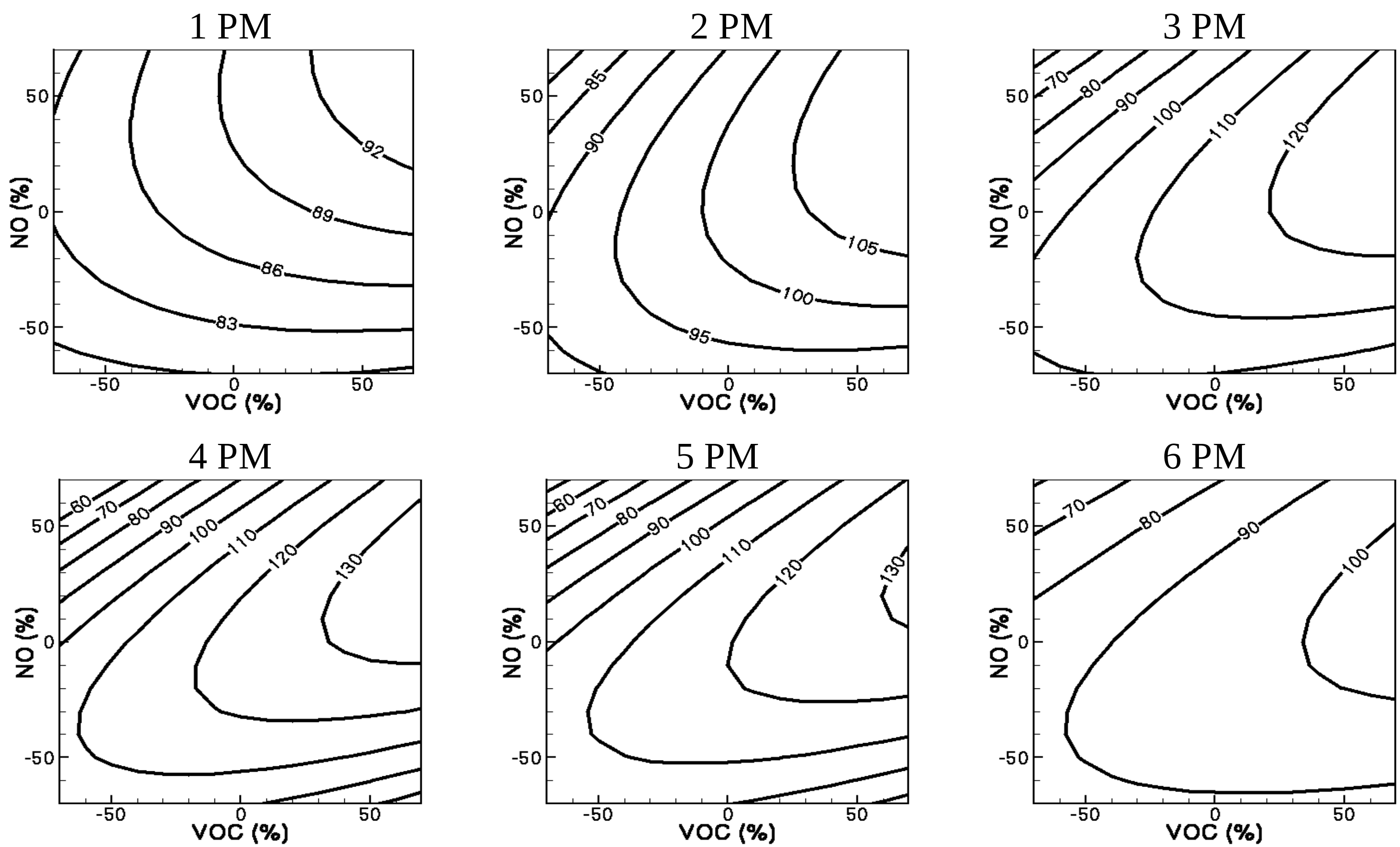
3rd day



4th day

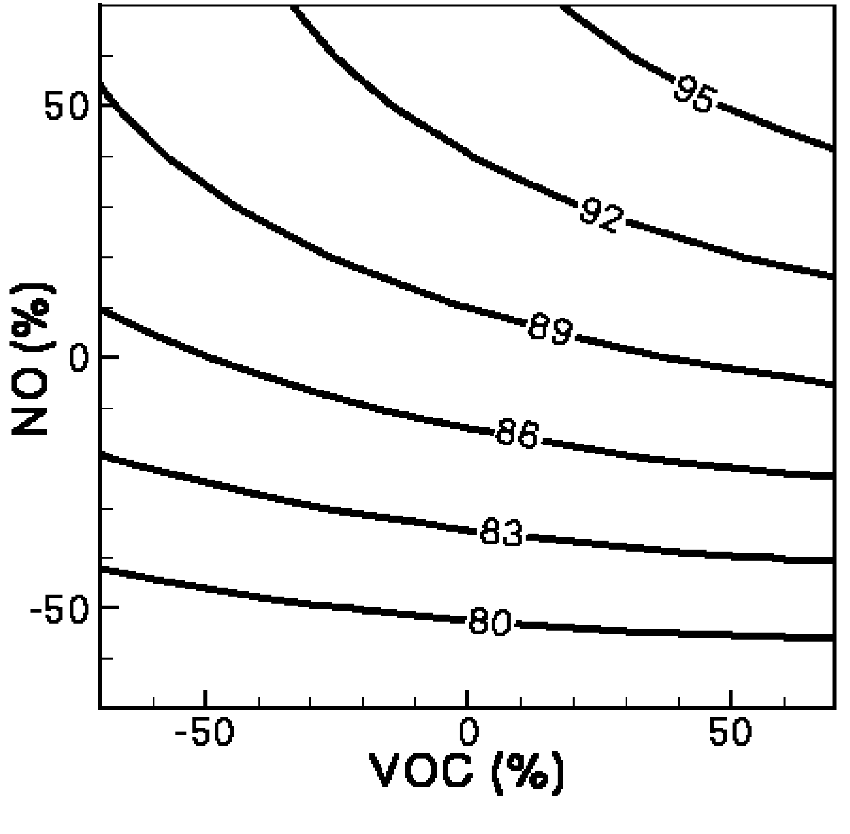


Ozone Isopleths, peak location (1-6 PM)

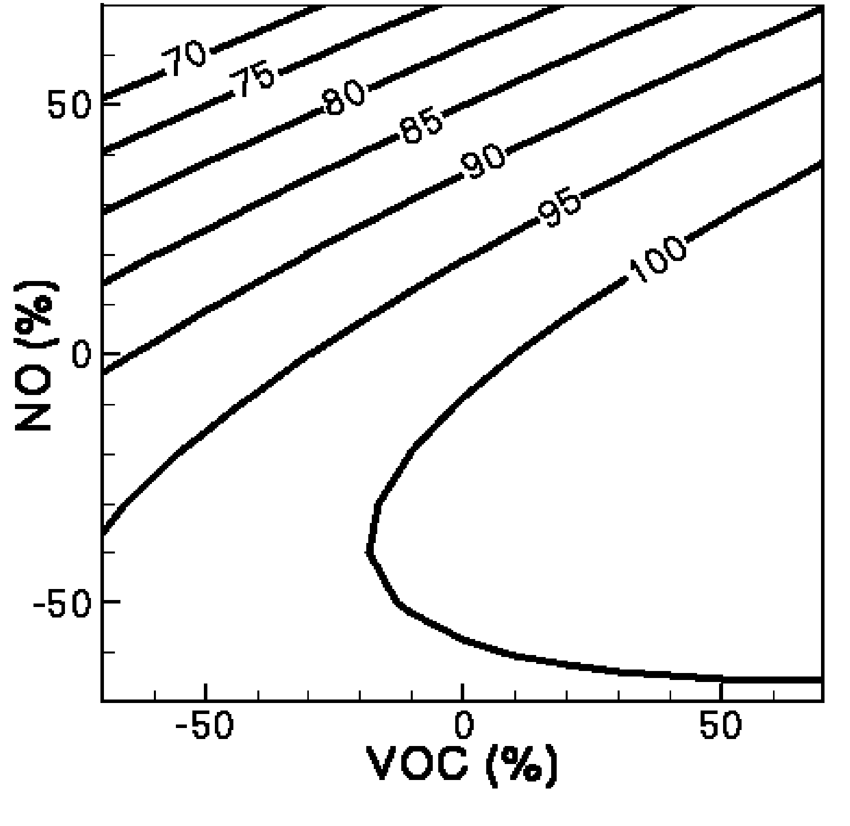


Ozone Isopleths, different regimes (2 PM)

NOx-limited



VOC-limited



Summary

- HDDM provides an efficient tool for the calculation of high-order sensitivity coefficients.
- Higher-order sensitivity analysis can, to a large extent, explain the non-linearities in atmospheric response, e.g. ozone isopleths.
- Most of the improvement in accuracy is achieved by adding the 2nd order coefficients (curvature).
- Non-linear behavior in photochemistry usually happens at the time/location of a transition between different chemical regimes.
- The location and time of the peak ozone is usually subject to a high degree of non-linearity. This is potentially important for the case of NOx control in an urban plume.

What should be done? What can be done?

- Higher-order sensitivity analysis is a completely unexplored territory. More research to come.
- Higher-order derivatives become unrealistically large -- should investigate the numerical limitations.
- Despite efficiency of HDDM, calculating higher derivatives of too many sensitivity parameters will be costly.
- HDDM provides us with new approach to old problems:
  - Cost-effectiveness analysis of control strategies,
  - Inverse modeling of non-linear systems,
  - Uncertainty analysis of organic reactivities,
  - And in general, description of any non-linearity, e.g. ozone isopleths.

Acknowledgement

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