Novel Higher-Order Sensitivities of Secondary Organic Aerosols with Respect to Their Precursor Concentrations using a Box Model

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Finite difference (Brute-Force) method is easy to implement but prone to errors

- Finite Difference (Brute-Force) method
  - Apply a perturbation to the desired variable
  - Estimate the first and second-order sensitivities
- Advantages:
  - Easy to understand
  - Easy to implement
- Disadvantages:
  - Truncation Error
  - Cancellation Error

$$\frac{\partial Y_i}{\partial X_i} \approx \frac{f(X_i+h)-f(X_i-h)}{2h}$$

Central Difference, 1st order

$$\frac{\partial^2 Y_i}{\partial X_i^2} \approx \frac{f(X_i+h)-2f(X_i)+f(X_i-h)}{h^2}$$

Central Difference, 2nd order

Direct Decoupled Method (DDM) and Adjoint are accurate but also have limitations

- Advantages:
  - Can get exact first- and second-order sensitivities in certain cases
  - Adjoint has the advantage of calculating sensitivities of one output with respect to many input variables

- Disadvantages:
  - Either is more difficult to understand than BF methods.
  - DDM – write sensitivity equations for nonlinear steps, which are common in chemistry and advection steps
  - Hard to update when the equations change

First- and Second-order sensitivities can be calculated with hyperdual numbers

- A hyperdual number is defined as follow:

$$H = x_0 + x_1\epsilon_1 + x_2\epsilon_2 + x_{12}\epsilon_{12}$$

$$\epsilon_1^2 = \epsilon_2^2 = \epsilon_{12}^2 = 0$$

$$\epsilon_1 \neq \epsilon_2 \neq \epsilon_{12} \neq 0$$

$$\epsilon_1 * \epsilon_2 = \epsilon_{12}$$

- If we perturb the desired input variable with the hyperdual number below:

$$H_h = 0 + h_1\epsilon_1 + h_2\epsilon_2 + 0\epsilon_{12}$$

- The perturbed function is:

$$f(x + h_1\epsilon_1 + h_2\epsilon_2 + 0\epsilon_1\epsilon_2)$$

First- and Second-order sensitivities can be calculated with hyperdual numbers

- Expand the terms with Taylor expansion

$$f(x + h_1\epsilon_1 + h_2\epsilon_2 + 0\epsilon_1\epsilon_2) = f(x) + \boxed{(h_1\epsilon_1 + h_2\epsilon_2)f'(x)} + \boxed{\frac{1}{2!}(h_1\epsilon_1 + h_2\epsilon_2)^2 f''(x)} +$$

$$\cancel{\frac{1}{3!}(h_1\epsilon_1 + h_2\epsilon_2)^3 f'''(x) + \cdots}$$

- The first- and second-order sensitivities are:

$$f'(x) = \frac{\epsilon_1 part[f(x+H_h)]}{h_1} = \frac{\epsilon_2 part[f(x+H_h)]}{h_2} \qquad f''(x) = \frac{\epsilon_{12} part[f(x+H_h)]}{h_1 h_2}$$

First- and Second-order sensitivities can be calculated with hyperdual numbers

- The cross sensitivities can also be calculated with the following perturbation for two different variables of interest:

$$H_1 = 0 + h_1\epsilon_1 + 0\epsilon_2 + 0\epsilon_{12} \qquad H_2 = 0 + 0\epsilon_1 + h_2\epsilon_2 + 0\epsilon_{12}$$

- The first- and cross-order sensitivities are:

$$\frac{\partial f}{\partial x_1} = \frac{\epsilon_1 part[f(x + H_1 + H_2)]}{h_1}$$

$$\frac{\partial f}{\partial x_2} = \frac{\epsilon_2 part[f(x + H_1 + H_2)]}{h_2}$$

$$\frac{\partial f^2}{\partial x_1 \partial x_2} = \frac{\epsilon_{12} part[f(x + H_1 + H_2)]}{h_1 h_2}$$

Introduction of CMAQ-hyd implementation

- Development of HDMod.f90 – a Fortran overloading library
  - Write out explicit calculation rules of hyperdual numbers
  - For example, the multiplication of two hyperdual numbers is shown below:

```
!----- Multiplication operator (*)
function hdual_mul_hdual(qleft, qright) result(res)

  implicit none
  TYPE(hyperdual), intent(in) :: qleft, qright
  TYPE(hyperdual)             :: res

  res%x = qleft%x * qright%x
  res%dx1 = qleft%x * qright%dx1 + qleft%dx1 * qright%x
  res%dx2 = qleft%x * qright%dx2 + qleft%dx2 * qright%x
  res%dx1x2 = qleft%x * qright%dx1x2 + qleft%dx1 * qright%dx2 + qleft%dx2 * qright%dx1 + qleft%dx1x2 * qright%x
```

First- and Second-Order Sensitivities from Hyperdual Calculations are exact

- Advantages
  - Intuitive, similar to finite difference
  - No truncation or cancellation errors can affect the results because the sensitivity information is stored in different variables and the calculation is exact.
  - The sensitivities do not depend on perturbation sizes
- Disadvantage
  - Slow, takes about 6 to 8 times longer than a regular CMAQ run.

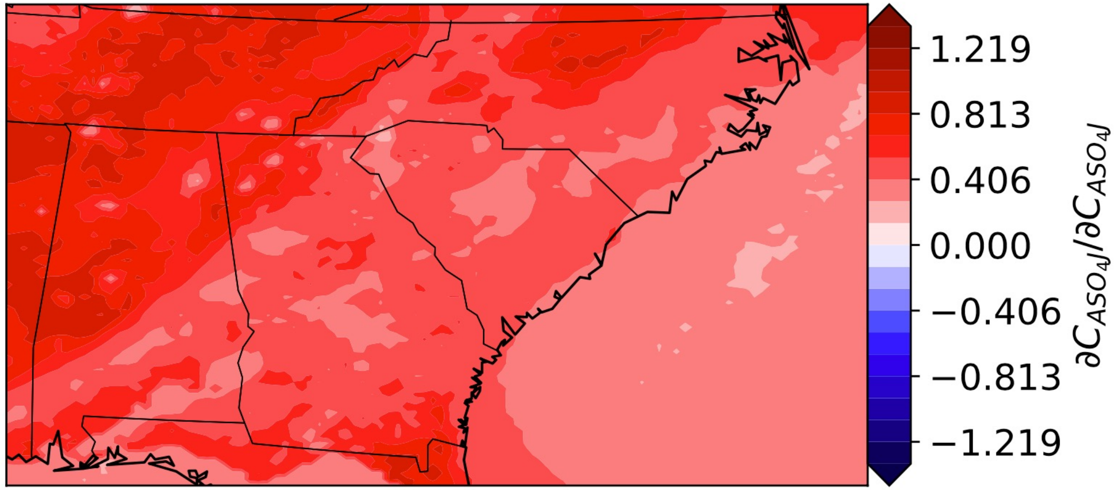
Hyperdual vs. CD evaluation framework

- BF sensitivities
  - Perturb 1st layer concentrations of a specific species, at time = 0.
  - Run the model twice, with positive and negative perturbations.
  - Calculate the central difference sensitivity.

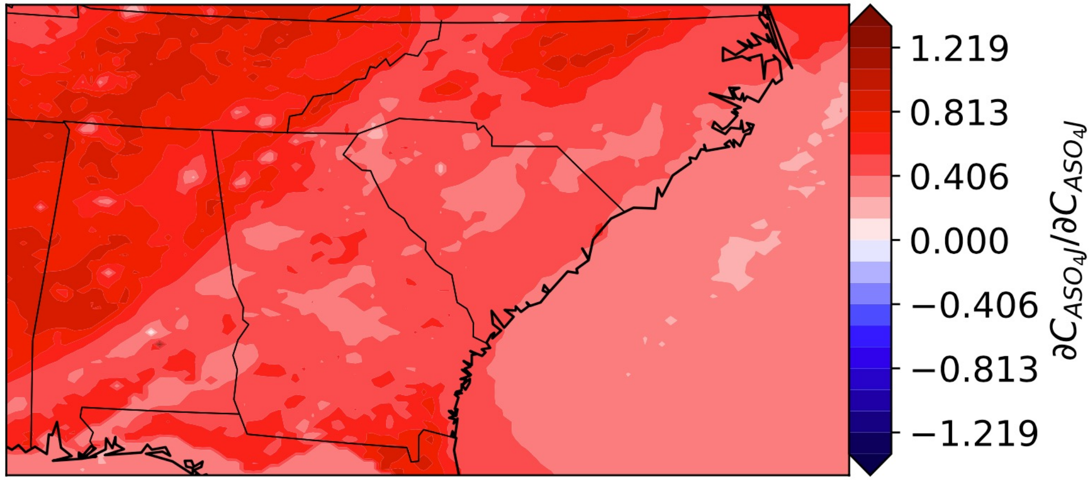
- Hyperdual Sensitivities
  - Perturb the 1st layer concentrations of a specific species with the hyperdual perturbation, at time = 0
  - Run the hyperdual model.
  - Calculate the sensitivity from ϵ_1, ϵ_2, or ϵ_{12} part.

Example of Hyperdual Sensitivity Calculations

- The sensitivity calculated here is $$\frac{\sum_{l=1}^{L} \partial C_{ASO_4J_{c,r,l,t=6hr}}}{\partial C_{ASO_4J_{c,r,l=1,t=0hr}}}$$



Hyperdual

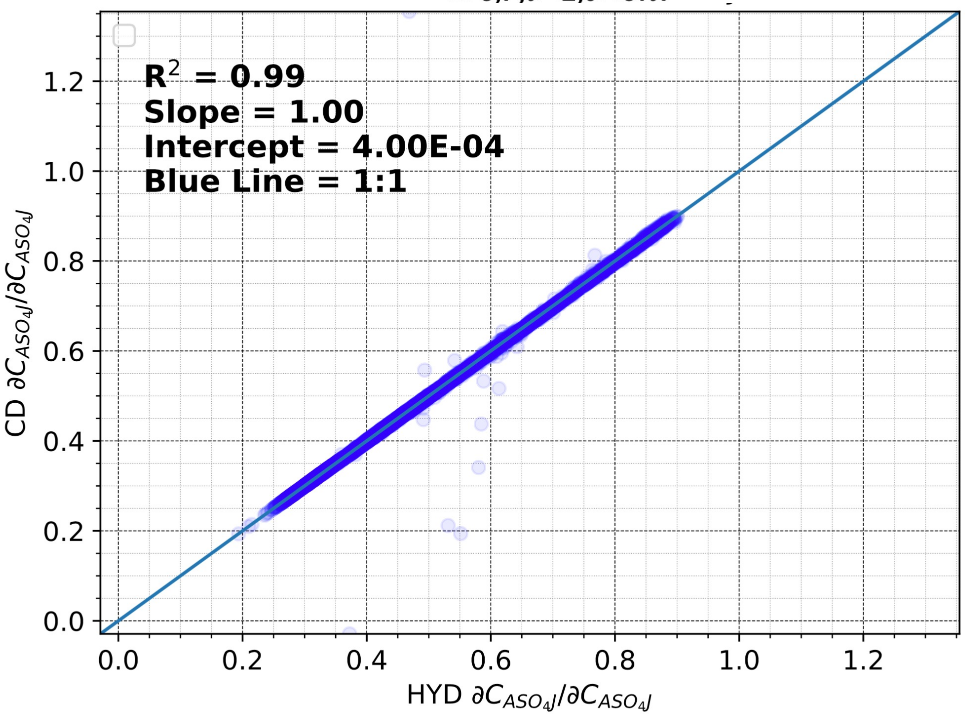


Central Difference

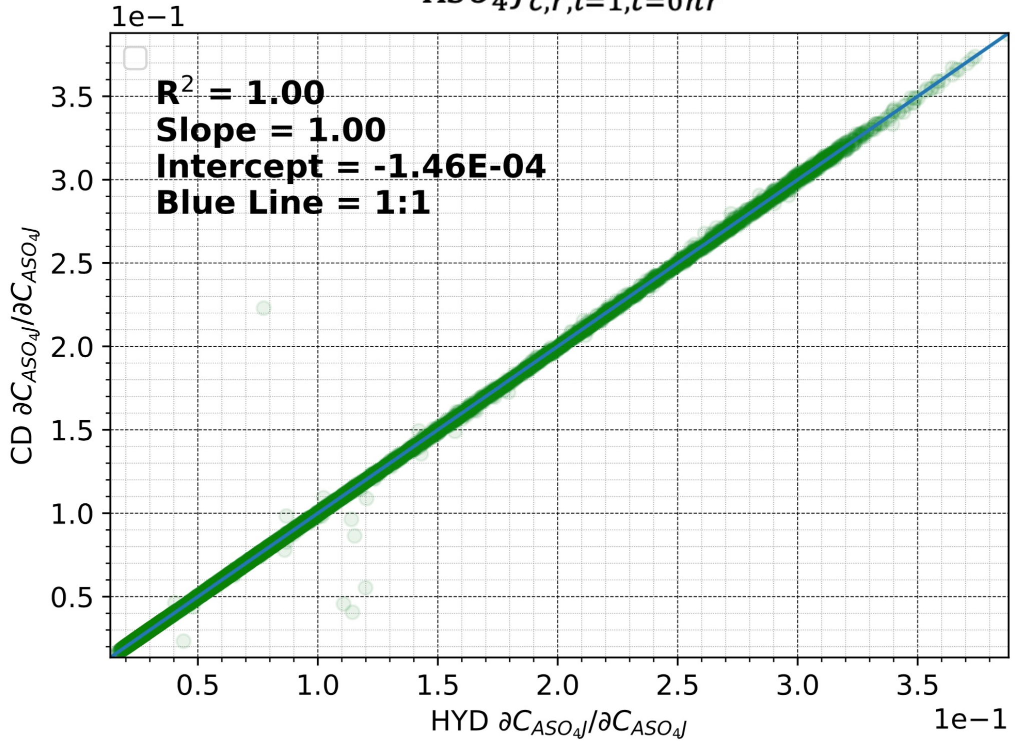
1:1 plots show that the sensitivities agree with each other

$$\frac{\sum_{l=1}^{L} \partial C_{ASO_4J_{c,r,l,t=6hr}}}{\partial C_{ASO_4J_{c,r,l=1,t=0hr}}}$$

$$\frac{\partial C_{ASO_4J_{c,r,l=2,t=6hr}}}{\partial C_{ASO_4J_{c,r,l=1,t=0hr}}}$$



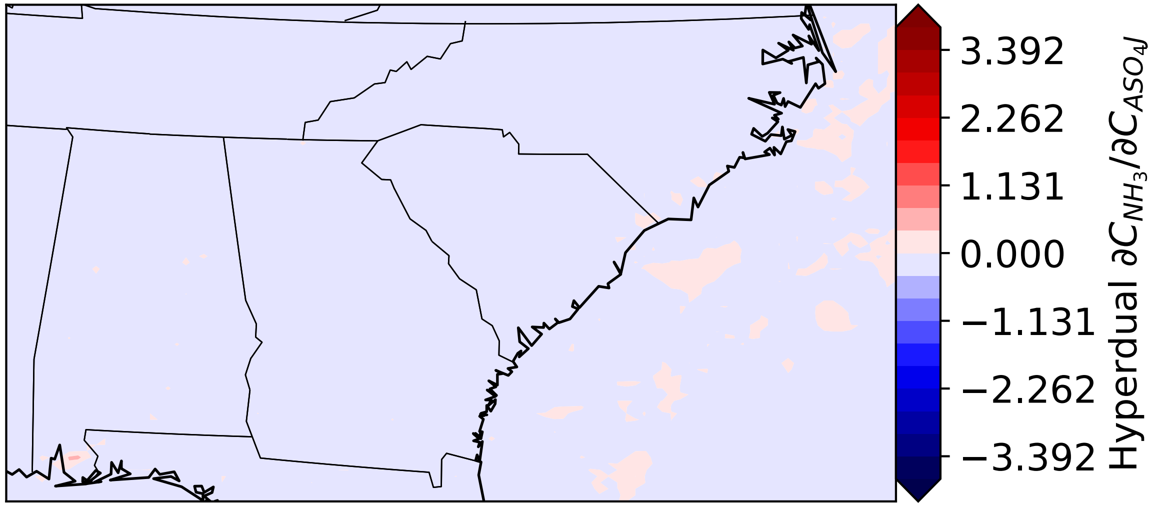
Sum of All Layers, CD vs. HYD

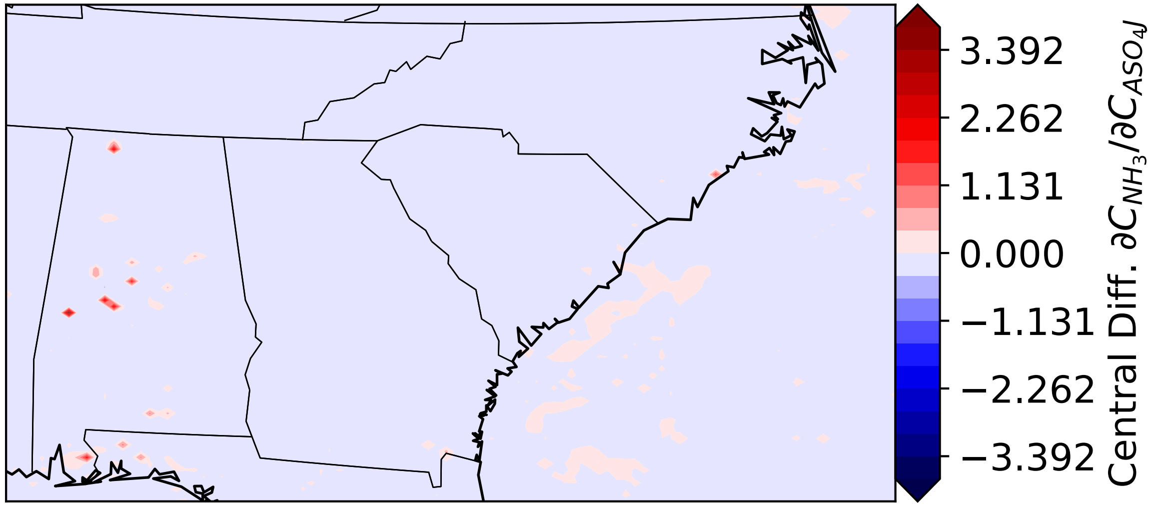


Second Layer, CD vs. HYD

Example of Hyperdual Sensitivity Calculations

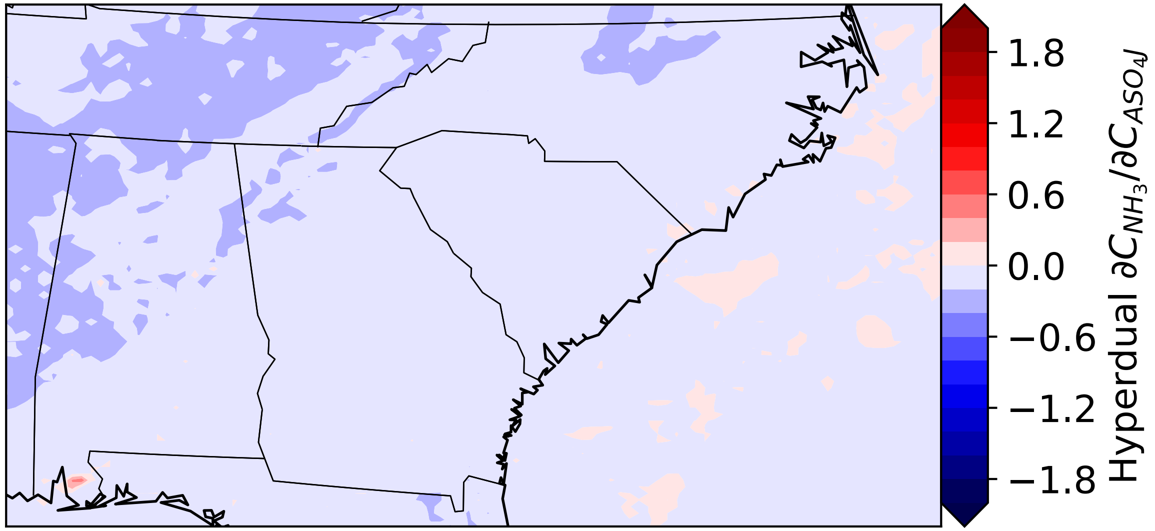
- The sensitivity calculated here is $\dfrac{\sum_{l=1}^{L} \partial C_{{NH_3}_{c,r,l,t=6hr}}}{\partial C_{{ASO_4J}_{c,r,l=1,t=0hr}}}$

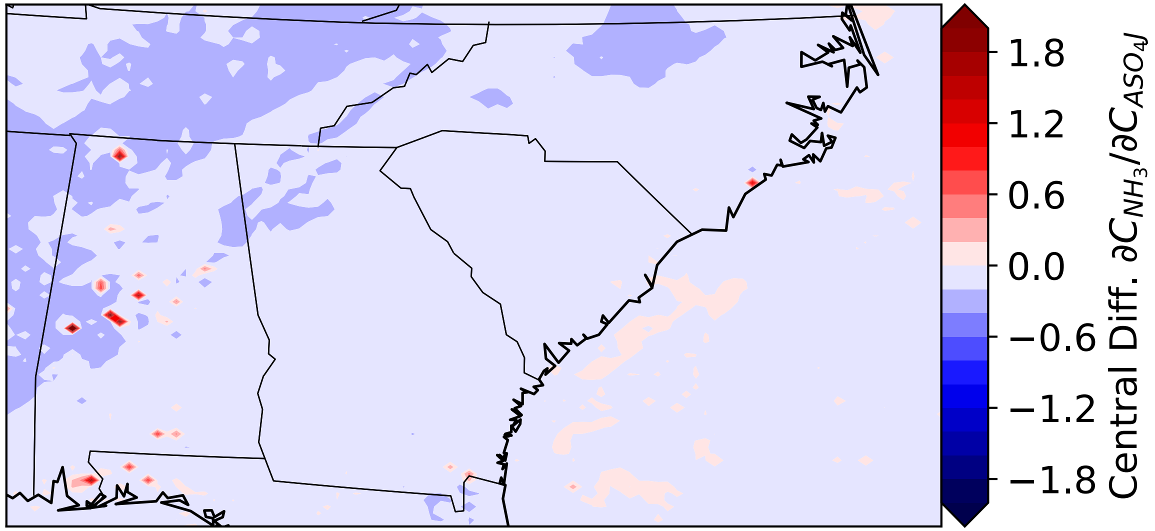




Example of Hyperdual Sensitivity Calculations

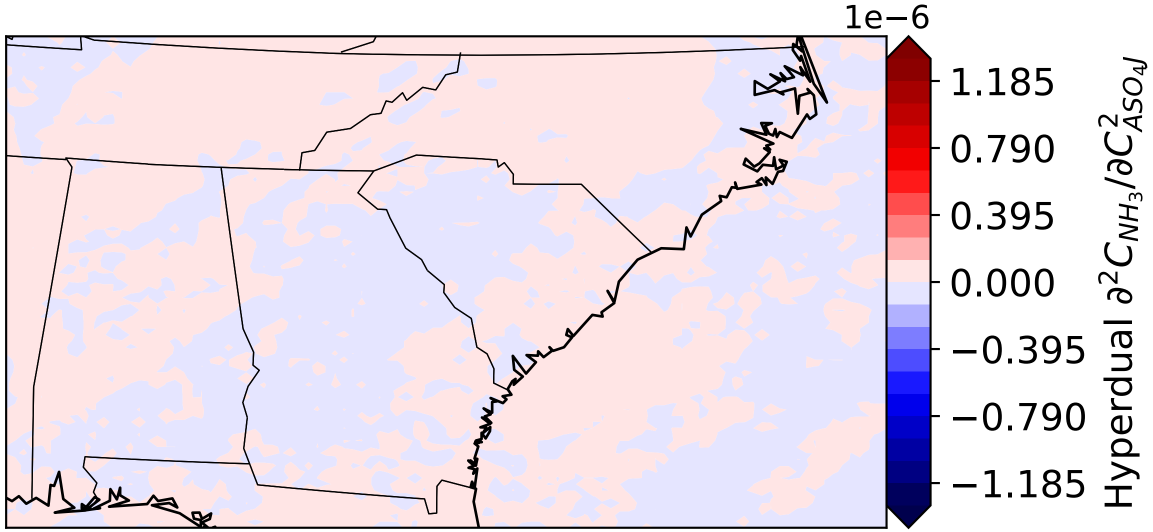
- The sensitivity calculated here is $$\frac{\sum_{l=1}^{L} \partial C_{NH_3}{}_{c,r,l,t=6hr}}{\partial C_{ASO_4J}{}_{c,r,l=1,t=0hr}}$$

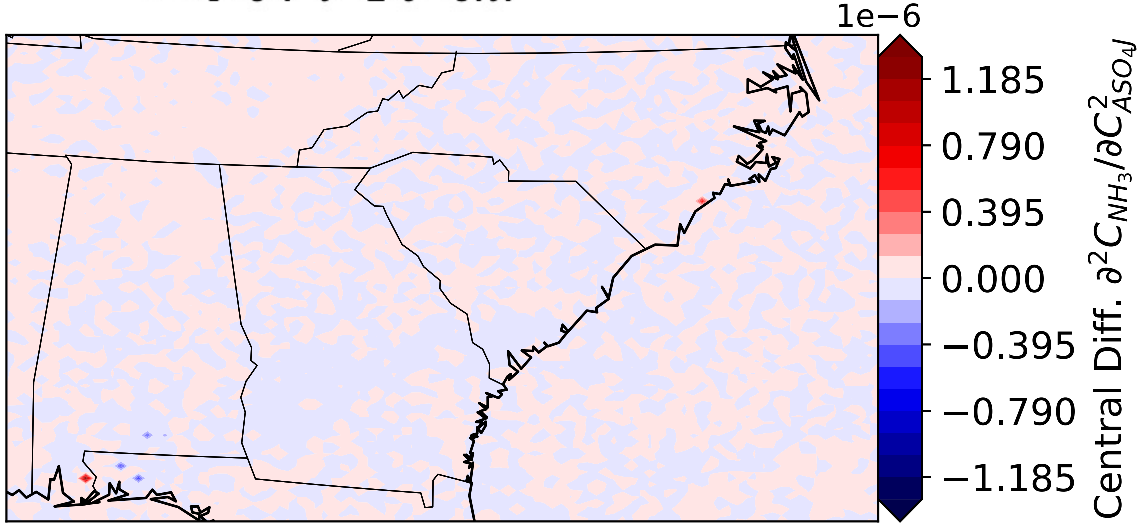




Example of Hyperdual Sensitivity Calculations

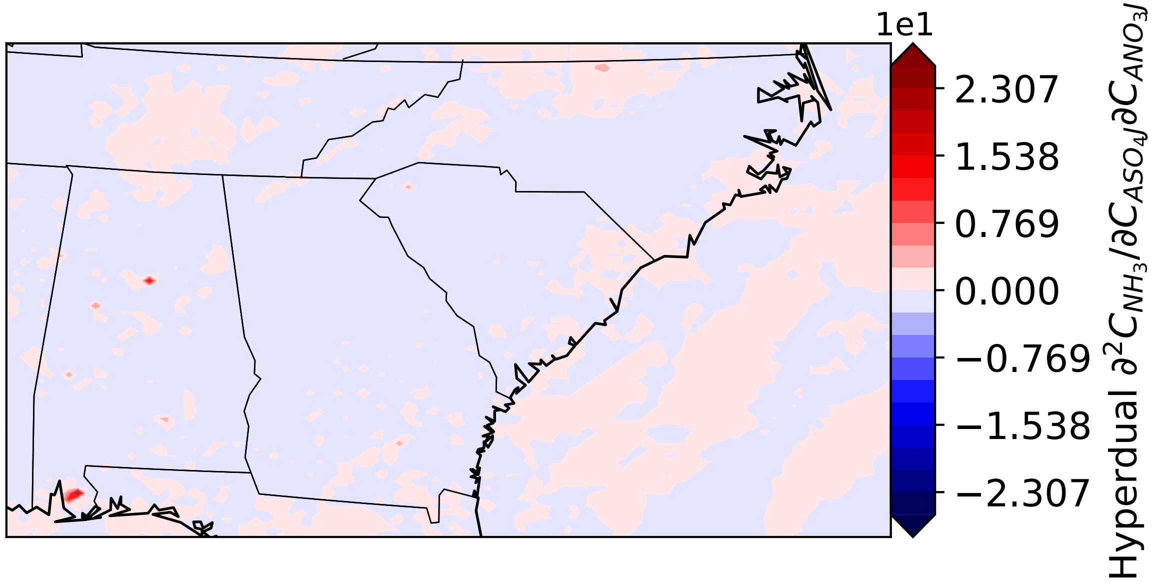
- The sensitivity calculated here is $\dfrac{\sum_{l=1}^{L} \partial^2 C_{NH_{3}{}_{c,r,l,t=6hr}}}{\partial C^2_{ASO_4J_{c,r,l=1,t=0hr}}}$

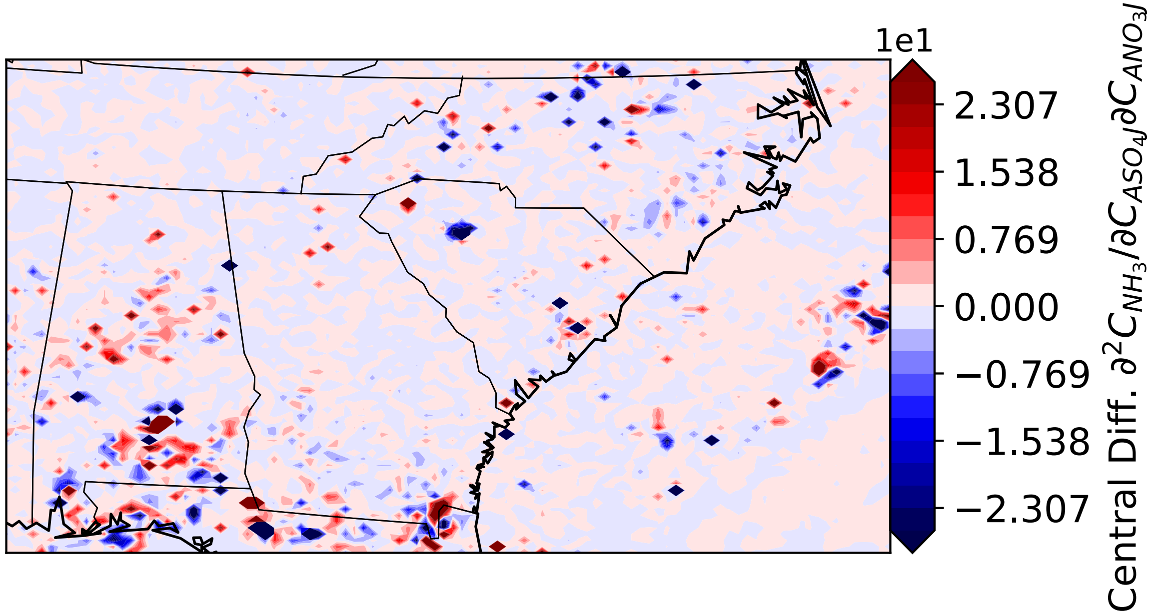




Cross Sensitivities of NH_3 with respect to ASO_4J and ANO_3J

- The sensitivity calculated here is $$\frac{\partial^2 C_{NH_3}{}_{c,r,l=1,t=6hr}}{\partial C_{ASO_4J}{}_{c,r,l=1,t=0hr}\,\partial C_{ANO_3J}{}_{c,r,l=1,t=0hr}}$$





Conclusion and Future Directions

- We present the partial development of the CMAQ-hyd model and the calculation of exact first and second order sensitivities.
- We will continue modifying the CMAQ modules to use hyperdual numbers.
- We will apply CMAQ-hyd to understand complex problems which are difficult to solve with traditional methods.