Application of the hyperdual-step method in the Community Multiscale Air Quality Model (CMAQ) for the assessment of aerosol formation from volatile chemical products (VCPs)

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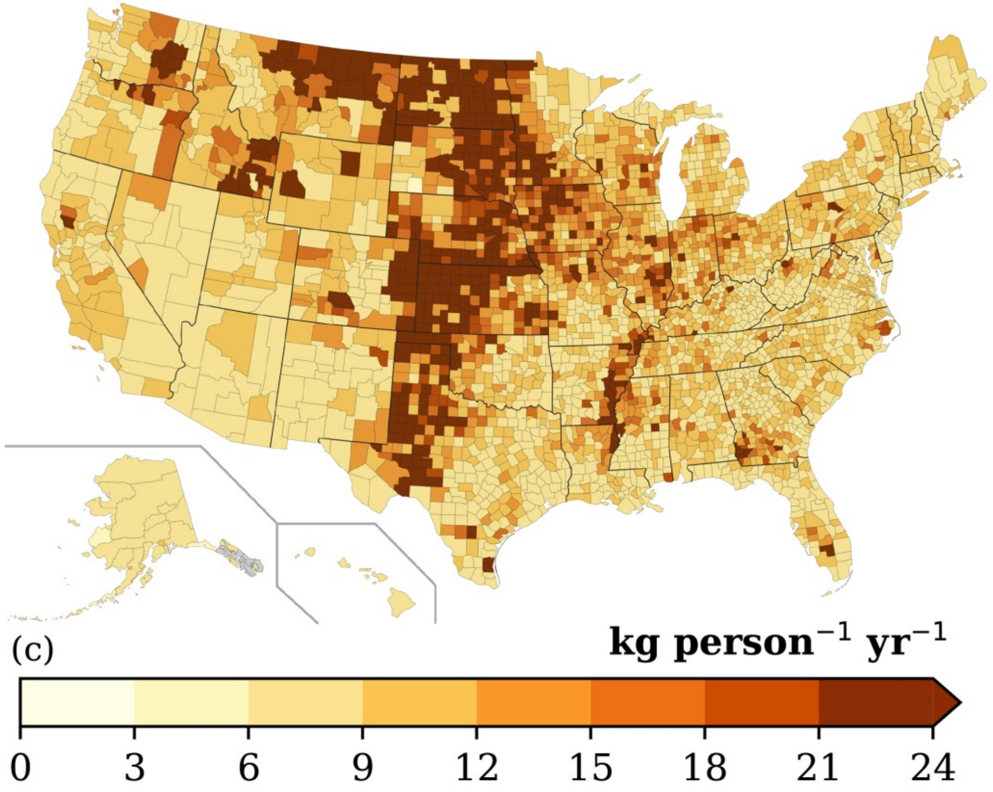
Substantial VCP contributions to VOC emissions

- Volatile chemical products (VCPs), such as personal care products and coatings, have become a major concern for human health due to direct VOC exposure and secondary organic aerosol (SOA) formation (McDonald et al., 2018).
- Seltzer et al. (2021b) found that VCPs contribute ~10% of population-weighted SOA mass in continental US.
- Further reducing the $PM_{2.5}$ concentrations in the US requires a deeper understanding of VCP emissions and SOA contributions to $PM_{2.5}$ (Qin et al., 2021).

VCP emissions for continental U.S. modeling

- Seltzer et al. (2021) developed a new framework, VCPy, to estimate the VCP emissions in CONUS.
- The average per-capita emissions from VCPs are 9.5 kg per-person per-year.
- The nationwide total is broadly consistent with the EPA's 2017 National Emission Inventory (NEI); however, county-level and categorical estimates can be different.



county-level per capita VCP emissions from Seltzer et al. (2021a)

CMAQ-hyd: a novel sensitivity analysis method in CMAQ

- We have applied the **hyperdual-step method** in CMAQ to generate a novel, augmented version of CMAQ called **CMAQ-hyd** (Liu et al., GMDD, 2023).
- This model can compute first- and second-order sensitivities of output concentrations with respect to input emissions to machine precision.
- We applied the model to assess aerosol formation from VCPs in CMAQ.

Properties of hyperdual numbers

- A hyperdual number *H* was defined by Fike and Alonso (2011) as:

$$H = a_0 + a_1\epsilon_1 + a_2\epsilon_2 + a_3\epsilon_{12}$$

- The a_0, a_1, a_2 and a_3 are real and ϵ_1, ϵ_2, and ϵ_{12} are non-real parts.
- The key property of hyperdual numbers is that the squares of three non-real components are zero, while they are not equal to zero or any value in the real space.

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

Exact calculation of derivatives with hyperdual numbers

- Suppose that we have a function of interest, $f(x)$.
- We multiply the variable of interest by $H_a = 1.0 + \ a_1\epsilon_1 + a_2\epsilon_2$ and expand with Taylor expansion.

$$f(xH_a) = f(x) + (xa_1\epsilon_1 + xa_2\epsilon_2)f'(x) + \ \frac{1}{2!}(xa_1\epsilon_1 + xa_2\epsilon_2)^2 f''(x) + \frac{1}{3!}(xa_1\epsilon_1 + xa_2\epsilon_2)^3 f'''(x) + \cdots$$

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Elimination of higher order terms by definition

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$$(x_1\epsilon_1 + x_2\epsilon_2)^2 = x_1^2\epsilon_1^2 + 2x_1x_2\epsilon_1\epsilon_2 + x_2^2\epsilon_2^2$$

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$$(x_1\epsilon_1 + x_2\epsilon_2)^2 = \cancel{x_1^2\epsilon_1^2} + 2x_1x_2\epsilon_1\epsilon_2 + \cancel{x_2^2\epsilon_2^2}$$

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$$(x_1\epsilon_1 + x_2\epsilon_2)^3 = x_1^3\epsilon_1^3 + 3x_1^2x_2\epsilon_1^2\epsilon_2 + 3x_1x_2^2\epsilon_1\epsilon_2^2 + x_2^3\epsilon_2^3$$

$$(x_1\epsilon_1 + x_2\epsilon_2)^4 = x_1^4\epsilon_1^4 + 4x_1^3x_2\epsilon_1^3\epsilon_2 + 6x_1^2x_2^2\epsilon_1^2\epsilon_2^2 + 4x_1x_2^3\epsilon_1\epsilon_2^3 + x_2^4\epsilon_2^4$$

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$$(x_1\epsilon_1 + x_2\epsilon_2)^4 = x_1^4\epsilon_1^4 + 4x_1^3x_2\epsilon_1^3\epsilon_2 + 6x_1^2x_2^2\epsilon_1^2\epsilon_2^2 + 4x_1x_2^3\epsilon_1\epsilon_2^3 + x_2^4\epsilon_2^4$$

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$$(x_1\epsilon_1 + x_2\epsilon_2)^4 = x_1^4\epsilon_1^4 + 4x_1^3x_2\epsilon_1^3\epsilon_2 + 6x_1^2x_2^2\epsilon_1^2\epsilon_2^2 + 4x_1x_2^3\epsilon_1\epsilon_2^3 + x_2^4\epsilon_2^4$$

The higher-order exponentials in this form will all be 0.

Exact calculation of derivatives with hyperdual numbers

- Suppose that we have a function of interest, $f(x)$.
- We multiply the variable of interest by $H_a = 1.0 + a_1\epsilon_1 + a_2\epsilon_2$ and expand with Taylor expansion.

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$$\Rightarrow \quad f(xH_a) = f(x) + (xa_1\epsilon_1 + xa_2\epsilon_2)f'(x) + x^2 a_1 a_2 \epsilon_{12} f''(x)$$

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- **The real value, first-order derivative, and second-order derivatives are separated into different terms.**

New hyperdual operator overloading library in Fortran

- To apply the hyperdual-step method in chemical transport models, we must first develop an operator overloading library which includes mathematical operation rules for hyperdual numbers.
- Over 400 operations involved in the CTM are defined by this operator overloading library.
- These operations are tested offline for their accuracy in both real-number calculations and sensitivity calculations before being applied to the CTM of interest.

HDMod.f90

```
!----- Addition operator (+)
function hdual_plus_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%dx1 + qright%dx1
  res%dx2   = qleft%dx2 + qright%dx2
  res%dx1x2 = qleft%dx1x2 + qright%dx1x2

end function hdual_plus_hdual
```

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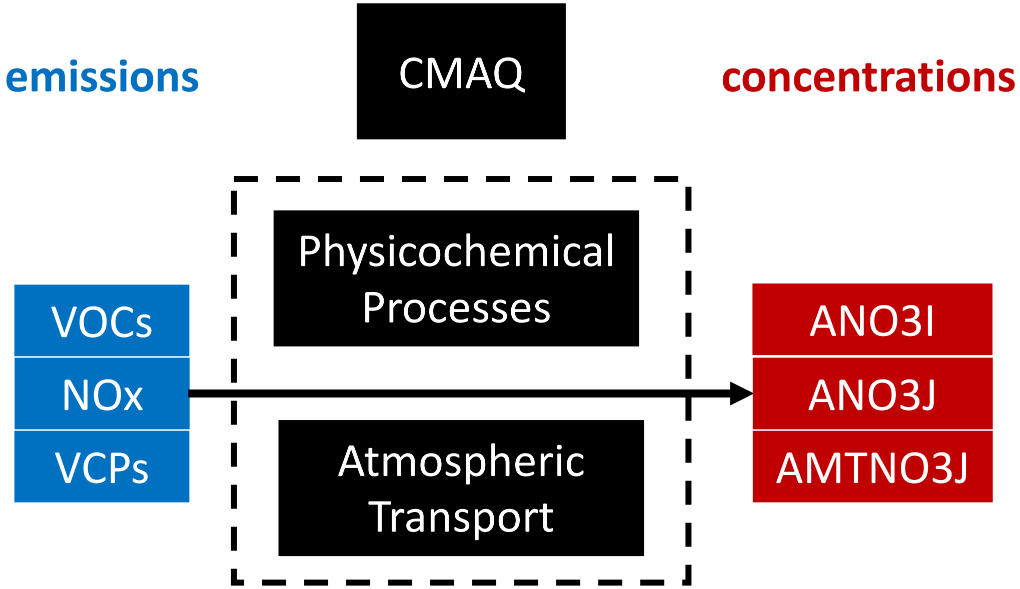
```



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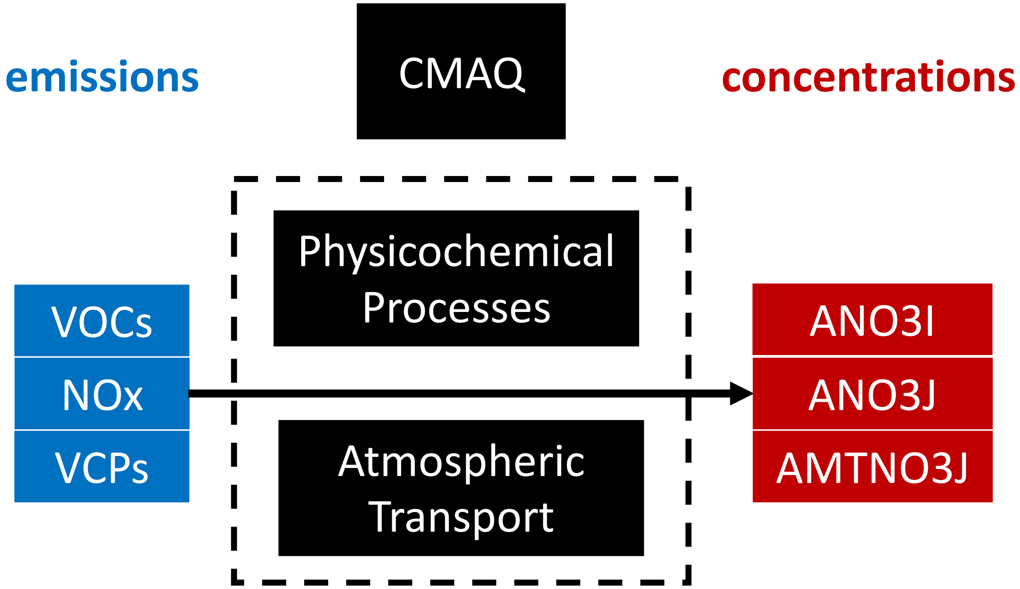

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```

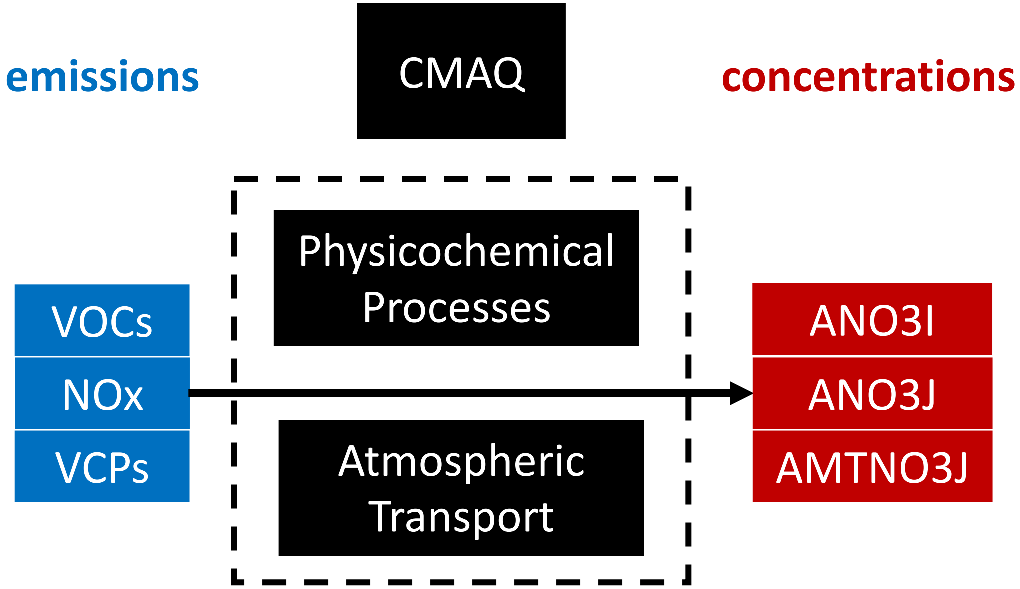

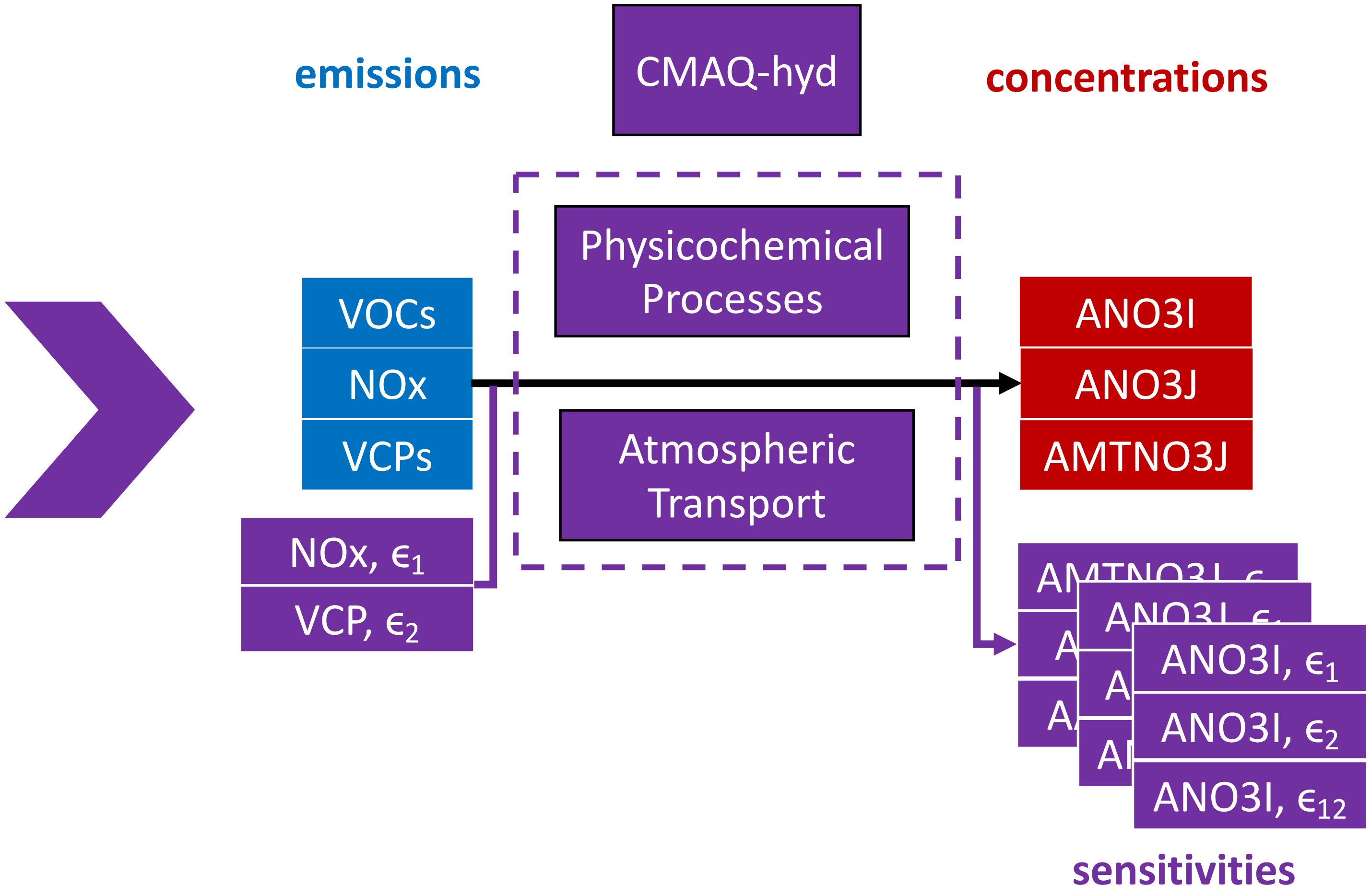

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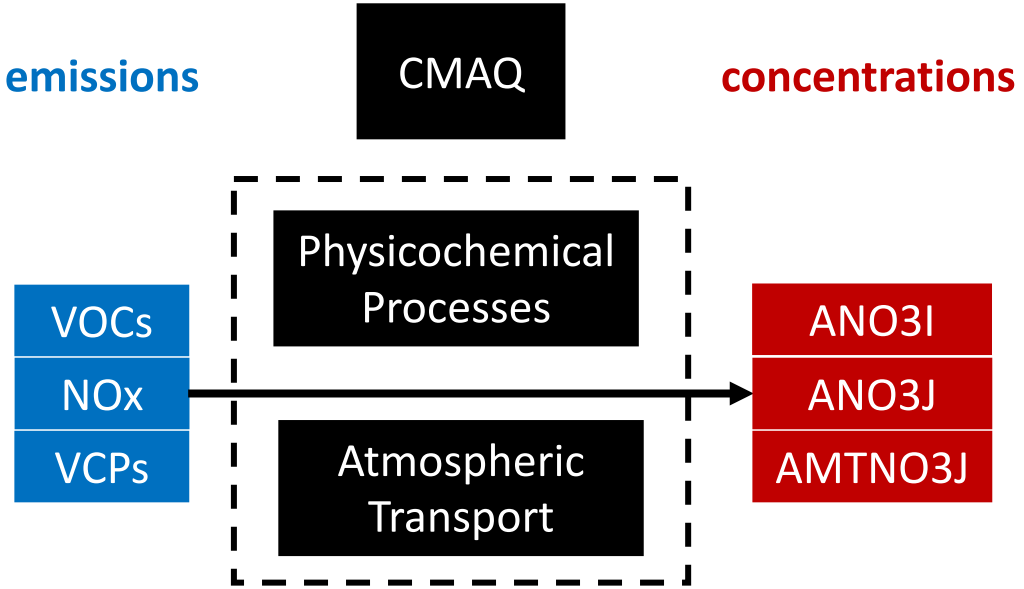
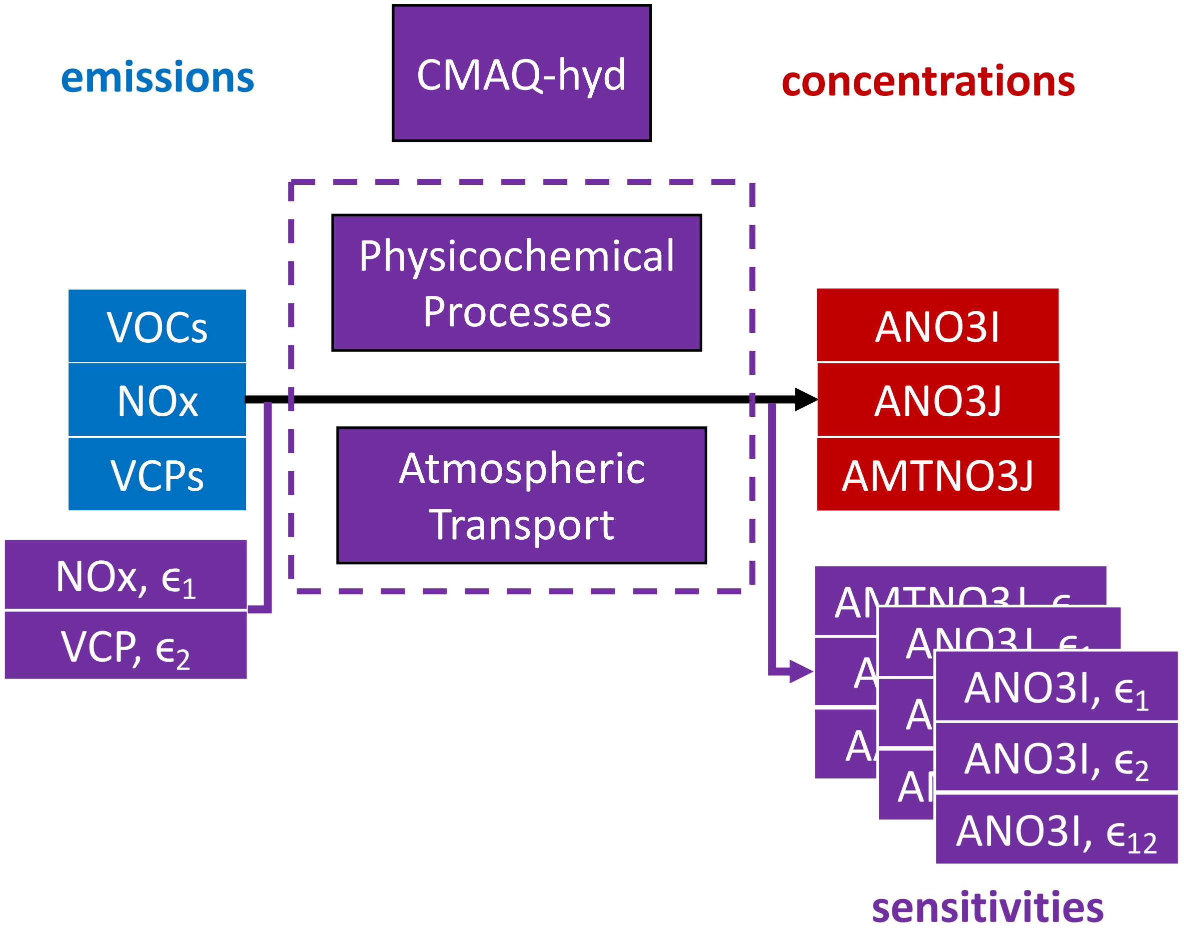


Source-code modifications



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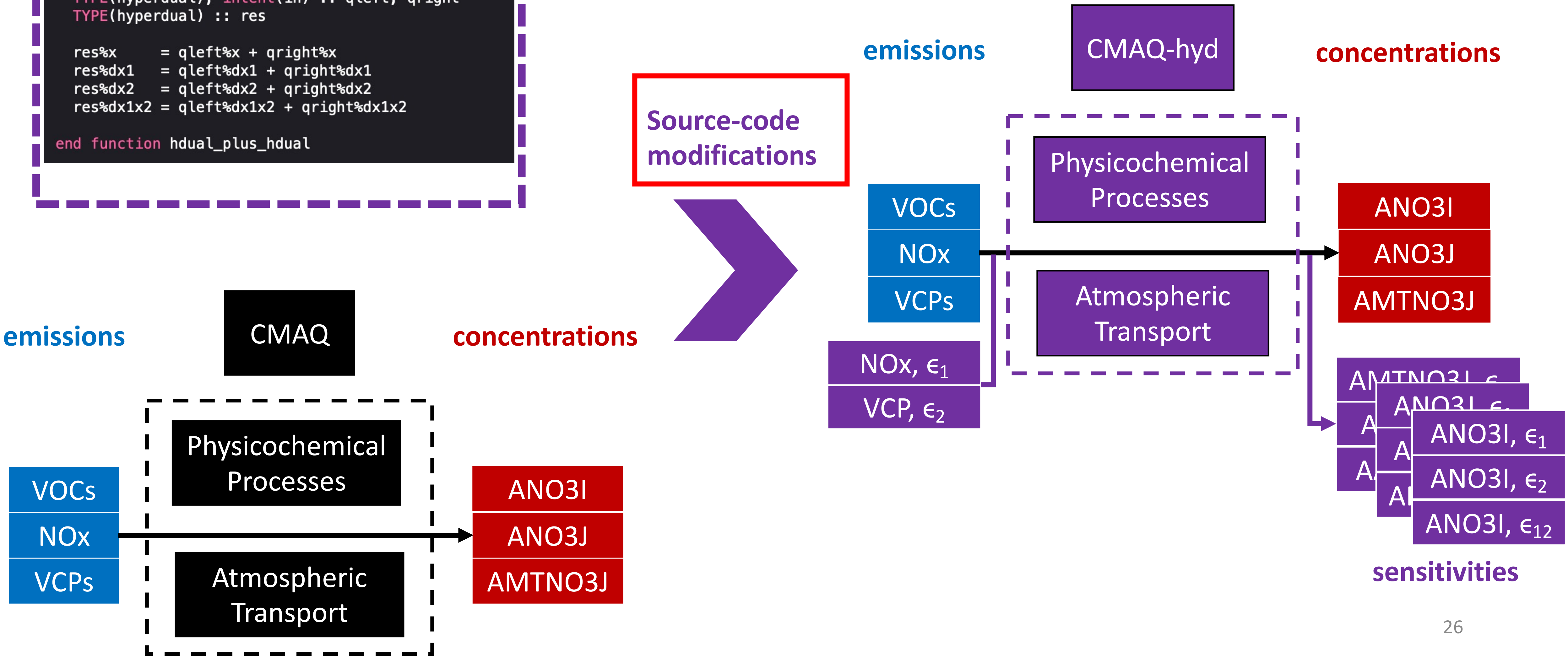


Source code modifications

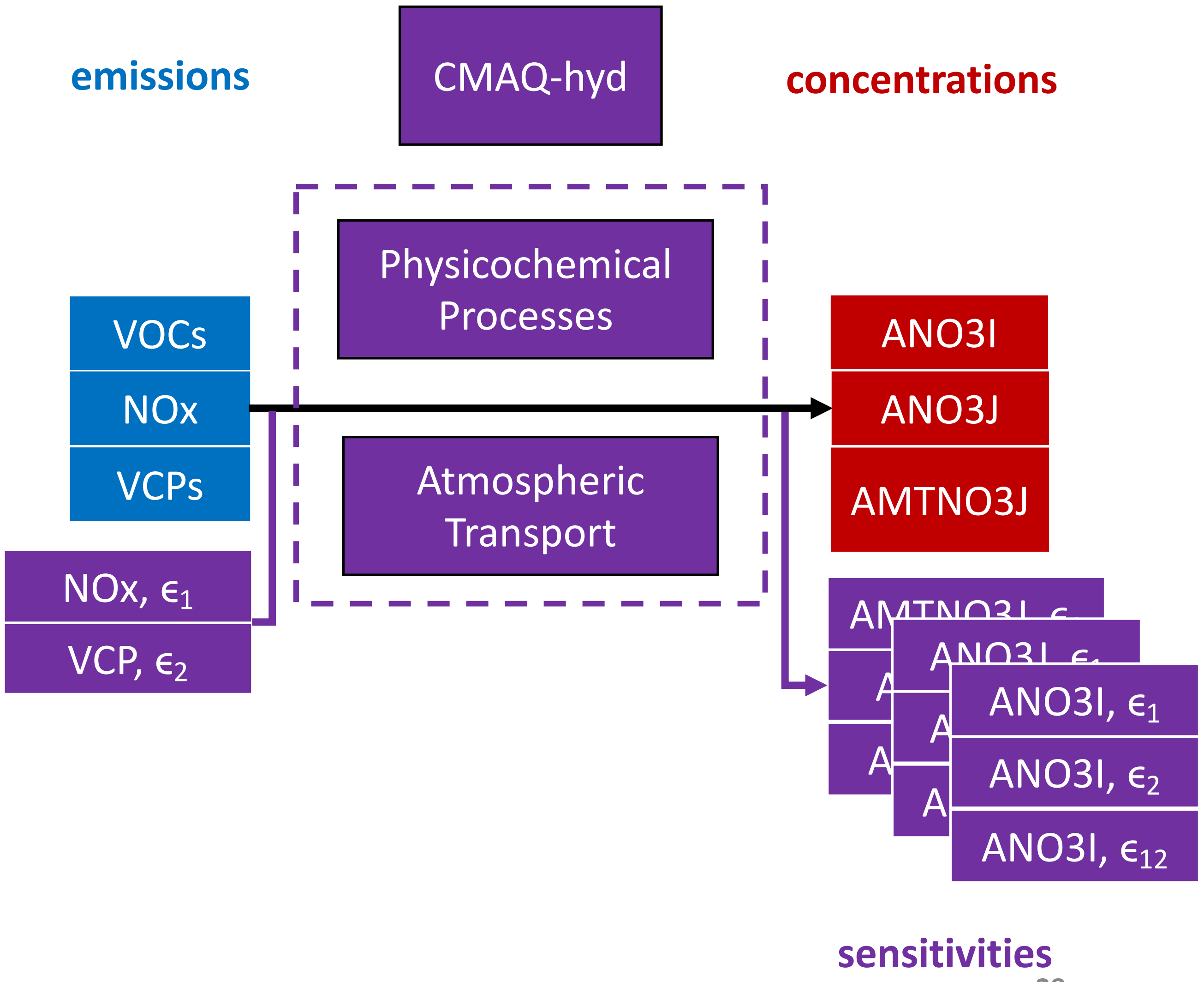
- Vertical diffusion in CMAQ:
- The only change to the source code is changing the real variables of interest to hyperdual.
- Not all variables in the model need to be changed to hyperdual to reduce the computational overhead of the model.
- A typical CMAQ-hyd run in regional scale is 2.5 times as expensive as the original CMAQ model.


```
SUBROUTINE VDIFF ( CGRID, JDATE, JTIME, TSTEP )
   USE HDMod
   TYPE(hyperdual), POINTER   :: CGRID( :,:,:,: )
   INTEGER, INTENT( IN )      :: JDATE, JTIME
   INTEGER, INTENT( IN )      :: TSTEP( 3 )
```

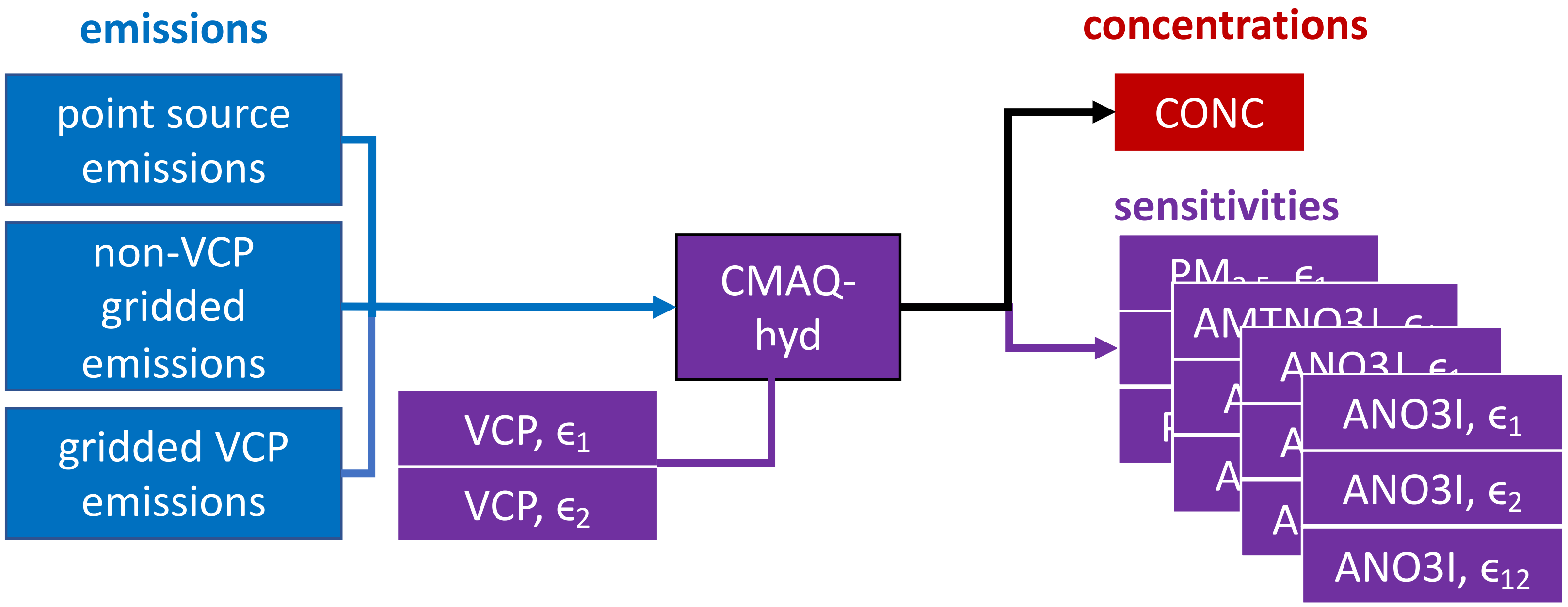
example of hyperdual CMAQ as described in Liu et al., GMDD, 2023

Sensitivity analysis with CMAQ-hyd

- CMAQ-hyd v.5.3.2 has been modified to propagate sensitivities from emissions to concentrations through hyperdual numbers (Liu et al., GMDD, 2023).
- The real concentrations are unchanged, and we get additional outputs which contain both first- and second-order sensitivity information.



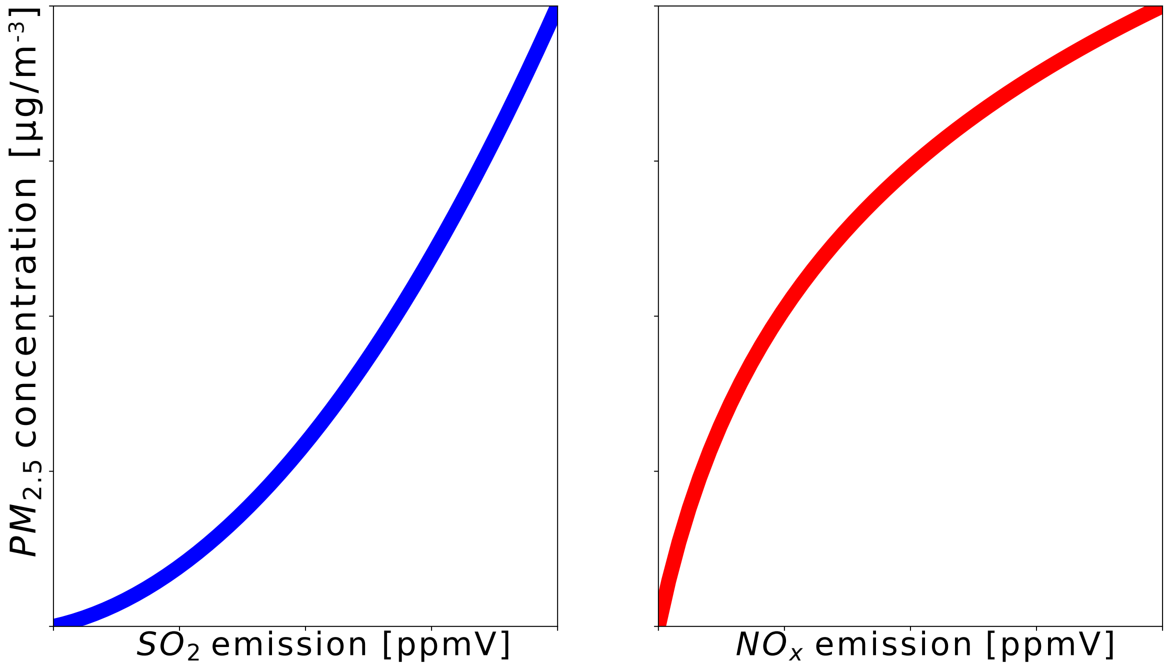
Modeling VCP contributions with CMAQ-hyd



- Using CMAQ-hyd, the VCP emissions are perturbed in hyperdual space to obtain first- and second-order sensitivities of all modeled concentrations with respect to total VCP emissions.

Physical meaning of first- and second-order sensitivities in models

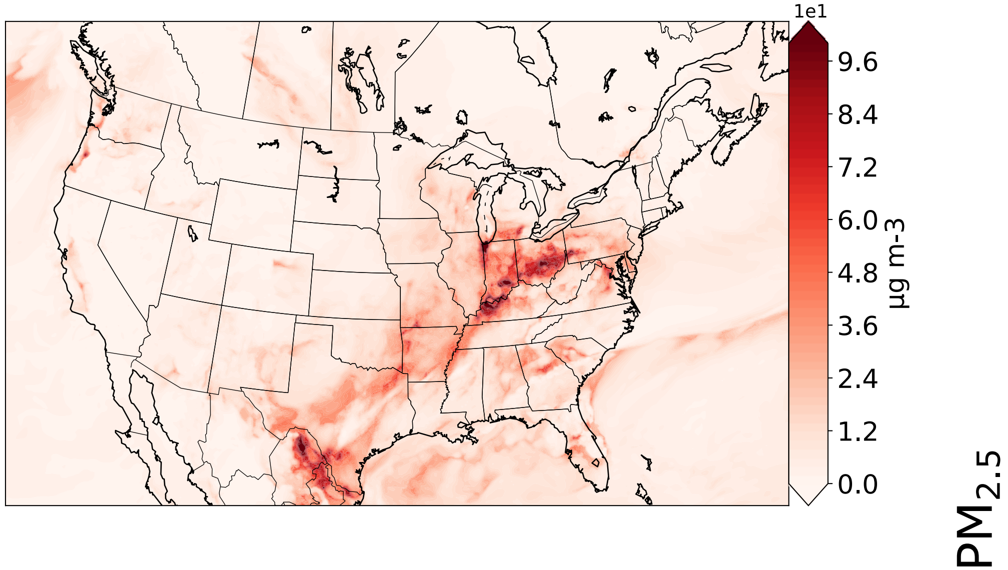
- The first-order sensitivity describes the slope of the relationship.
- The second-order sensitivity describes the curvature of the relationship.
- Since many processes in chemical transport models like CMAQ are highly nonlinear, understanding the second-order sensitivities is important.

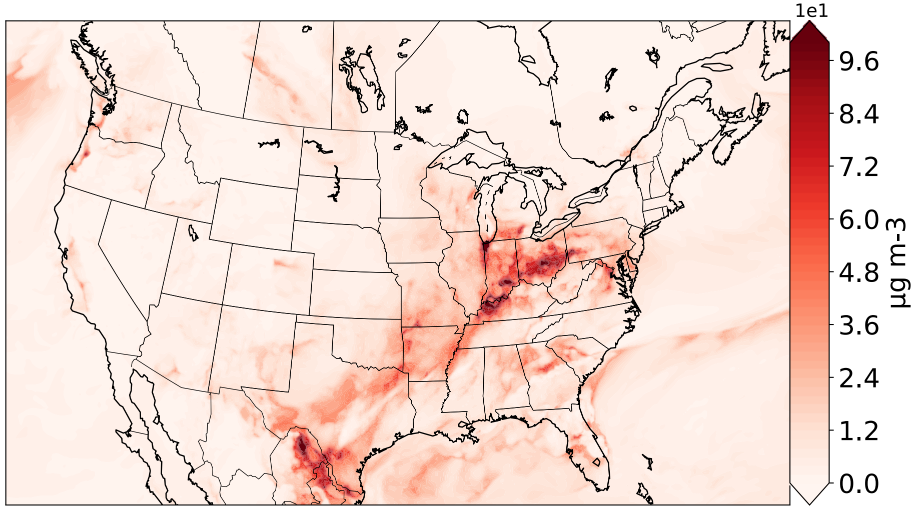


Proof-of-concept CMAQ-hyd application with VCP emissions

- Model configuration
  - 7:00 PM EST on Jan. 1st, 2019 to 7:00 AM EST on Jan. 2nd, 2019
  - Domain: 12US1 grid
  - Chemical mechanism: cb6r3_ae7_aq
  - Aerosol module: aero7

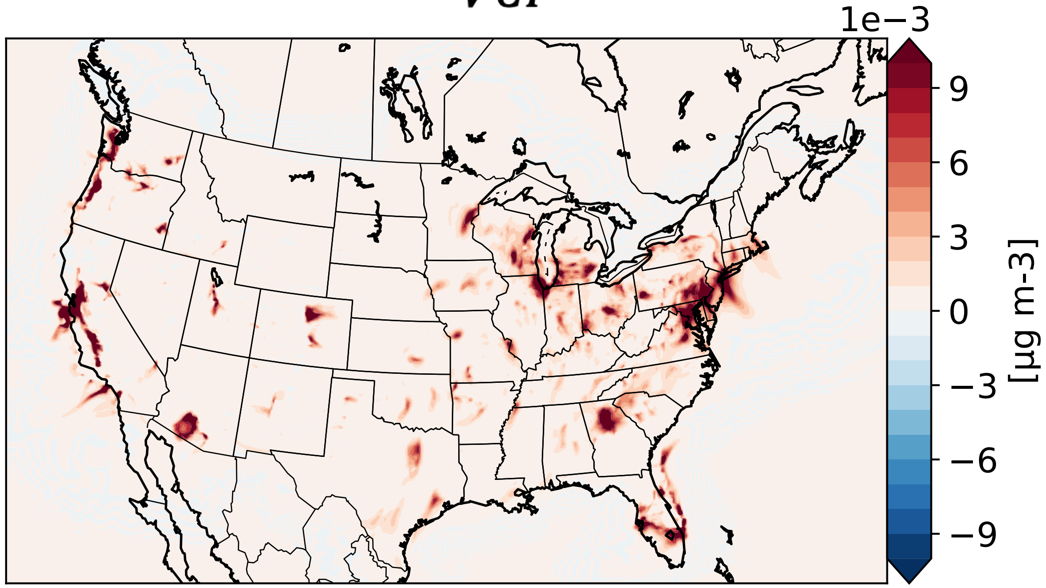
- The concentrations of $PM_{2.5}$ are nearly identical between the publicly released CMAQ version and our hyperdual version.



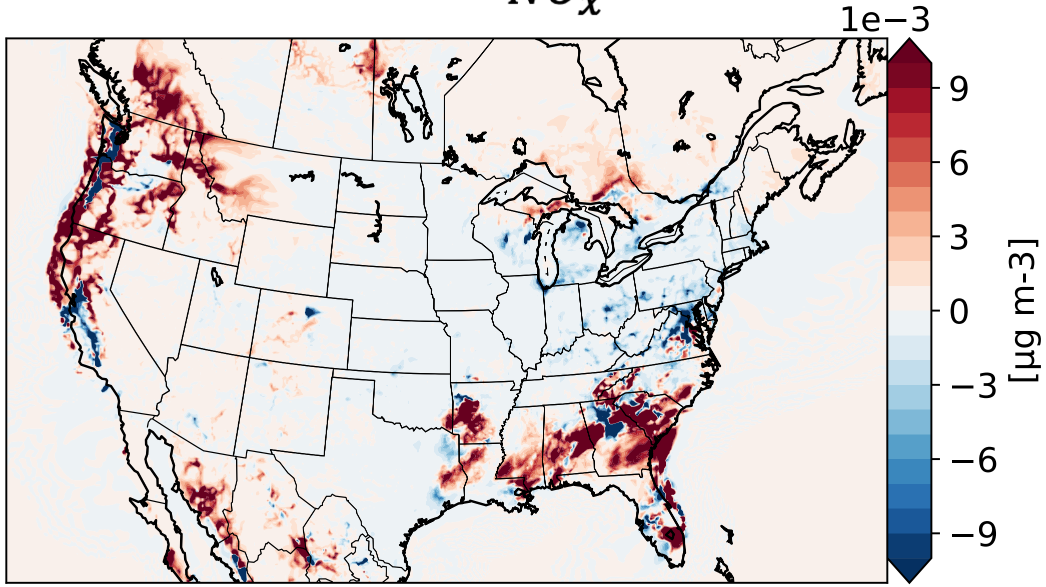


Probing contributions of VCPs and NOx to aerosol

$$\frac{\partial C_{AMTNO_3J}}{\partial \sigma_{VCP}}$$



$$\frac{\partial C_{AMTNO_3J}}{\partial \sigma_{NO_x}}$$



- The nighttime formation of accumulation-mode monoterpene nitrate aerosols (AMTNO3J) depends on both monoterpenes from VCP and NO_x emissions.

Probing contributions of VCPs and NOx to aerosol

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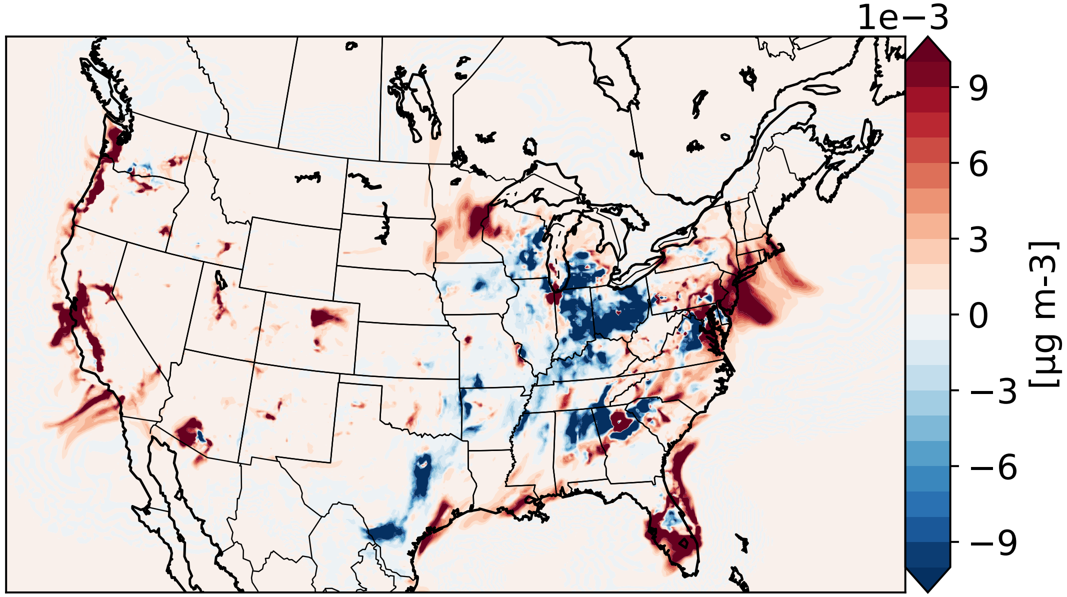
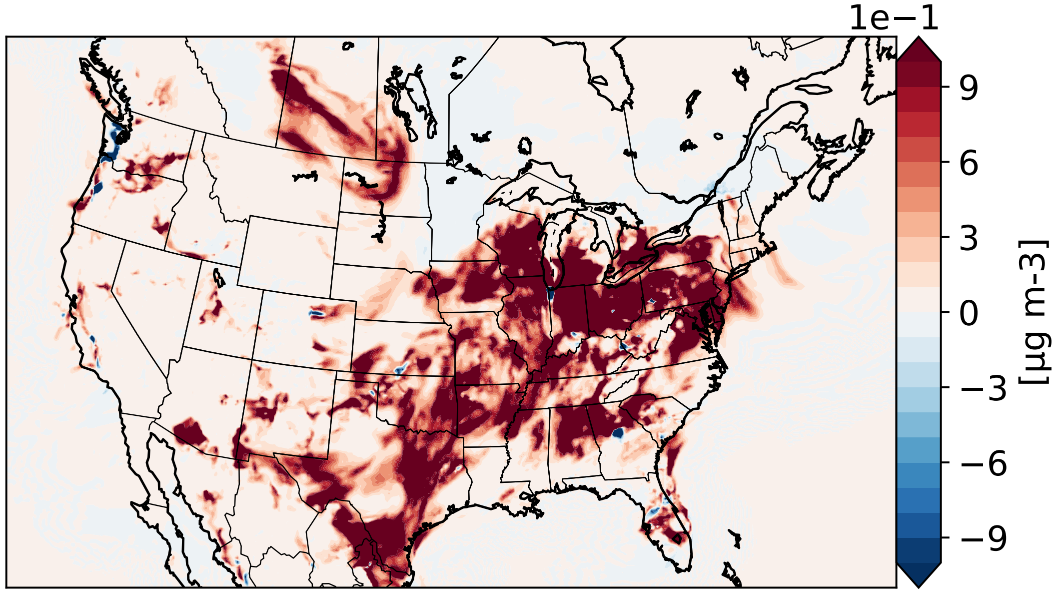


$$\frac{\partial C_{PM_{2.5}}}{\partial \sigma_{NO_x}}$$



- In parts of the central US, VCPs react with nitrate to form AMTNO3J, leading to less ammonium nitrate formation, which leads to a slightly negative sensitivity of $PM_{2.5}$ concentration to VCP emissions.

Probing the nonlinearity of contributions of VCPs and NOx to aerosols

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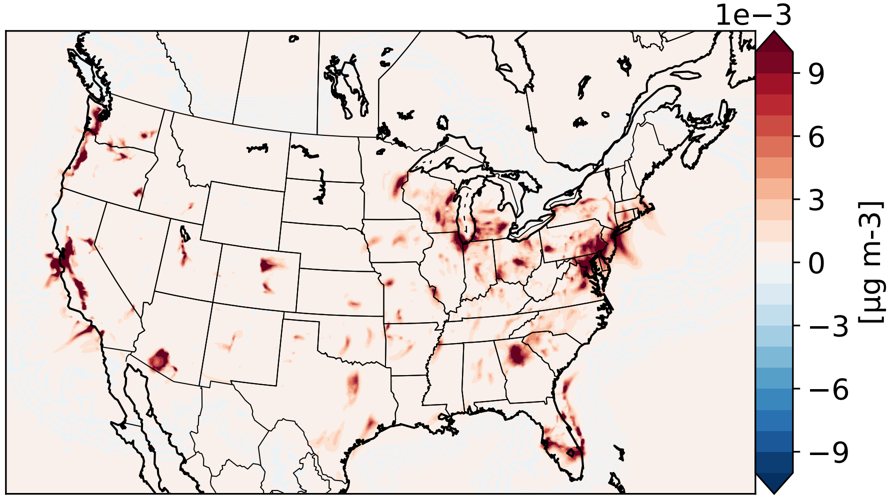
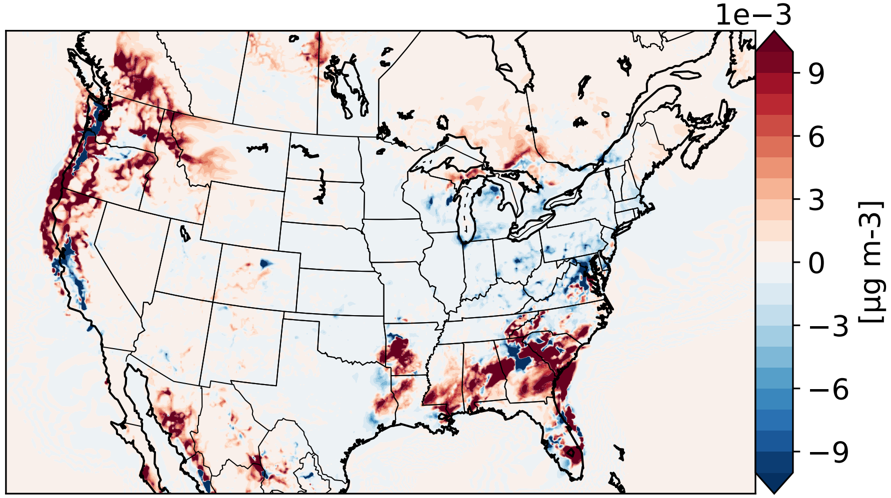
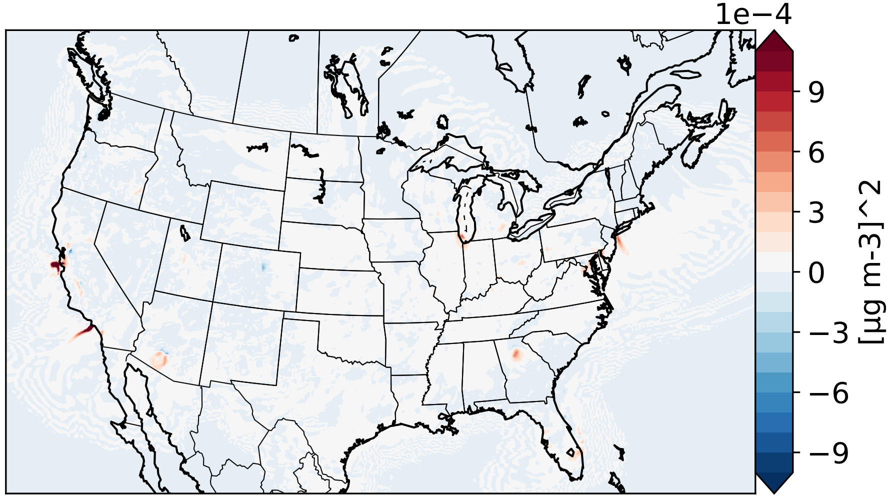


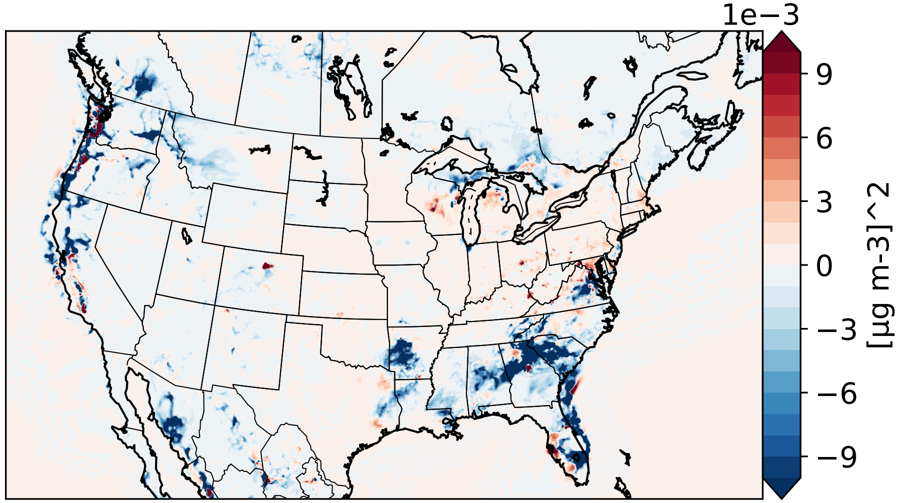
$$\frac{\partial C_{AMTNO_3J}}{\partial \sigma_{NO_x}}$$



$$\frac{\partial^2 C_{AMTNO_3J}}{\partial \sigma_{VCP}^2}$$



$$\frac{\partial^2 C_{AMTNO_3J}}{\partial \sigma_{NO_x}^2}$$



Probing the nonlinearity of contributions of VCPs and NOx to aerosols

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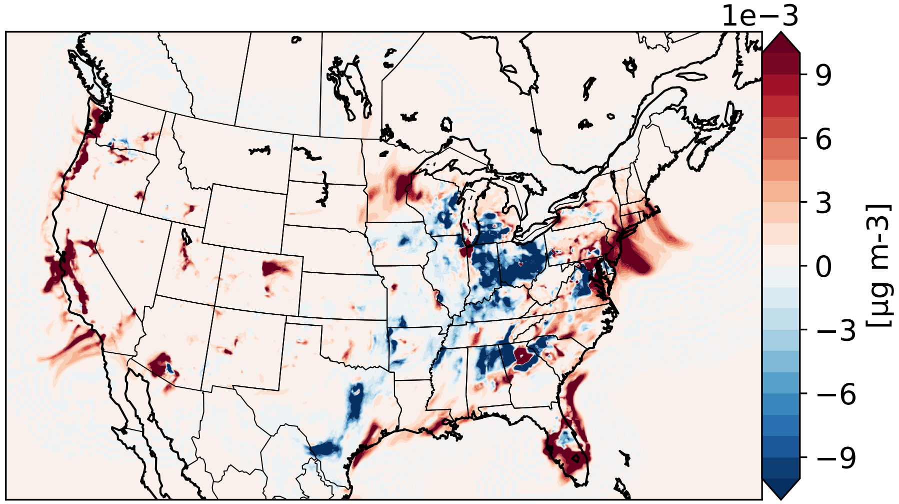
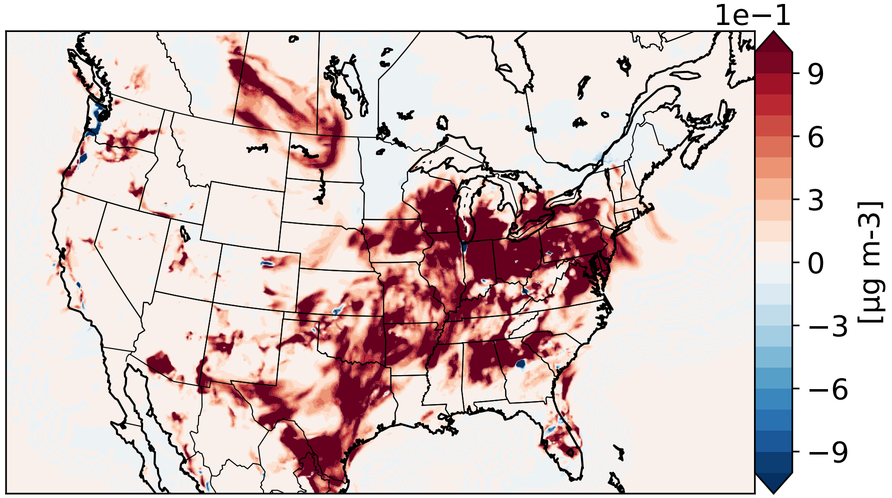
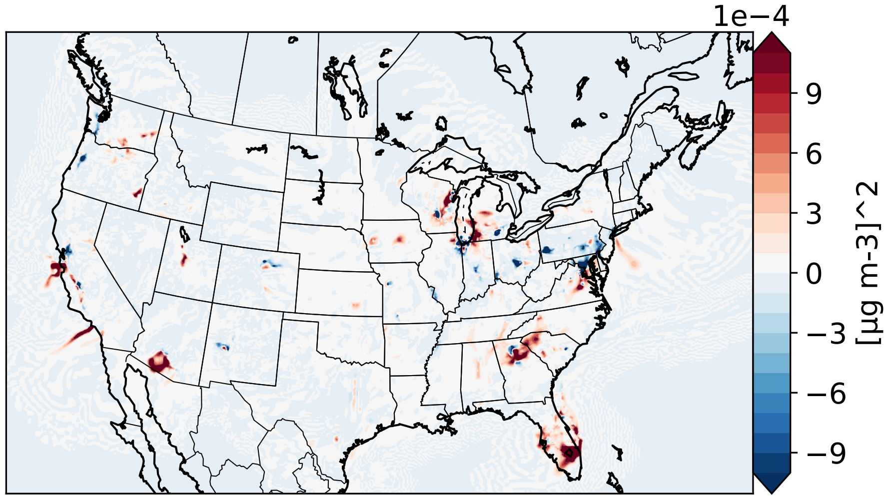


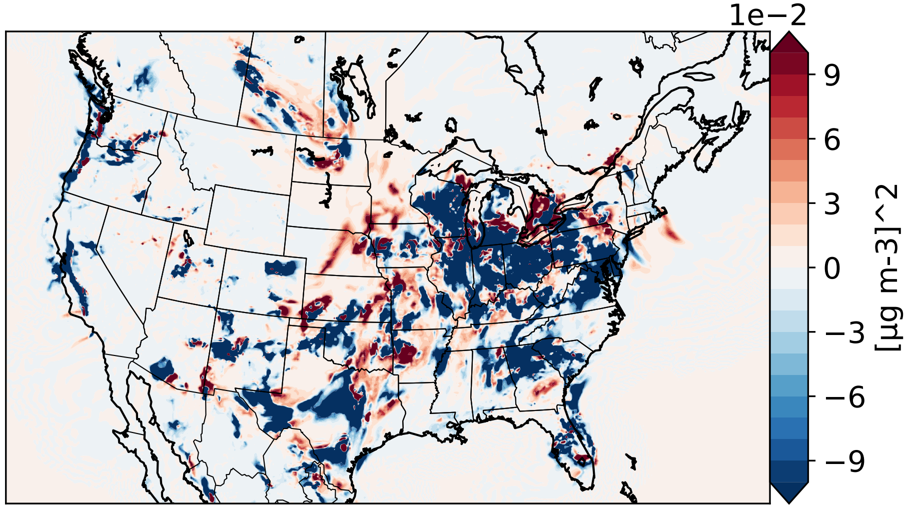
$$\frac{\partial C_{PM_{2.5}}}{\partial \sigma_{NO_x}}$$



$$\frac{\partial^2 C_{PM_{2.5}}}{\partial \sigma_{VCP}^2}$$



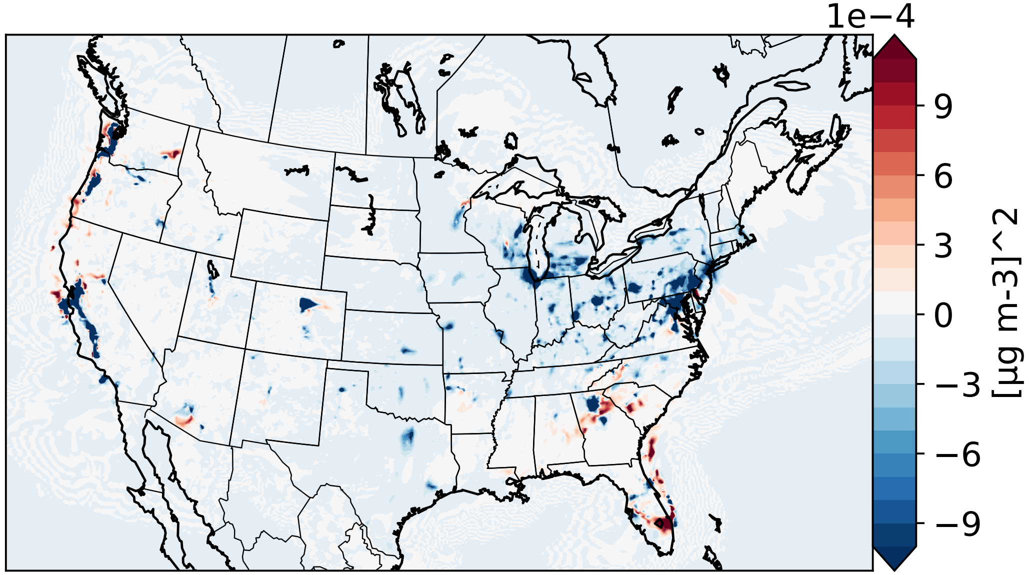
$$\frac{\partial^2 C_{PM_{2.5}}}{\partial \sigma_{NO_x}^2}$$

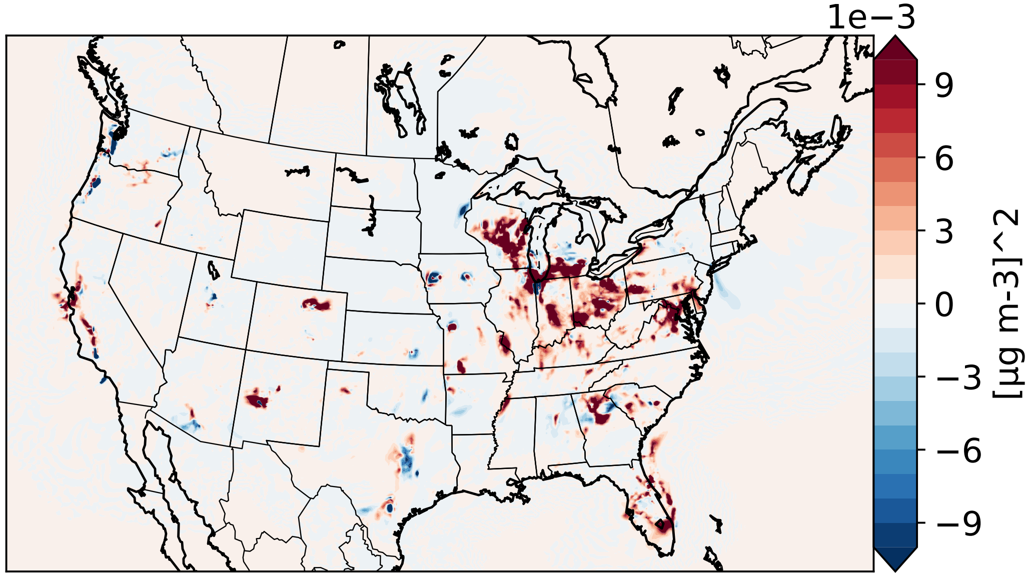


Probing the nonlinearity of contributions of VCPs and NOx to aerosols

$$\frac{\partial^2 C_{AMTNO_3J}}{\partial \sigma_{NO_x} \partial \sigma_{VCP}}$$

$$\frac{\partial^2 C_{PM_{2.5}}}{\partial \sigma_{NO_x} \partial \sigma_{VCP}}$$





Conclusions & Future Directions

- We have developed an augmented version of CMAQ which can compute first- and second-order sensitivities to machine precision.
- We plan to run monthly simulations across different seasons.
- We also plan to evaluate the health-related impact of VCP emissions based on the calculated sensitivities.
- In theory, the hyperdual-step method can be easily extended to other numerical models where sensitivities are of interest.

HDMod.f90

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- Emissions data: Dr. Kristen Foley provided the requisite pre-merged files for the VCP emissions analysis for EQUATES.
- Debugging: CMAS Forum

HDMod.f90

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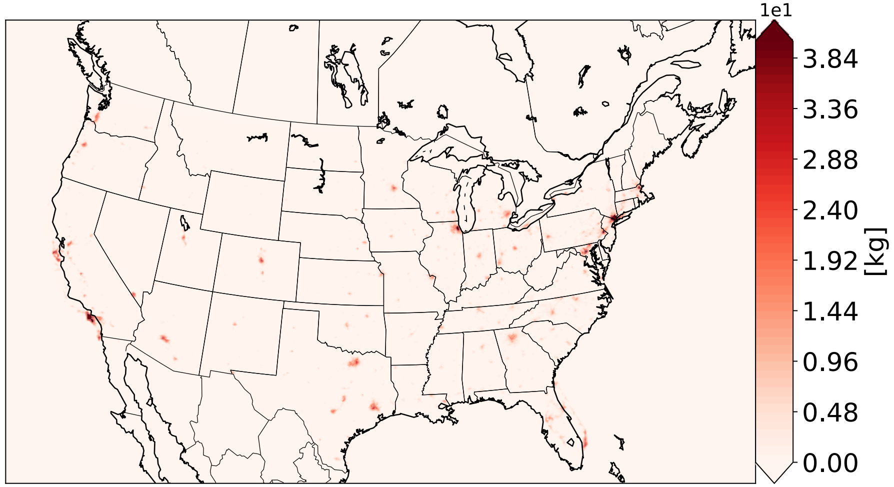
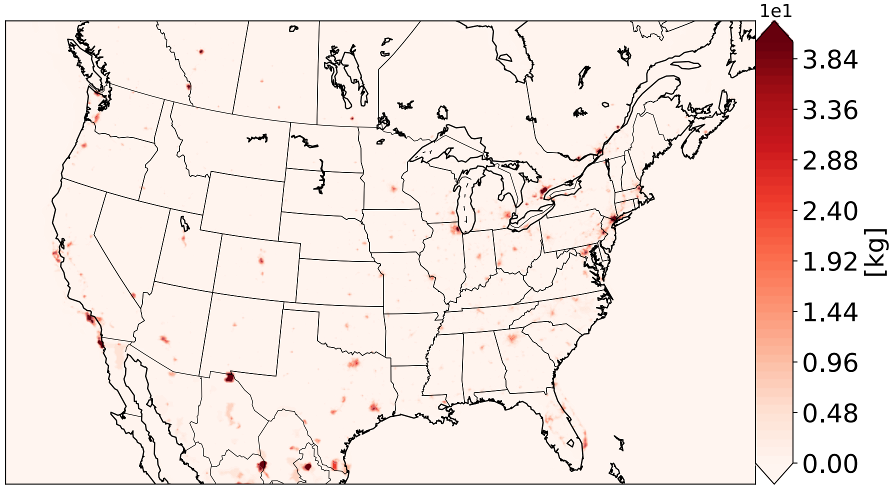
Verifying VCP emissions separation

Total daily gridded CONUS monoterpene emissions in kilograms on Jan 2[nd], 2019

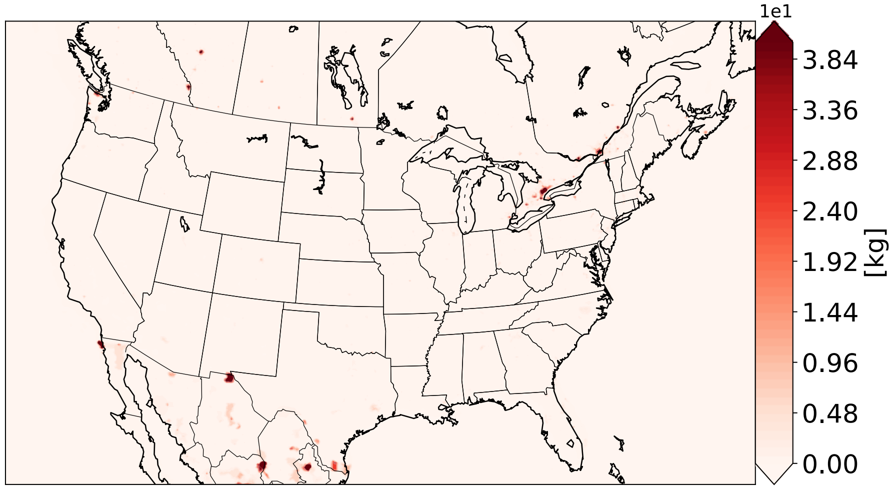
VCP gridded emissions (np_solvents)



EQUATES gridded emissions

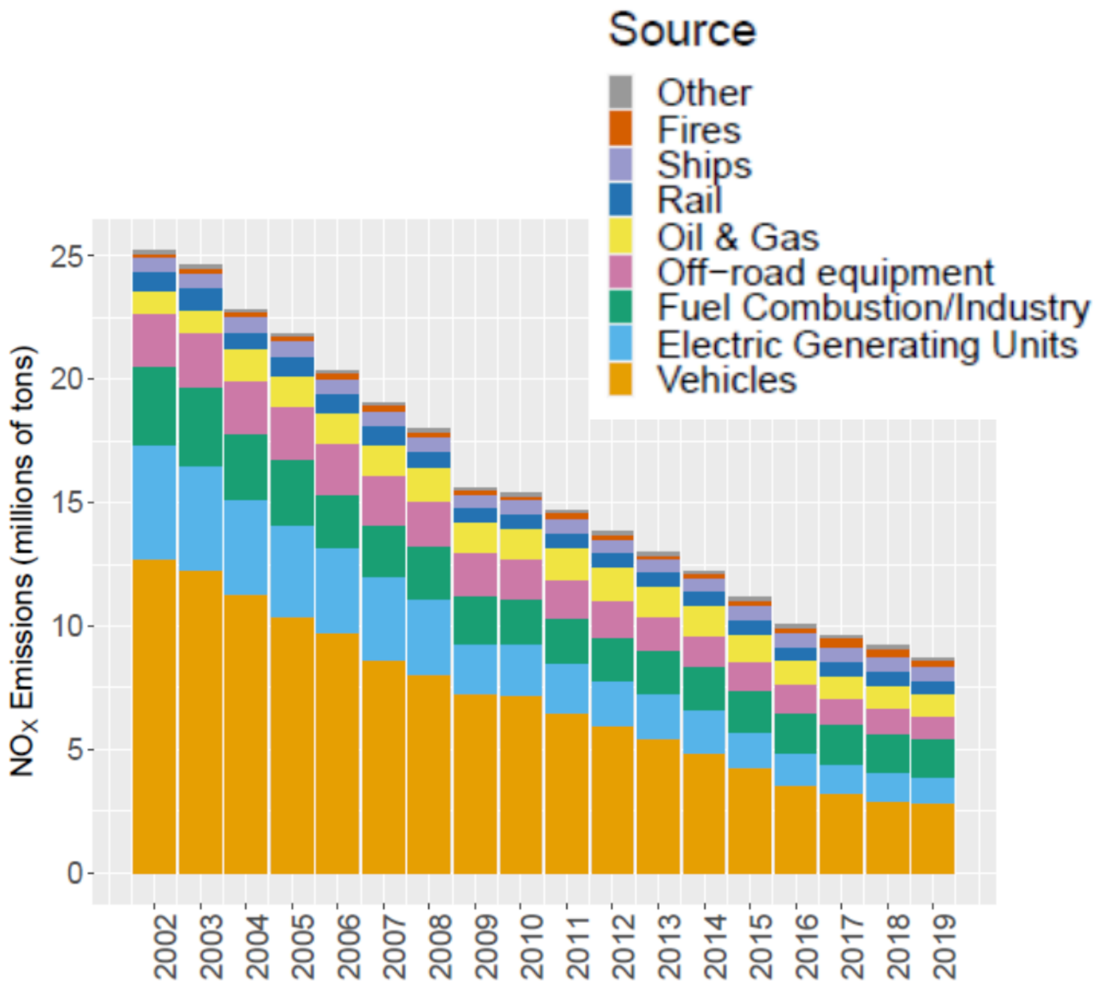


non-VCP gridded emissions



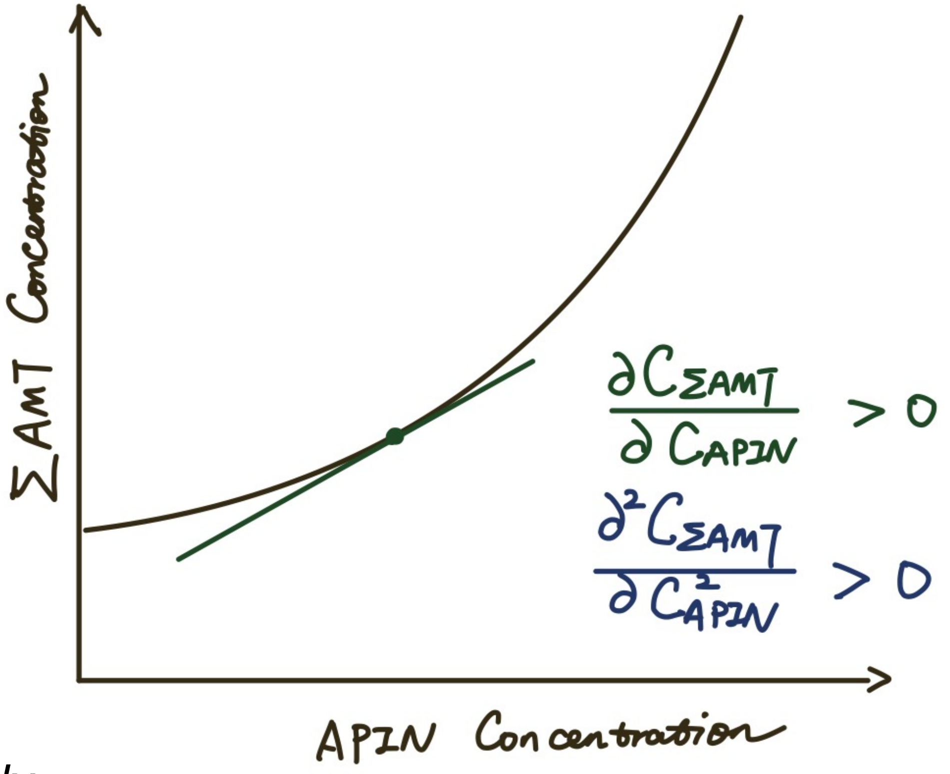
EQUATES modeling platform

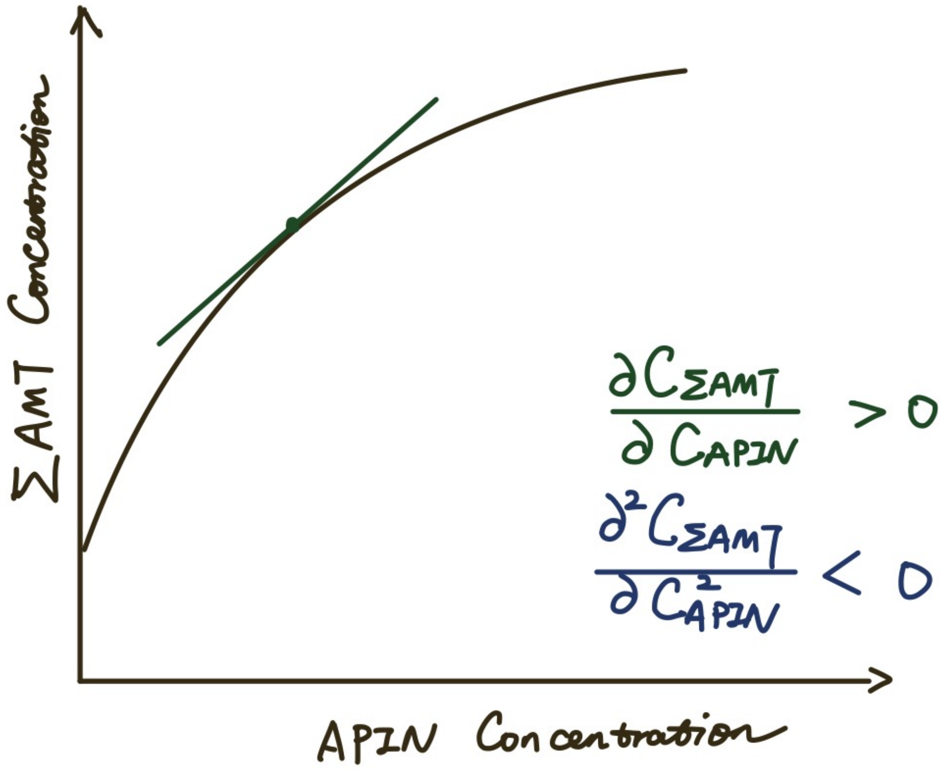
- EQUATES provides a unified set of modeled meteorology, emissions, air quality and pollutant deposition from year 2002 to 2019 (Foley et al., 2023).
- The EQUATES emissions were developed using consistent input data and methods.
- Importantly for this work, EQUATES includes VCP emissions following Seltzer et al. (2021).



annual NO_x emissions from 2002 to 2019
from Foley et al., 2023

The Physical Meaning of Second-Order Derivatives





If we only consider the first-order sensitivity of a reduction of demominator:

Underestimate the effect

Overestimate the effect

12US1 domain over CONUS: 299
rows x 459 columns x 35 layers