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Plan de Recuperación, Transformación y Resiliencia

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Modeling of secondary organic aerosol formation: how much do the choices of chemical mechanism and parameterization really matter?

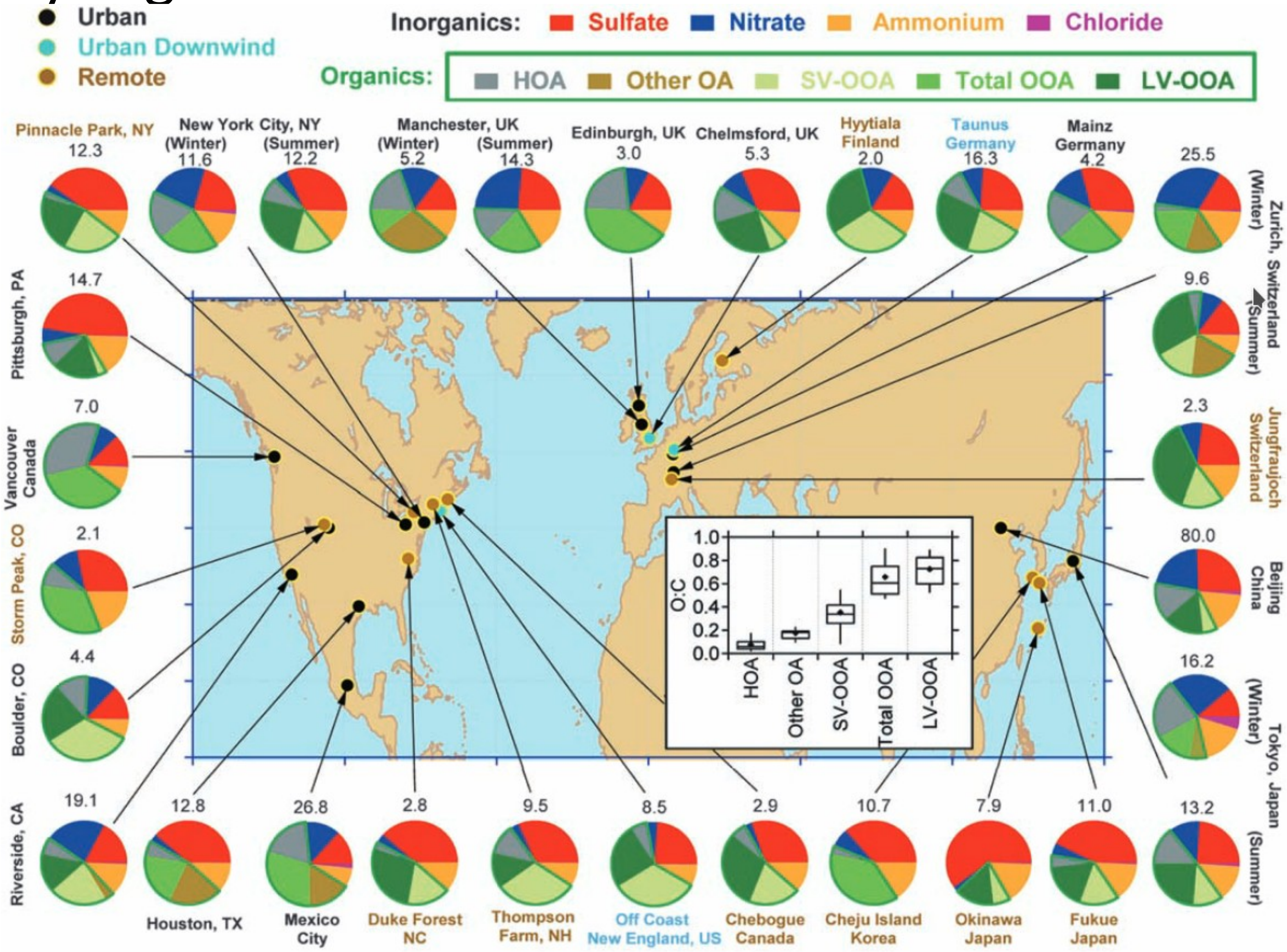
ITM, Copenhagen, 17/10/2024

Camille Mouchel-Vallon, Hervé Petetin, Alessio Melli, and Oriol Jorba

Air quality: secondary organic aerosol

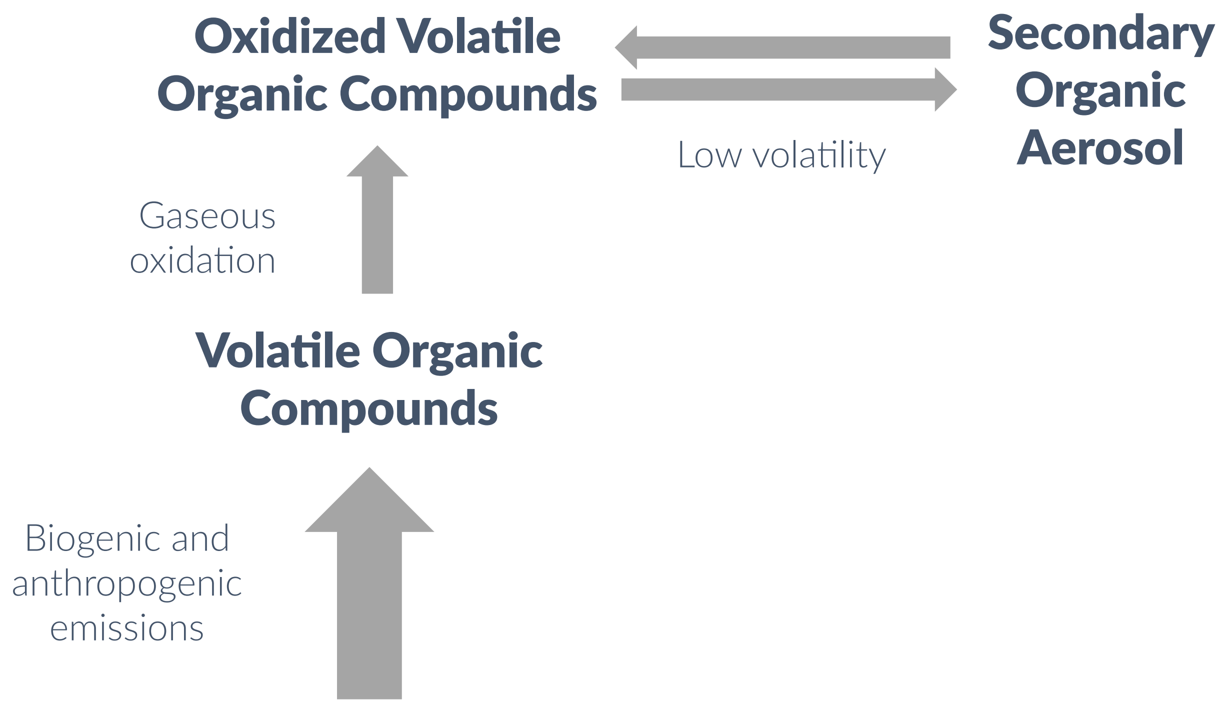
- Significant contribution of organics to particulate matter

- Crucial importance of representing formation of secondary organic aerosol (SOA)



Jimenez et al. (2009)

Atmospheric Evolution of Organics : in models

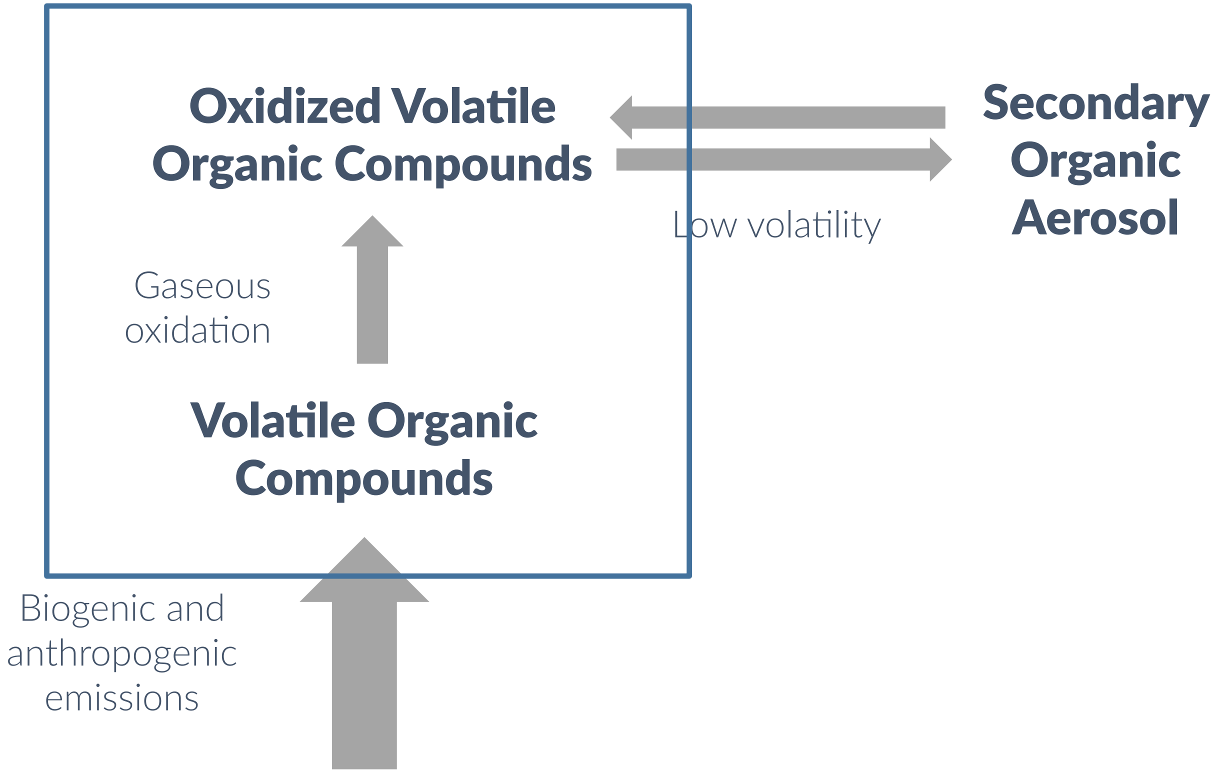


Atmospheric Evolution of Organics : in models

Large choice of gas phase mechanisms

Carbon bond,
RACM,
SAPRC, ...

Focused on the impact of VOCs on ozone



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Atmospheric Evolution of Organics : in models

Expected representation of SOA formation

Condensation of low volatility organics

Only done with sufficiently detailed chemical mechanisms (MCM, GECKO-A, ...)

Large choice of gas phase mechanisms

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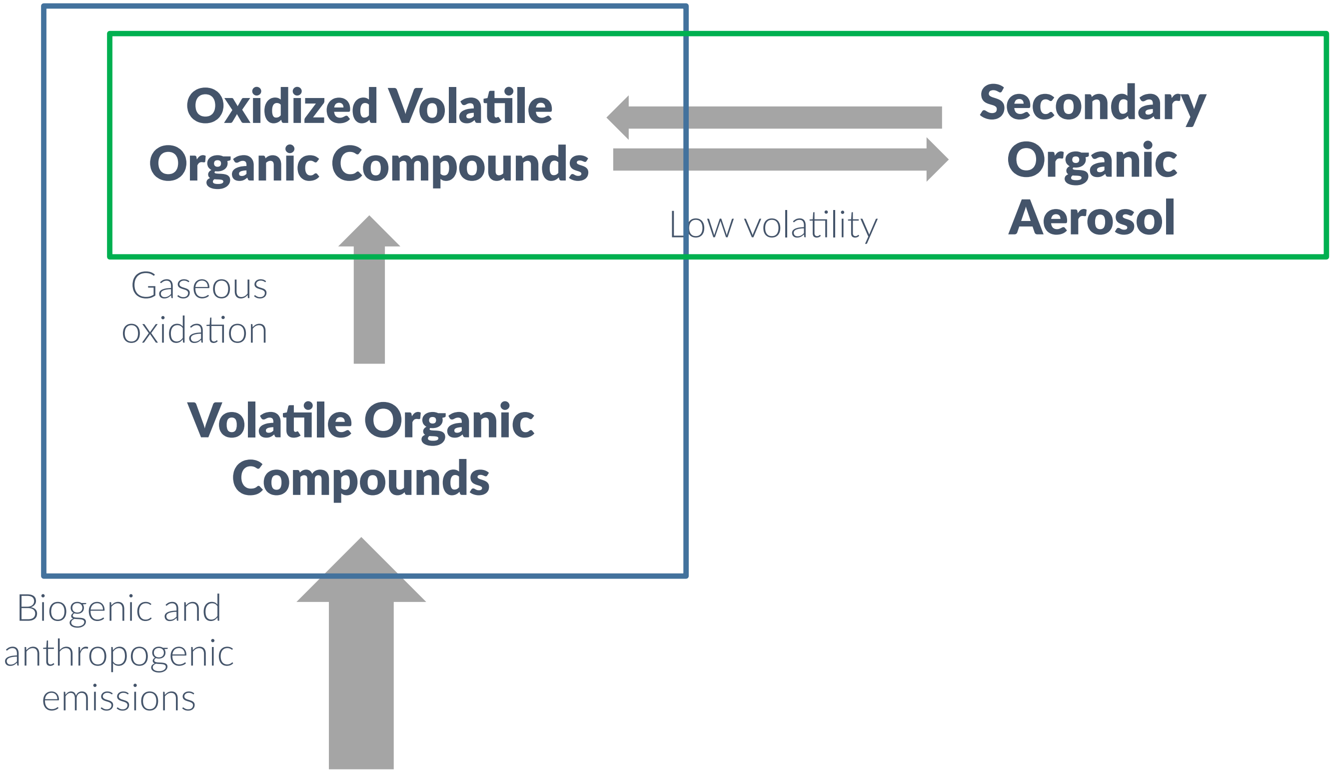

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Atmospheric Evolution of Organics : in models

Large choice of gas phase mechanisms

Carbon bond,
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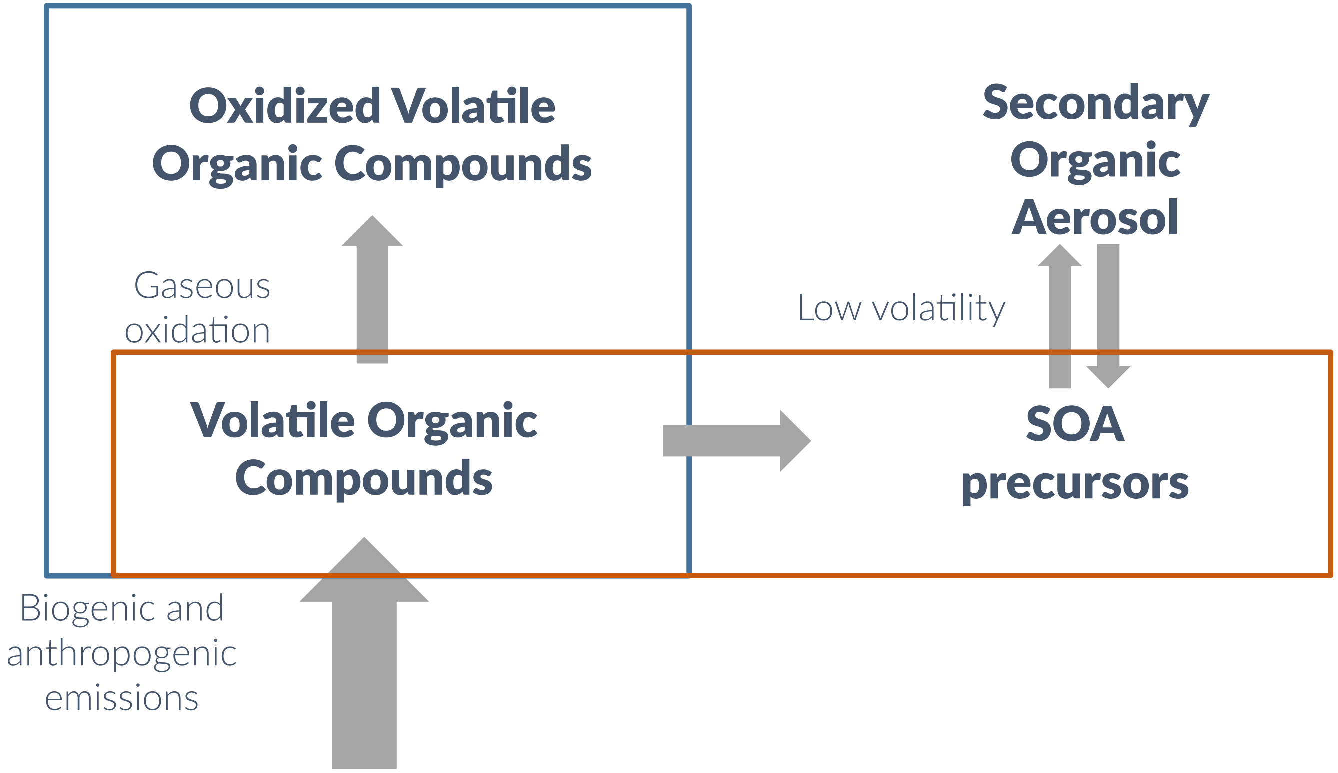
Focused on the impact of VOCs on ozone

SOA formation in practice

A parametrization running independently of the gas phase mechanism

2 product models, VBS, 2D VBS, SOM, ...



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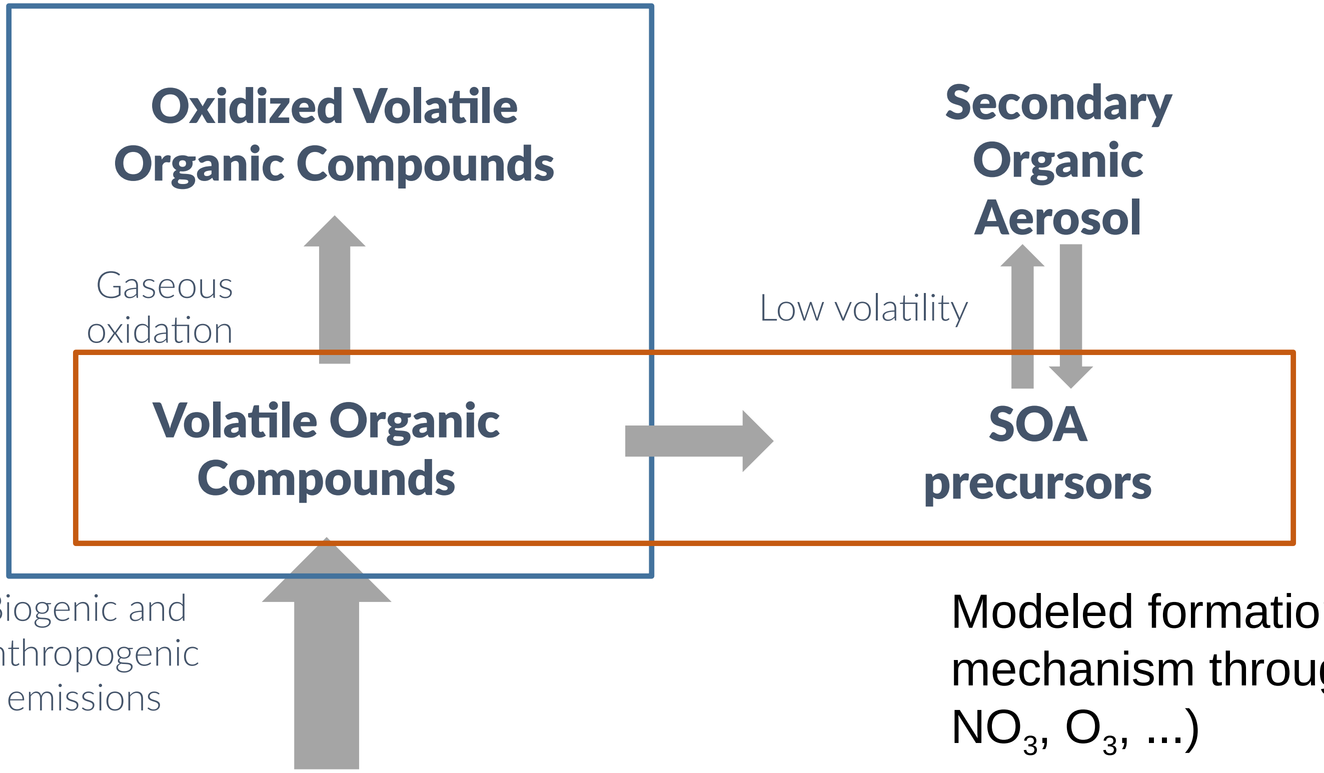
Atmospheric Evolution of Organics : in models



Modeled formation of SOA is only **indirectly** affected by the gas phase mechanism through the evolution of VOCs and their oxidants (OH, NO_3, O_3, ...)

More complex SOA parametrization also depend on the modeled **chemical regime** (NOx vs HO_2) for particle phase aging

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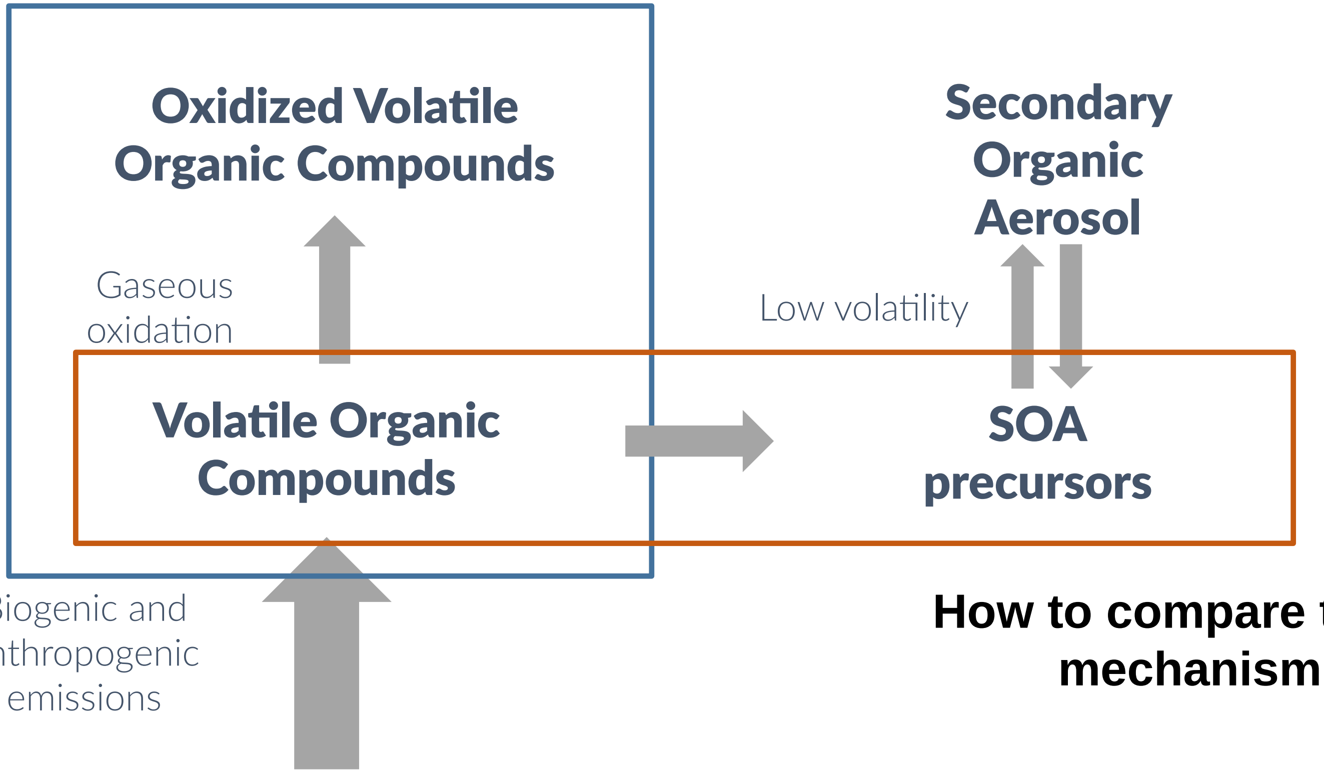
Atmospheric Evolution of Organics : chemical mechanisms comparisons



How to compare the impact of different combinations of chemical mechanism + SOA parametrization on SOA formation?

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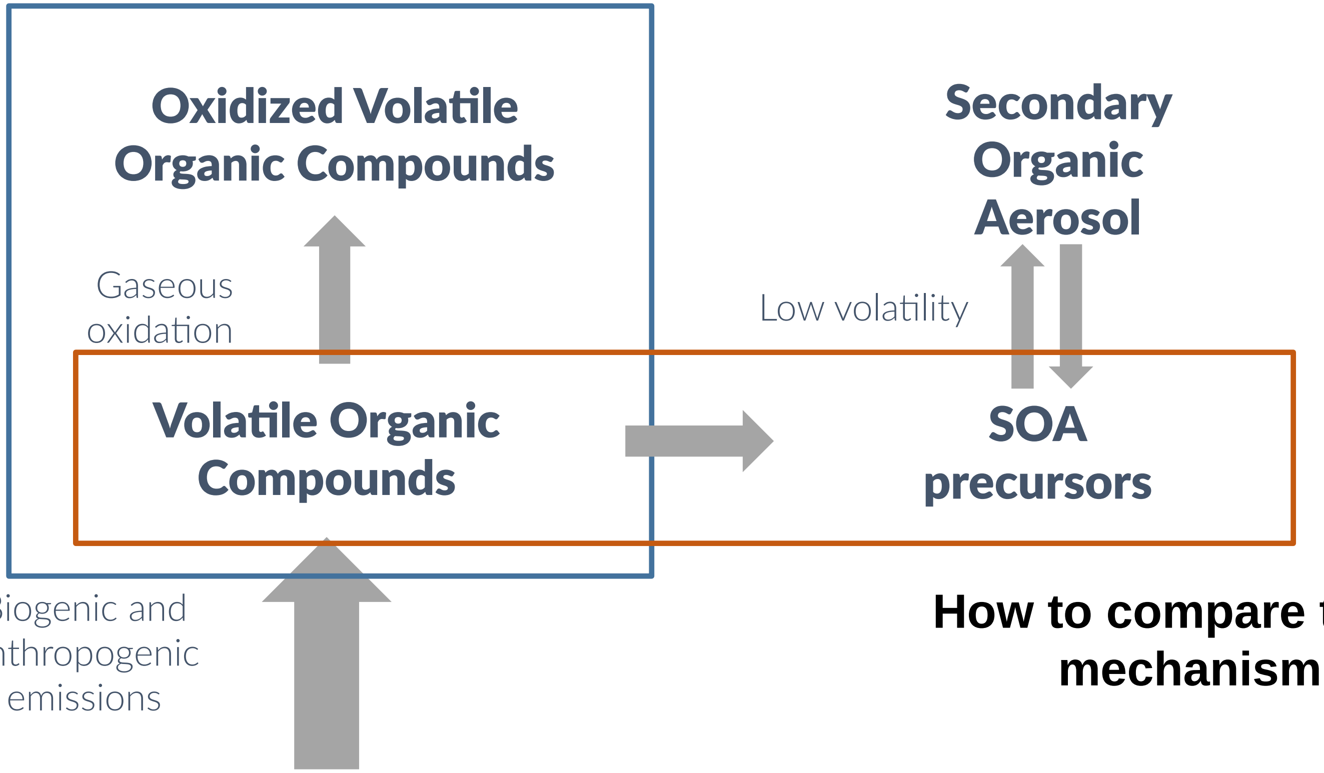
Atmospheric Evolution of Organics : chemical mechanisms comparisons



How to compare the impact of different combinations of chemical mechanism + SOA parametrization on SOA formation?

Evaluating the sensitivity of chemical mechanisms to different factors
(NOx, VOC, POA)

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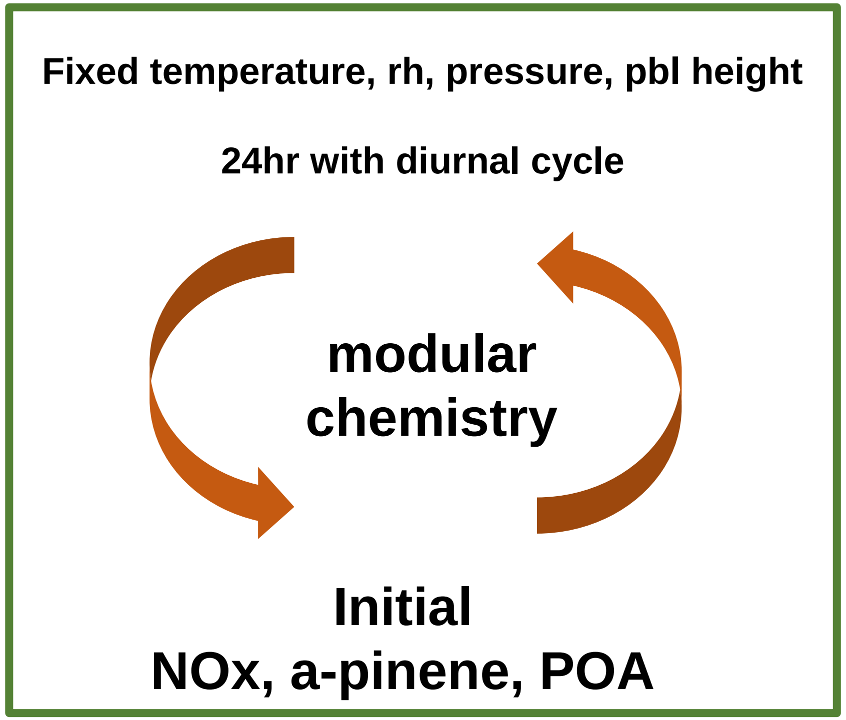
The CAMP boxmodel (Dawson et al., 2022)



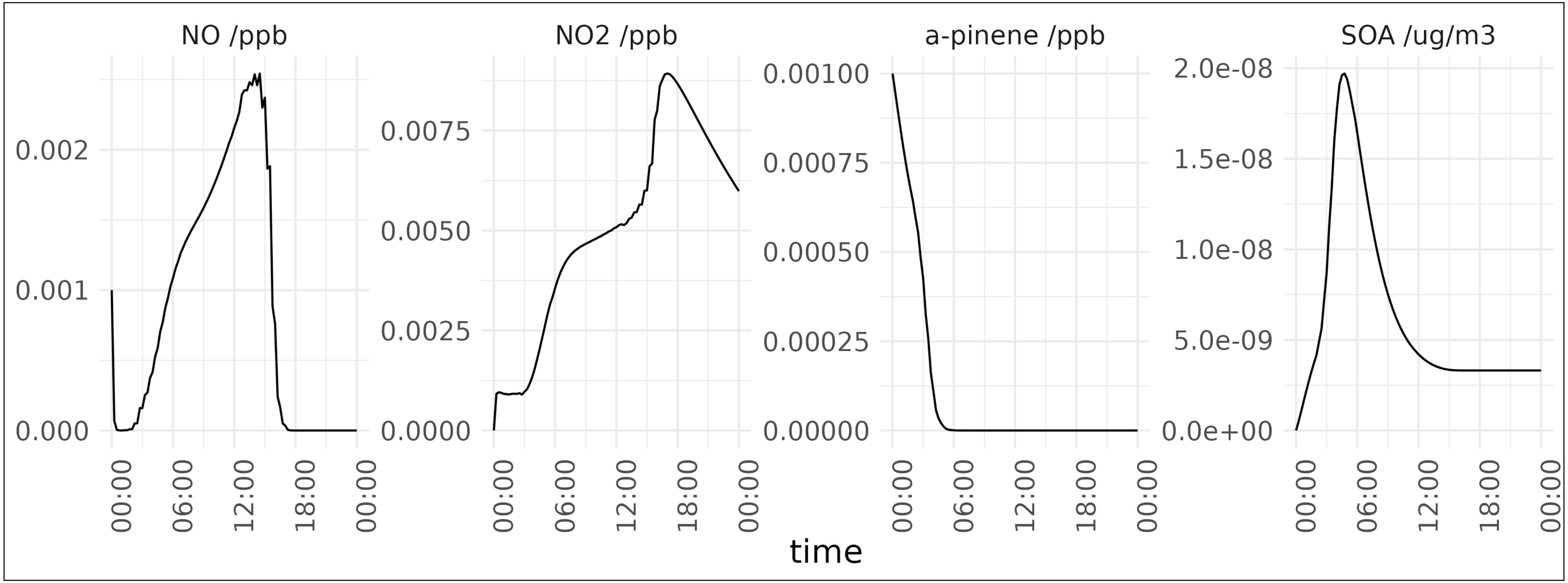
A boxmodel based on the CAMP (Chemistry Across Multiple Phases) atmospheric chemistry framework

Modular chemistry: easy to switch mechanisms, parametrizations

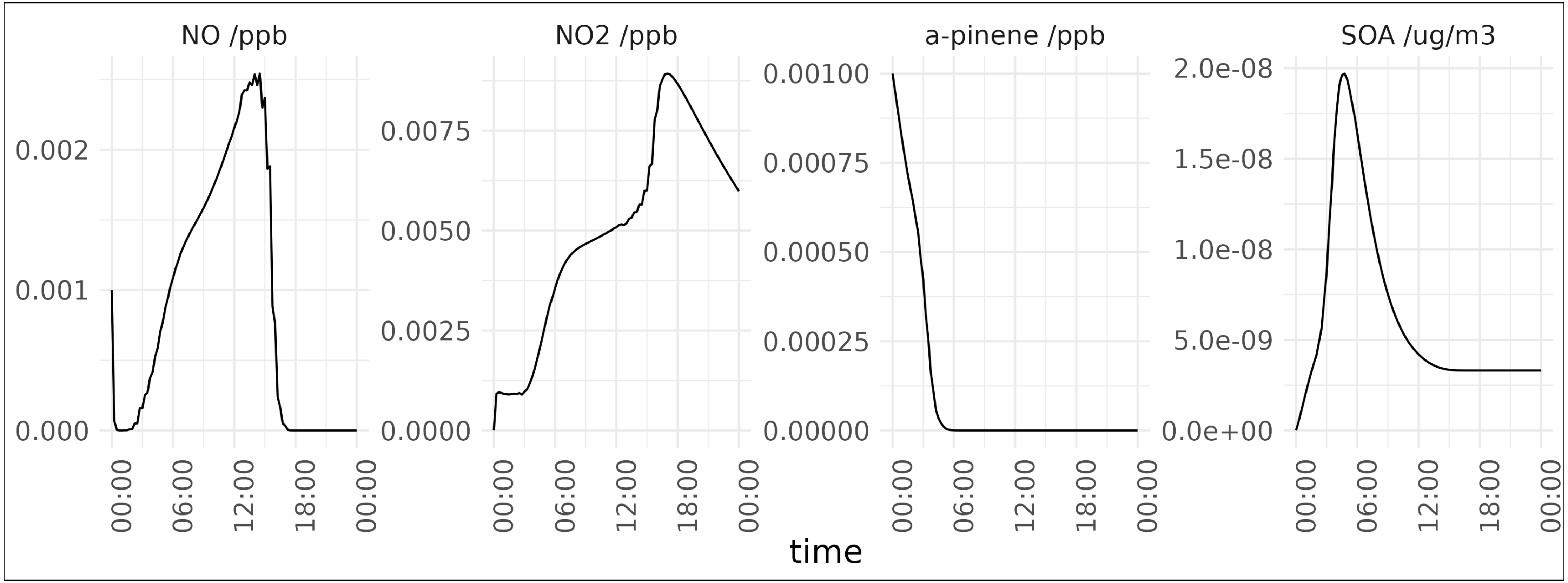
Parallel capabilities: run many boxes in parallel, with different initialization

Ideal tool to run sensitivity tests

SOA formation sensitivities: example of CB05 + 2 products



SOA formation sensitivities: example of CB05 + 2 products



Let’s play with initial NOx, a-pinene and POA

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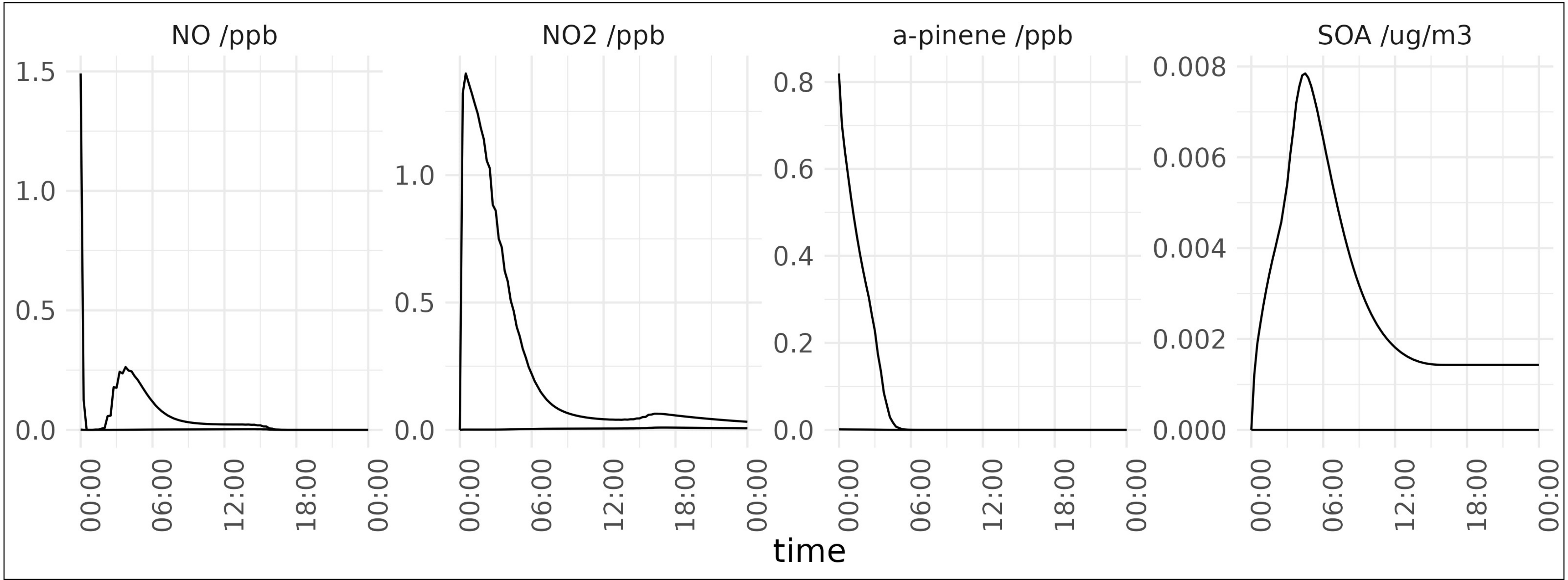

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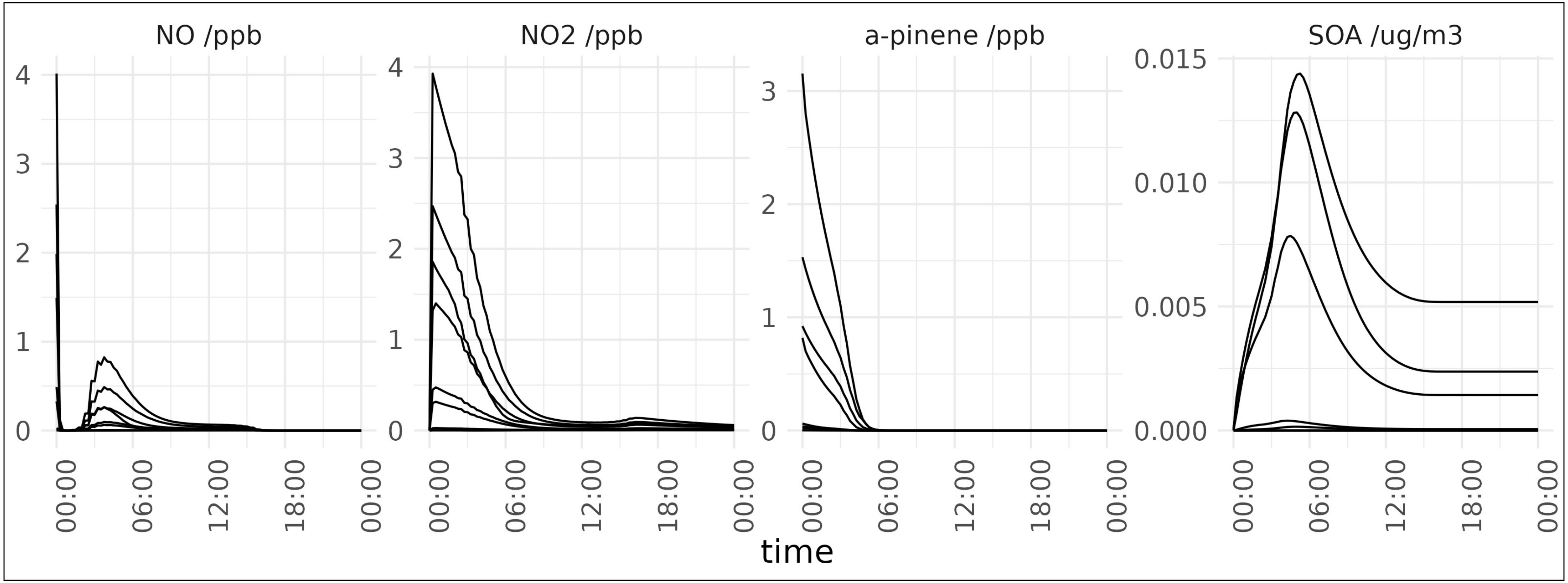

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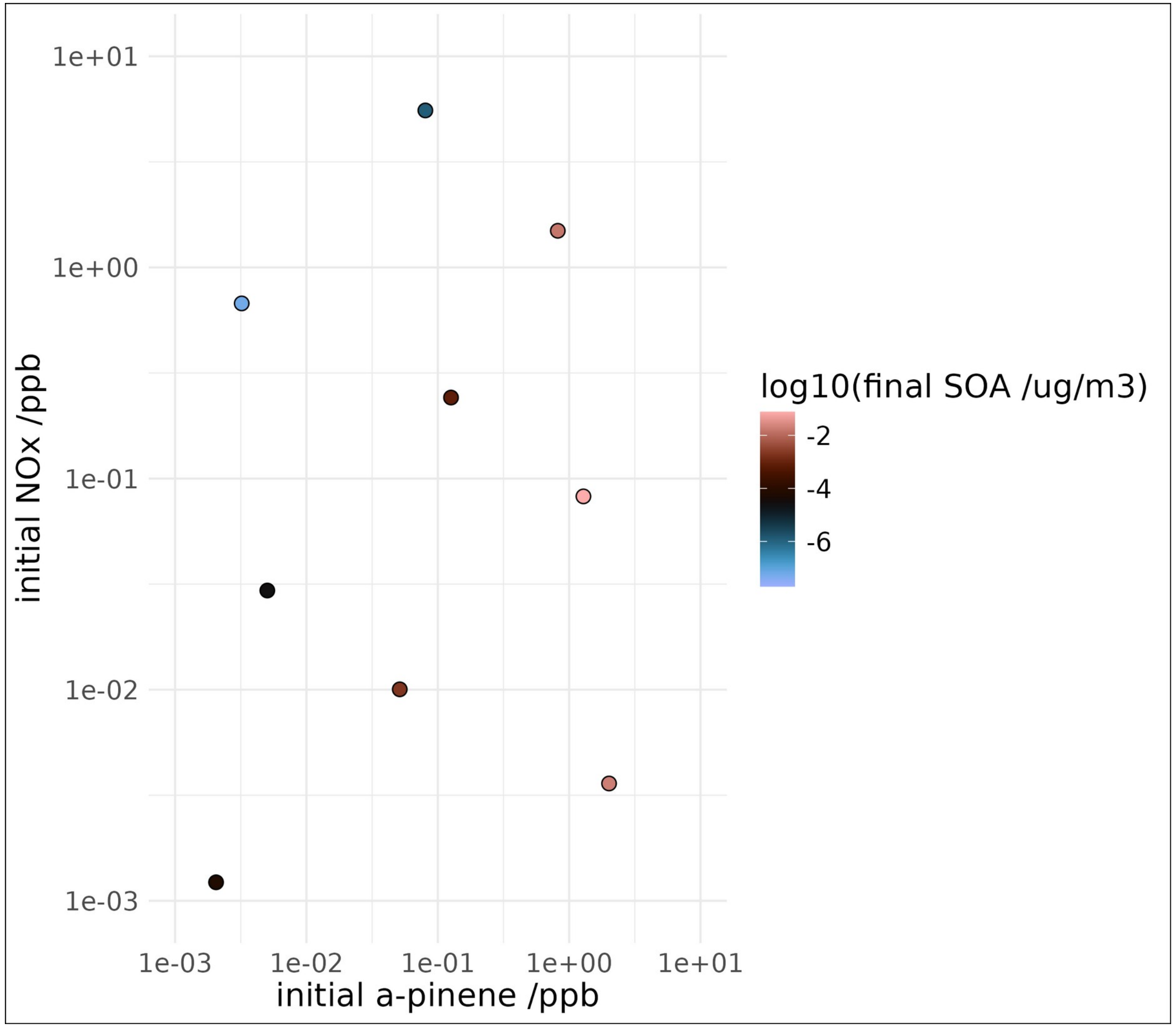

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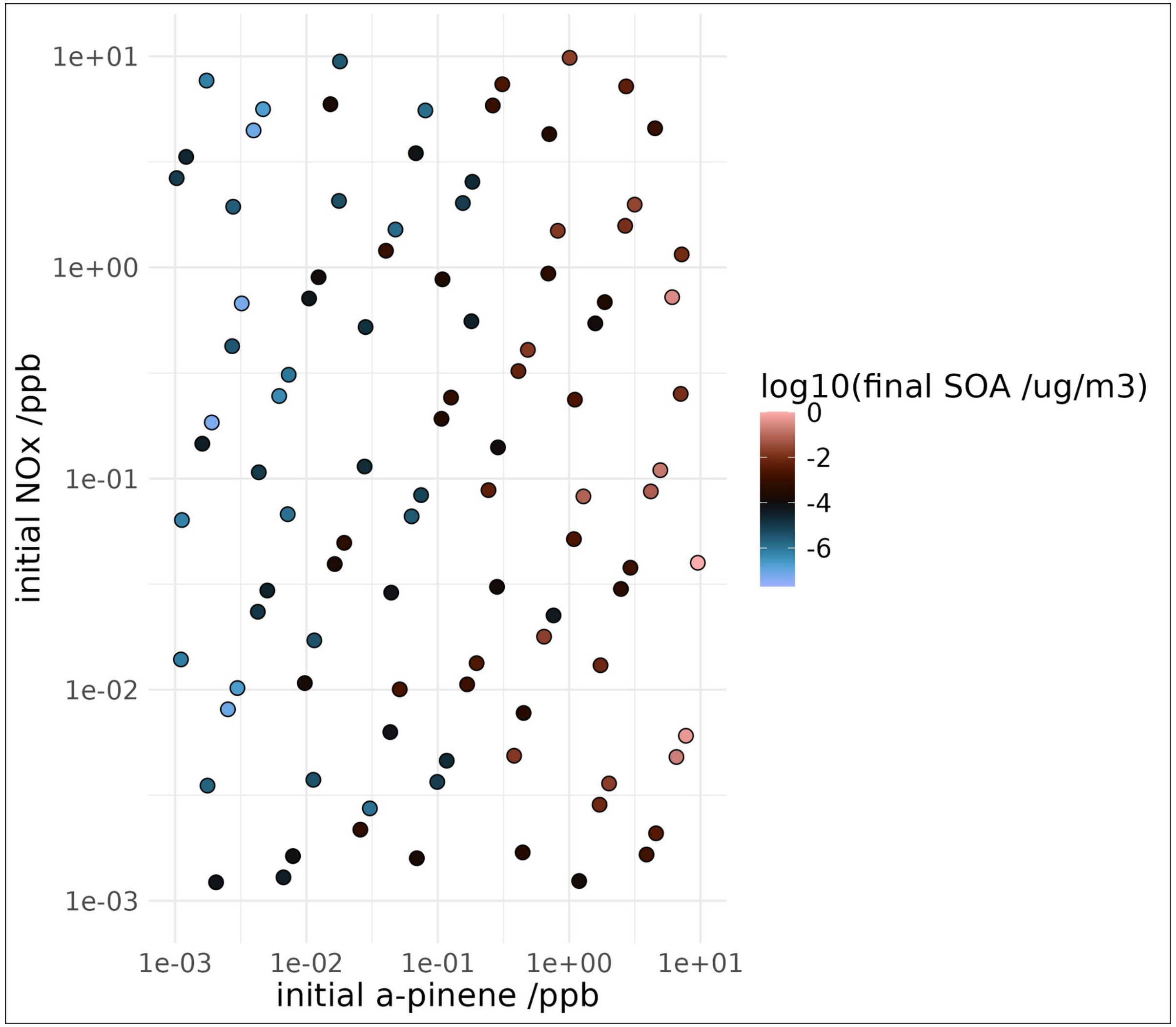

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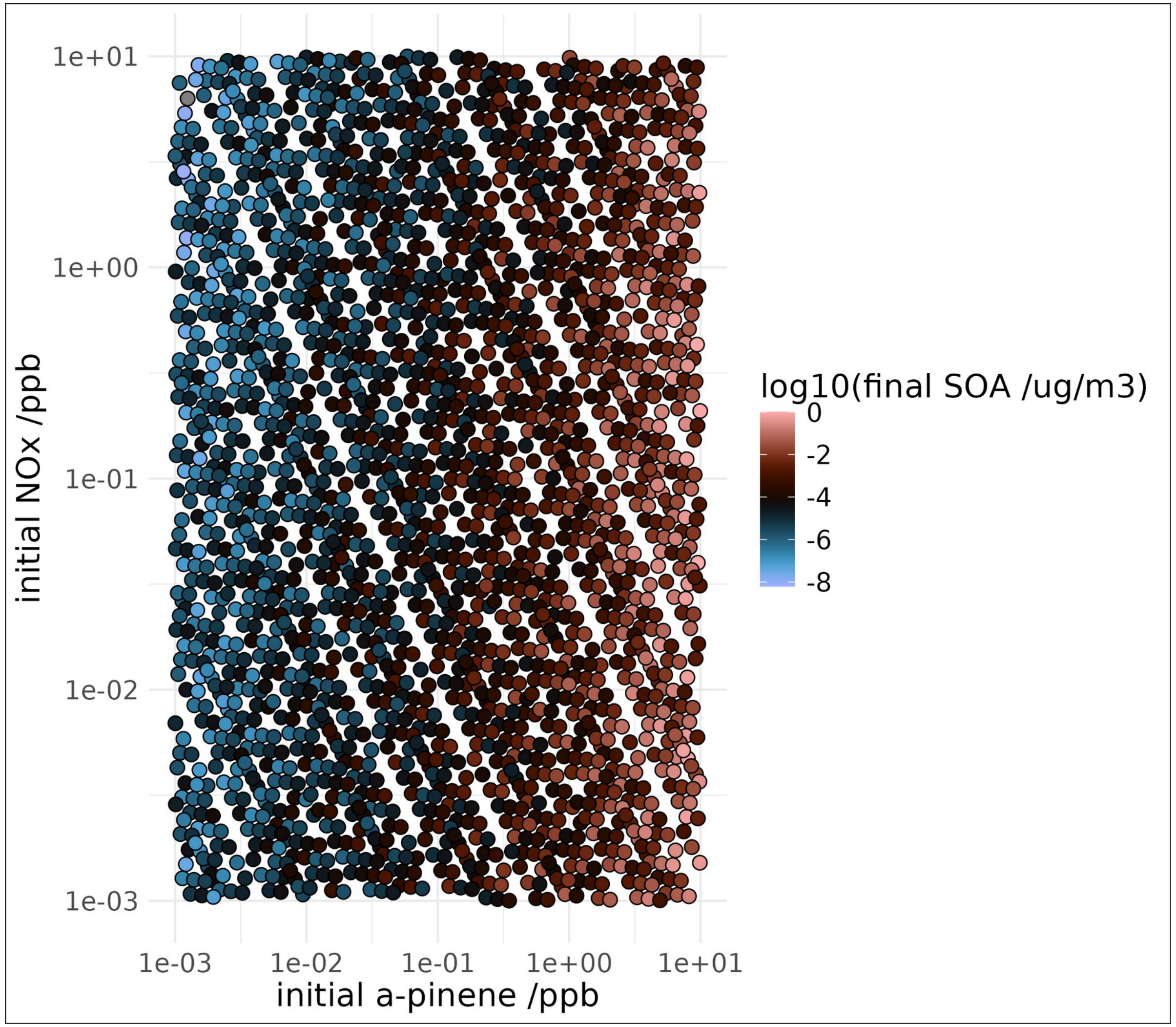
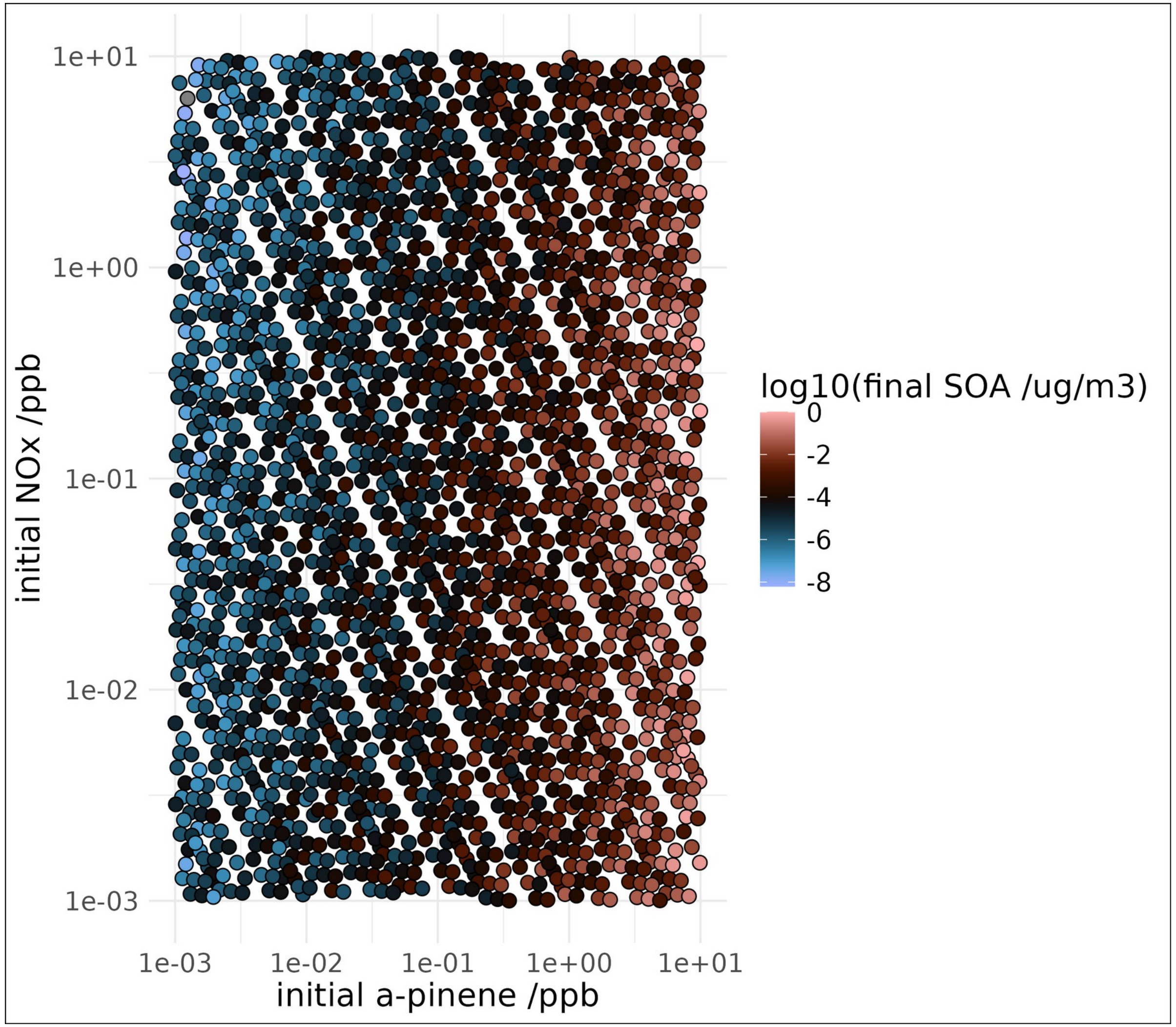
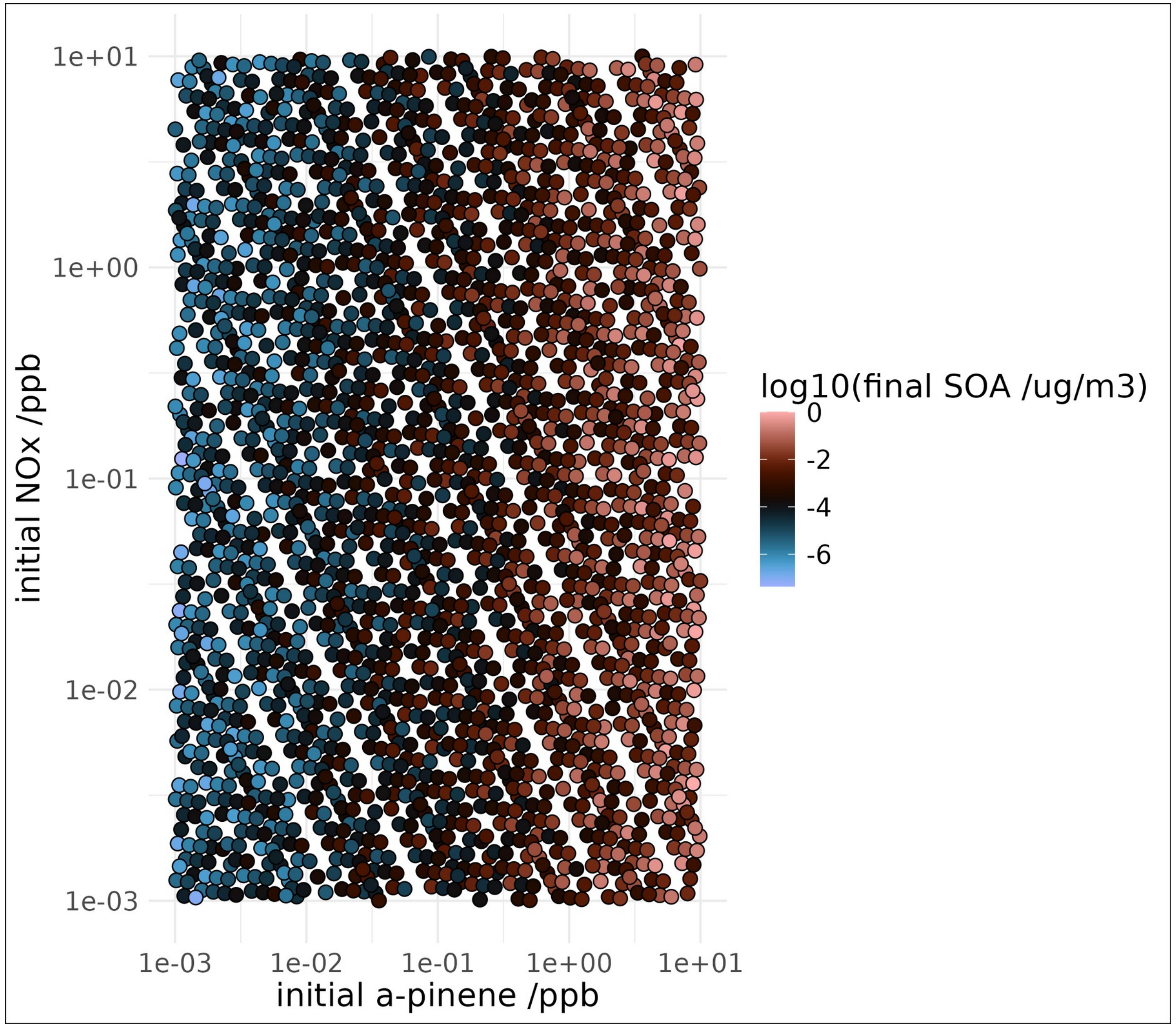


SOA formation sensitivities: CB05 vs CB6 ? + 2 products





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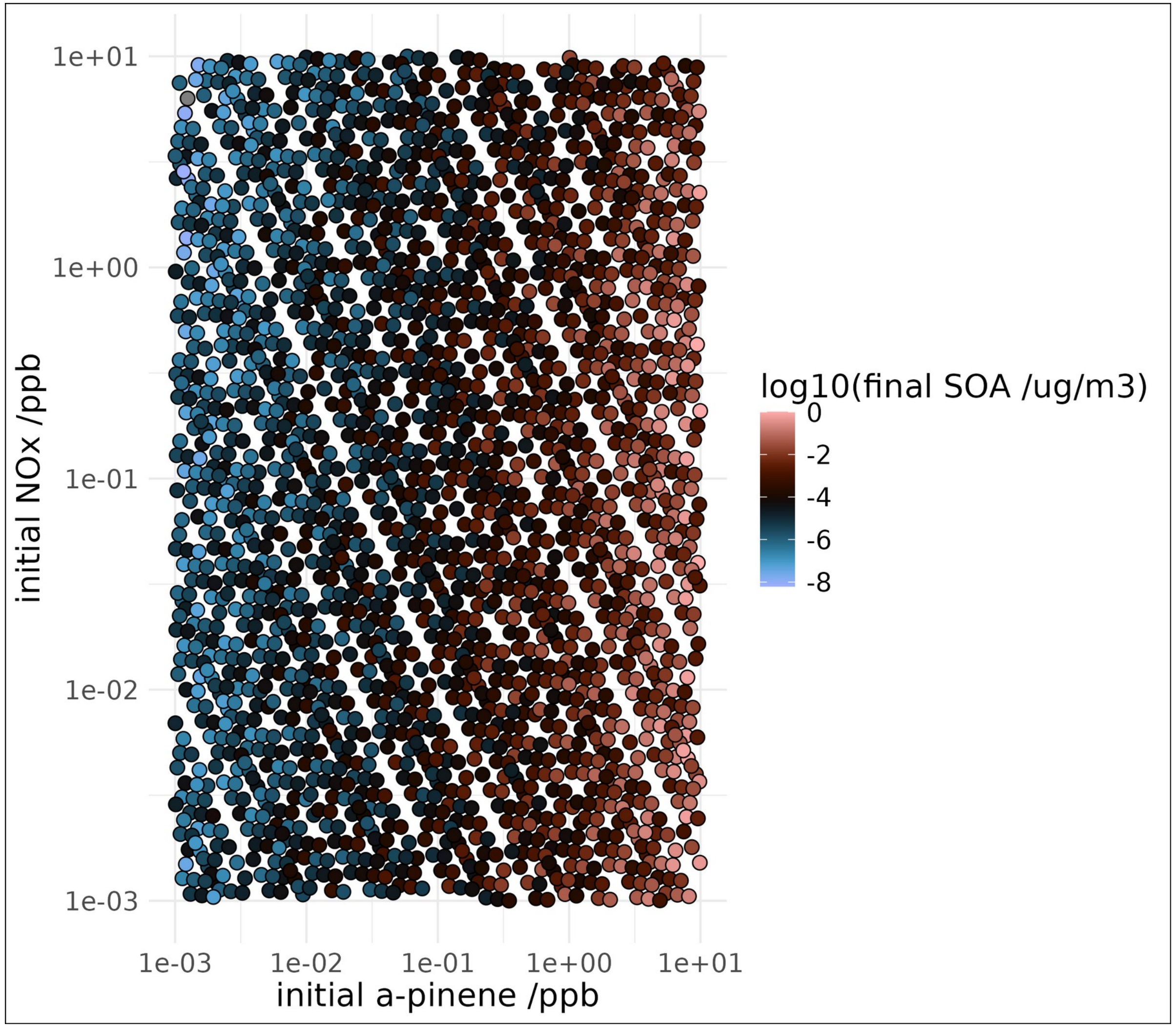
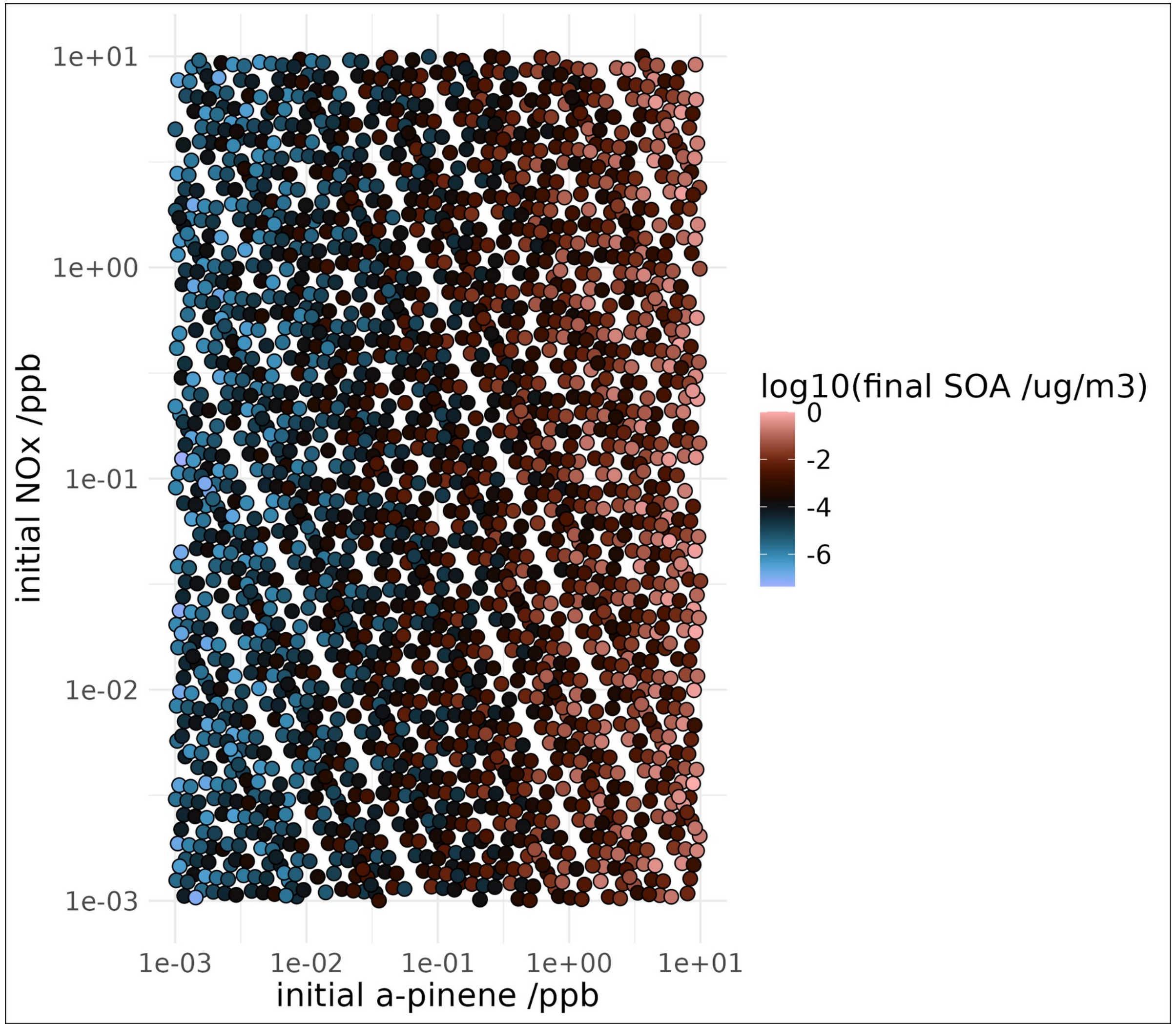
SOA formation sensitivities: CB05 vs CB6 ? + 2 products





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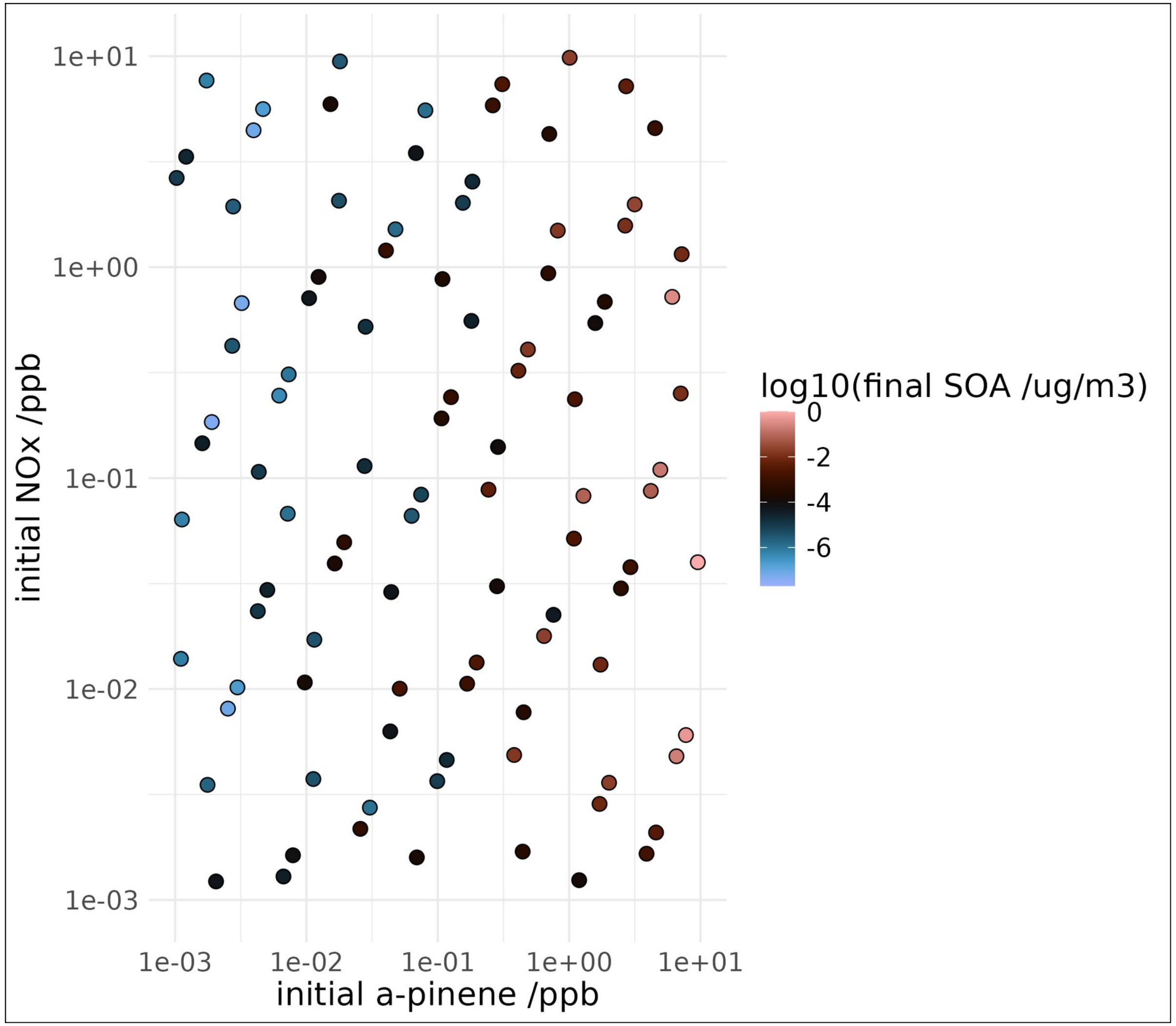
SOA formation sensitivity quantification needed

Sensitivities: introduction to IVARS



(inspired by) **Integrated Variogram Across a Range of Scales**
Razavi and Gupta (2016a, b)

Estimate variogram from variances of all pairs of points separated by a distance h in the considered dimension

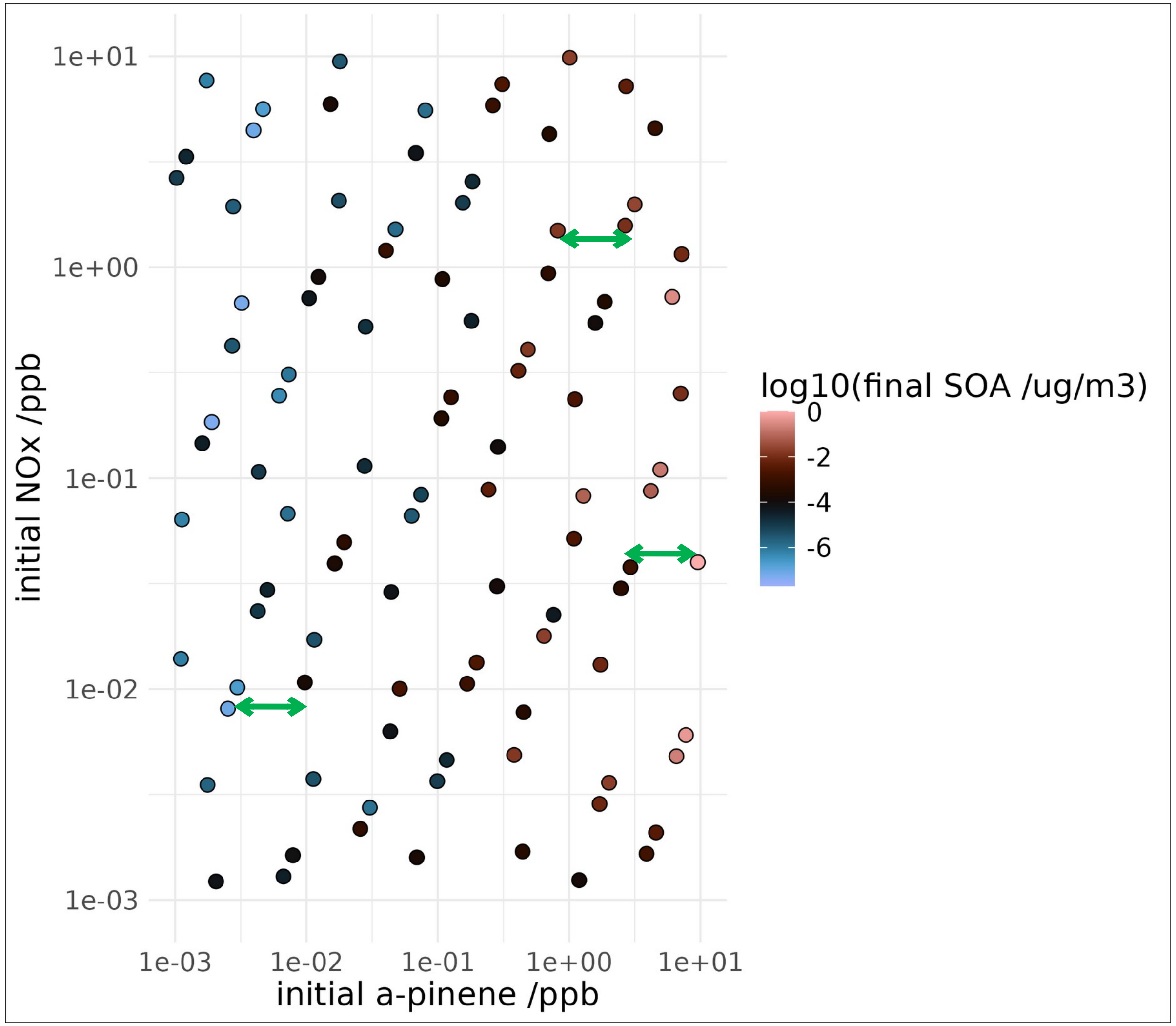
$$\gamma(h) = \frac{1}{2N(h)} \sum_{(i,j) \in N(h)} \left(y(x_i) - y(x_j) \right)^2$$

Integrate the variogram to the desired sensitivity scale H

$$\Gamma(H) = \int_0^H \gamma(h)\, dh$$

repeat for each factor

Sensitivities: introduction to IVARS



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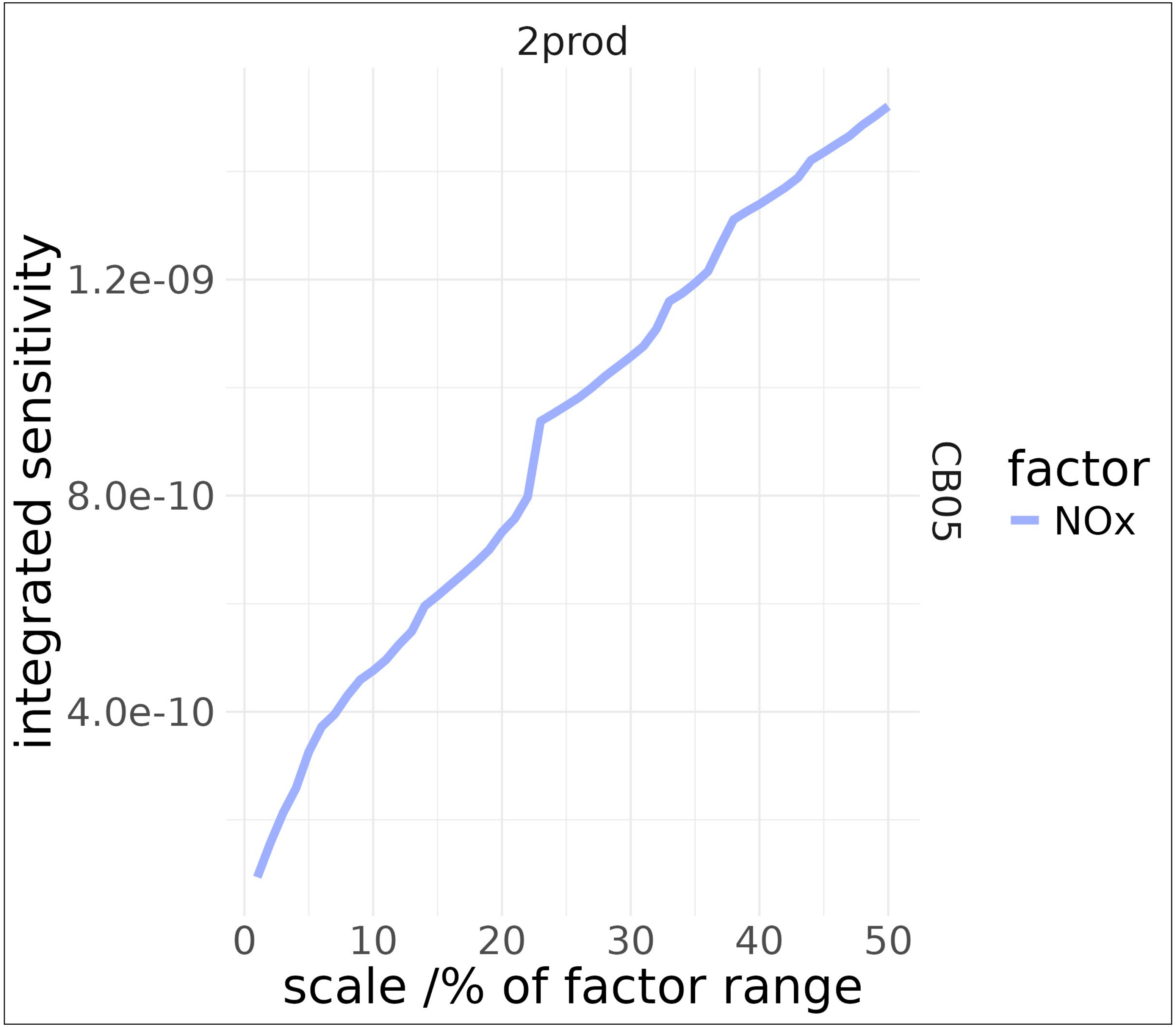
CB05 + 2 products

Sensitivity to the NOx factor similar across all scales



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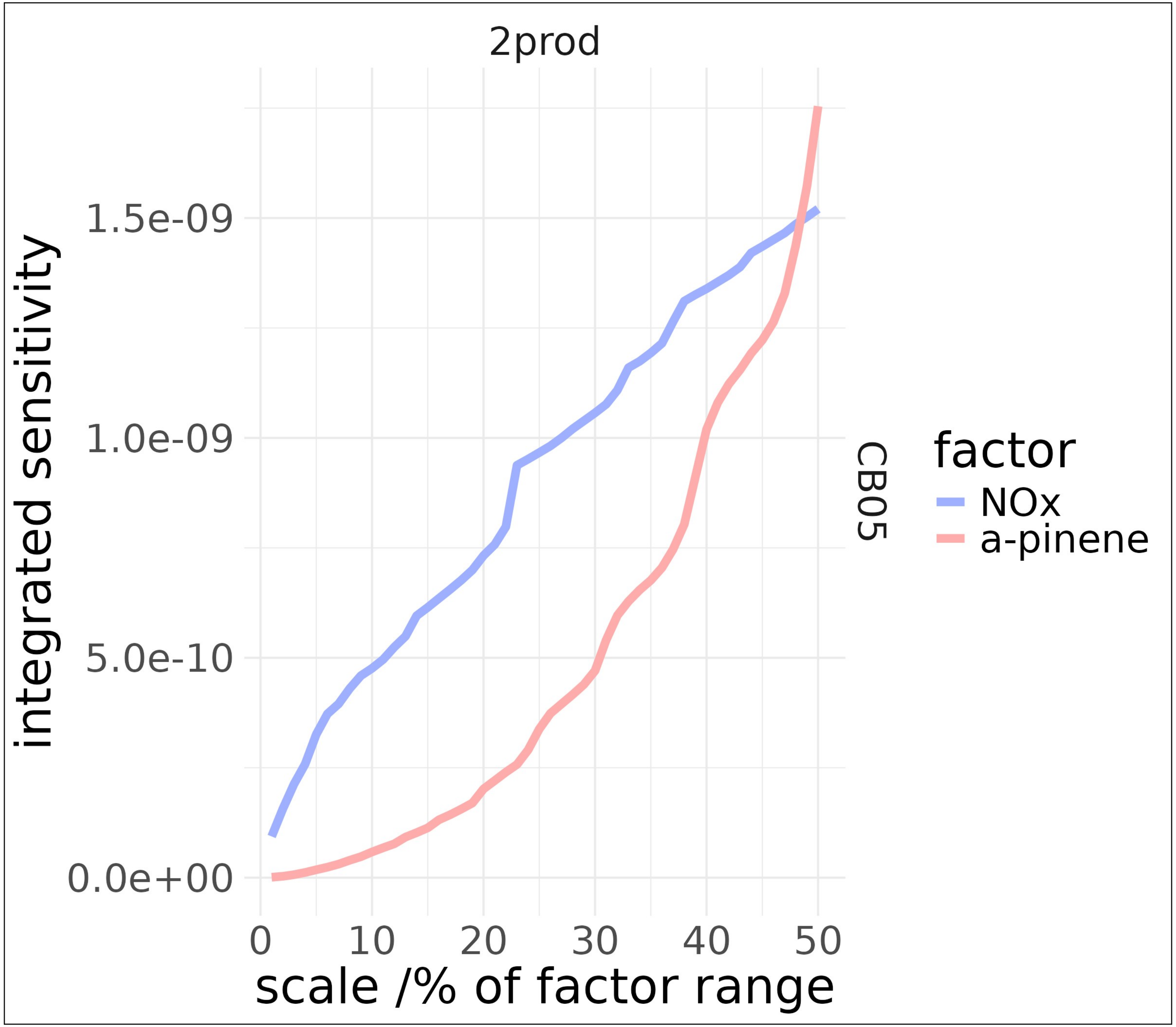
CB05 + 2 products

Sensitivity to the NOx factor similar across all scales

Sensitity to the a-pinene factor is stronger at larger scales



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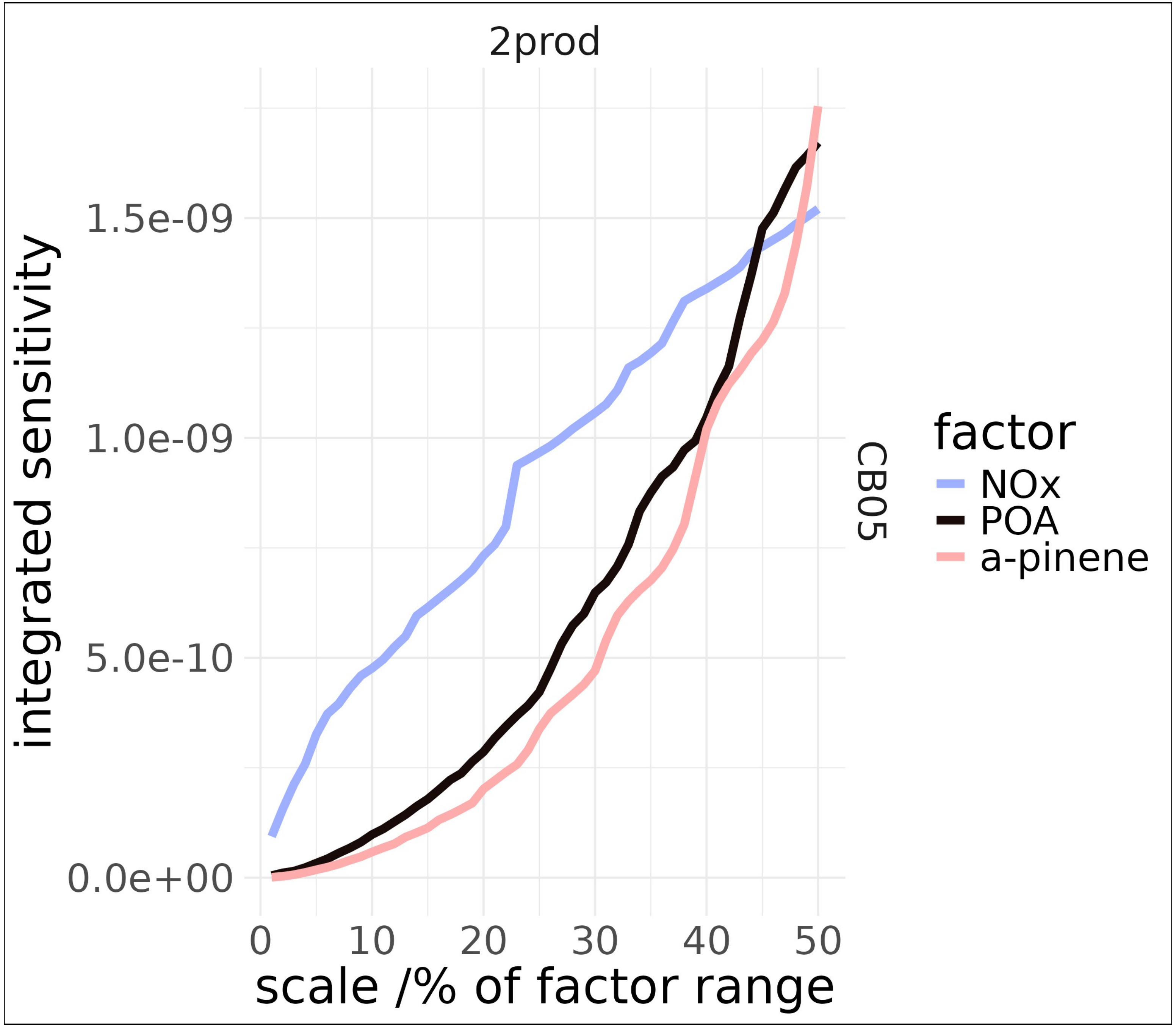
CB05 + 2 products

Sensitivity to the NOx factor similar across all scales

Sensitity to the a-pinene factor is stronger at larger scales

Sensitivity to POA factor is similar to sensitivity to a-pinene



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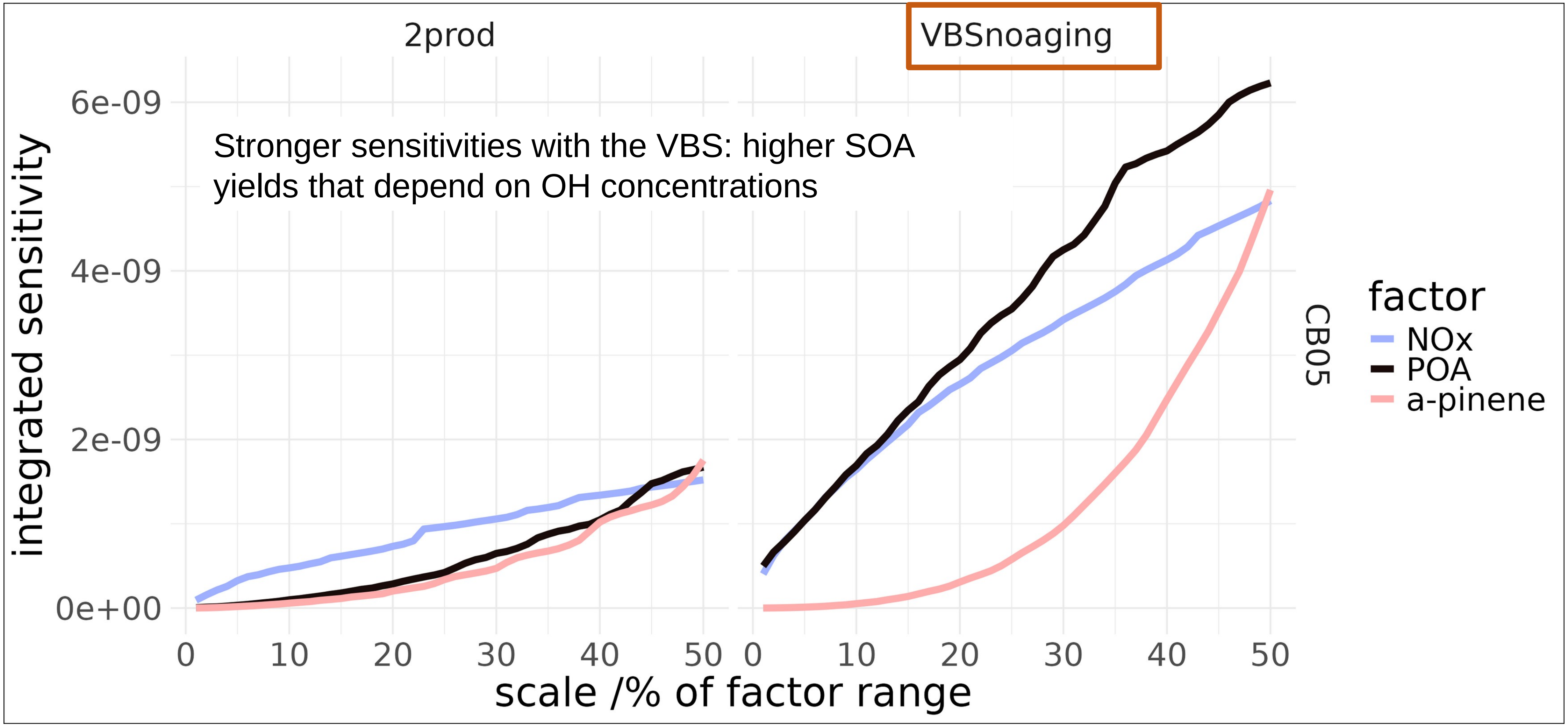
CB05: switching the SOA parameterization



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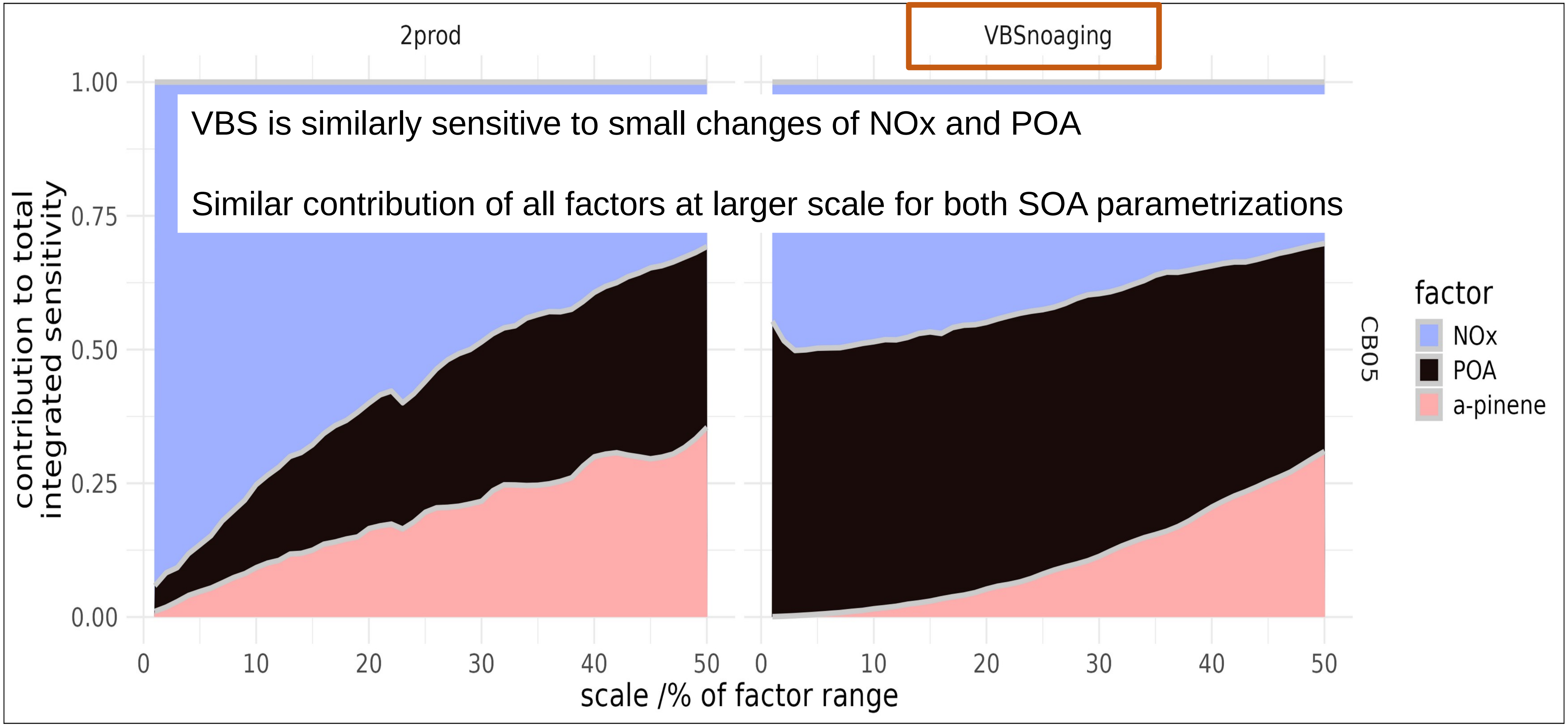
CB05: switching the SOA parameterization



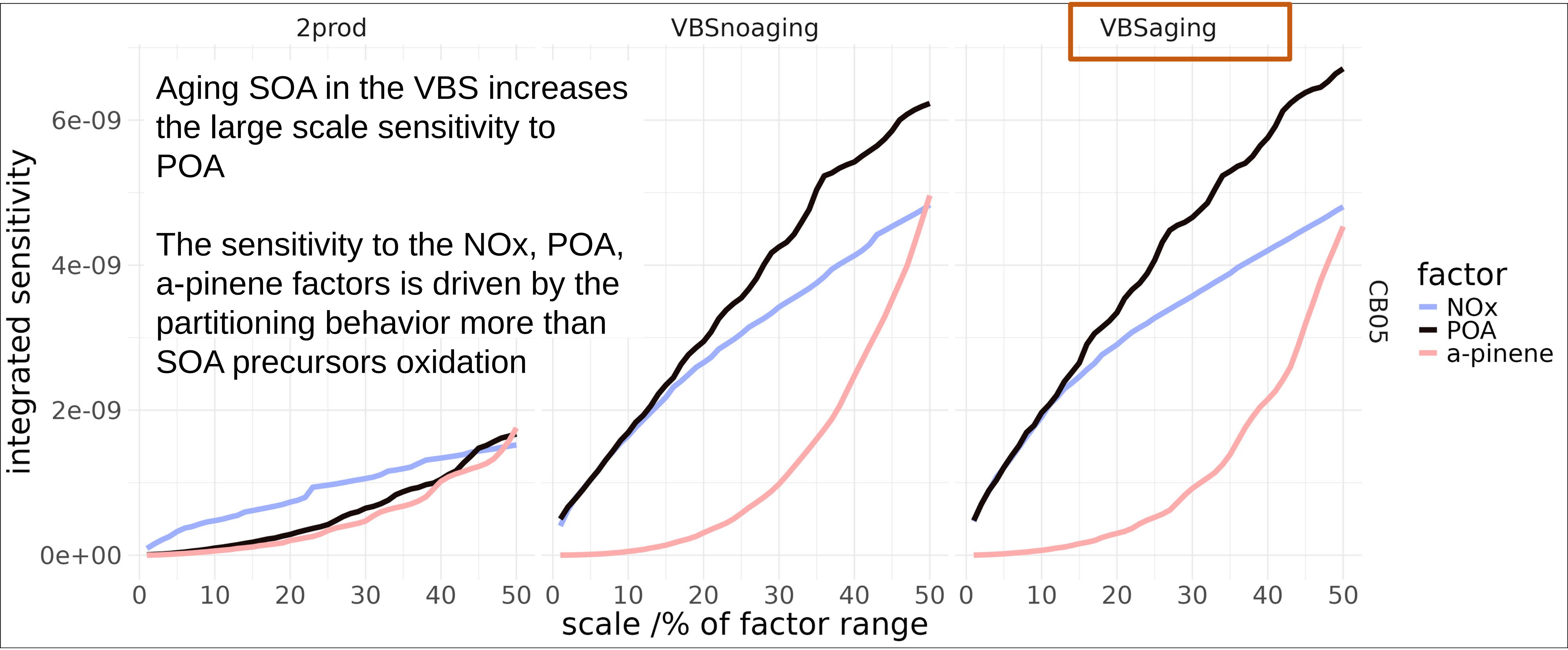
VBS is similarly sensitive to small changes of NOx and POA

Similar contribution of all factors at larger scale for both SOA parametrizations

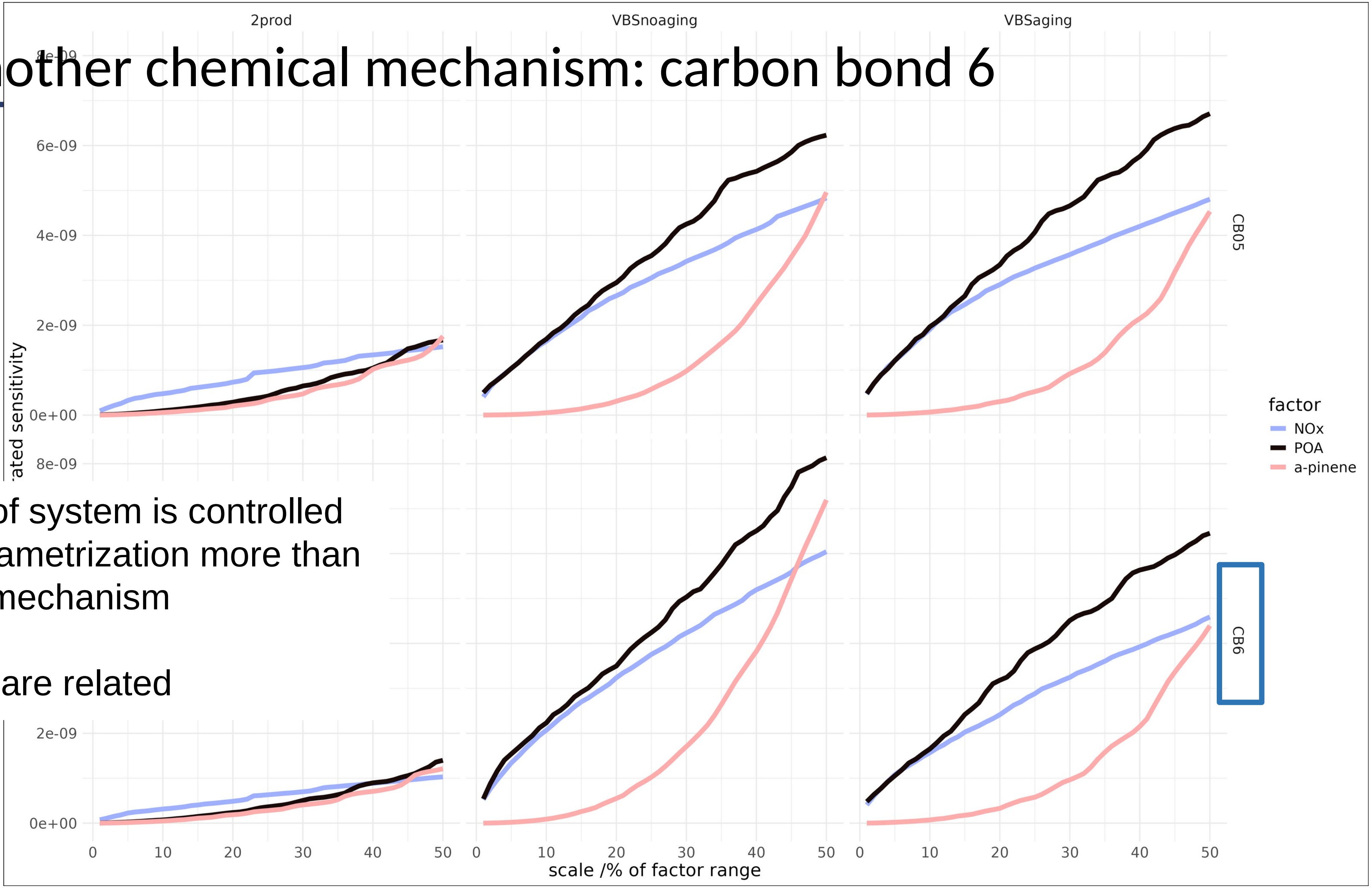
CB05: increasing the complexity of the SOA parameterization

Aging SOA in the VBS increases the large scale sensitivity to POA

The sensitivity to the NOx, POA, a-pinene factors is driven by the partitioning behavior more than SOA precursors oxidation



Let’s try another chemical mechanism: carbon bond 6



The sensitivity of system is controlled by the SOA parametrization more than the gas phase mechanism

CB05 and CB6 are related

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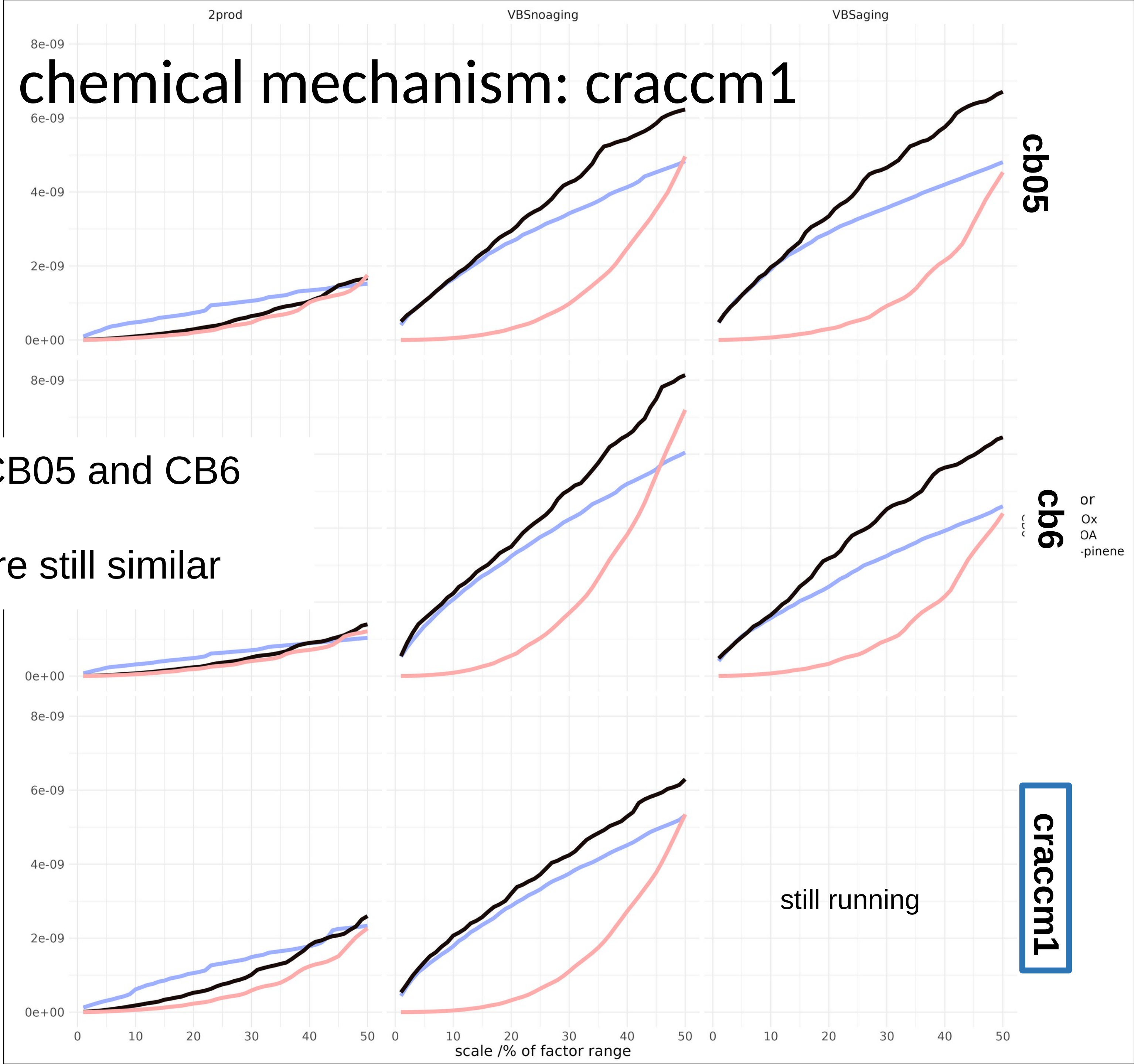
Let’s try an unrelated chemical mechanism: craccm1

CRACCM1 is not related to CB05 and CB6

SOA formation sensitivities are still similar



Preliminary conclusions

- Studies of emissions scenarios future effects rely on comparing the impacts of changes in factors (NO_X, POA, VOC) on targets (SOA)
- The selected SOA parametrization can strongly affect the model response to change at small and large scales
- The gas phase chemistry choice has a secondary impact on SOA sensitivity (this would be different for ozone!)
- This is only the beginning: need for more gas phase mechanisms and SOA parametrization representative of atmospheric community uses, investigation of more sensitivity factors and targets
- What is the SOA formation sensitivity of the real world?

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Thanks for your attention!

camille.mouchel@bsc.es