Trimming the Iterative Fat of Equilibrium Thermodynamic Models Using Neural Networks

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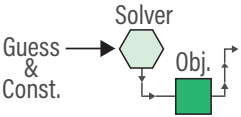
Bourses de recherche postdoctorale

IAMA December, 5 2019

The neural network shortcut

- Iterative solvers can be computationally expensive.

Typical approach



The neural network shortcut

- Iterative solvers can be computationally expensive.
- But, they are transparent and scientifically robust.
- Neural networks are the ultimate black box.

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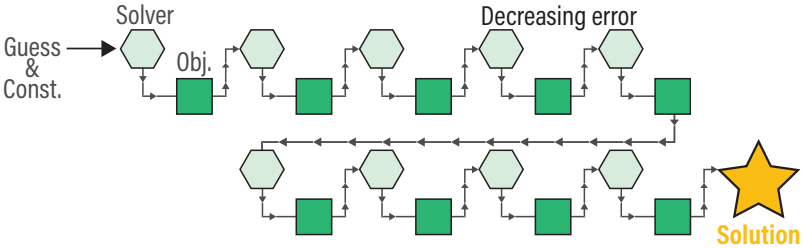
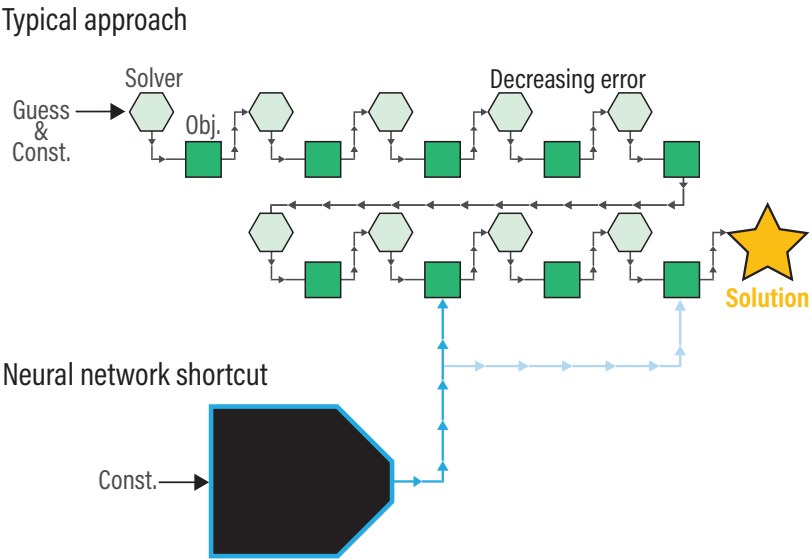


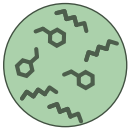
The neural network shortcut

- Iterative solvers can be computationally expensive.
- But, they are transparent and scientifically robust.
- Neural networks are the ultimate black box.
- But, they can be faster than iterative solvers.
- A combined approach is preferable.



Modeling aerosol mass concentrations

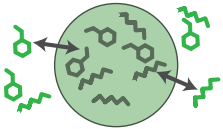
Organics

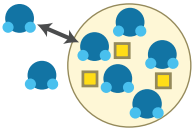


Inorganic Salts

Emission inventories and chemistry

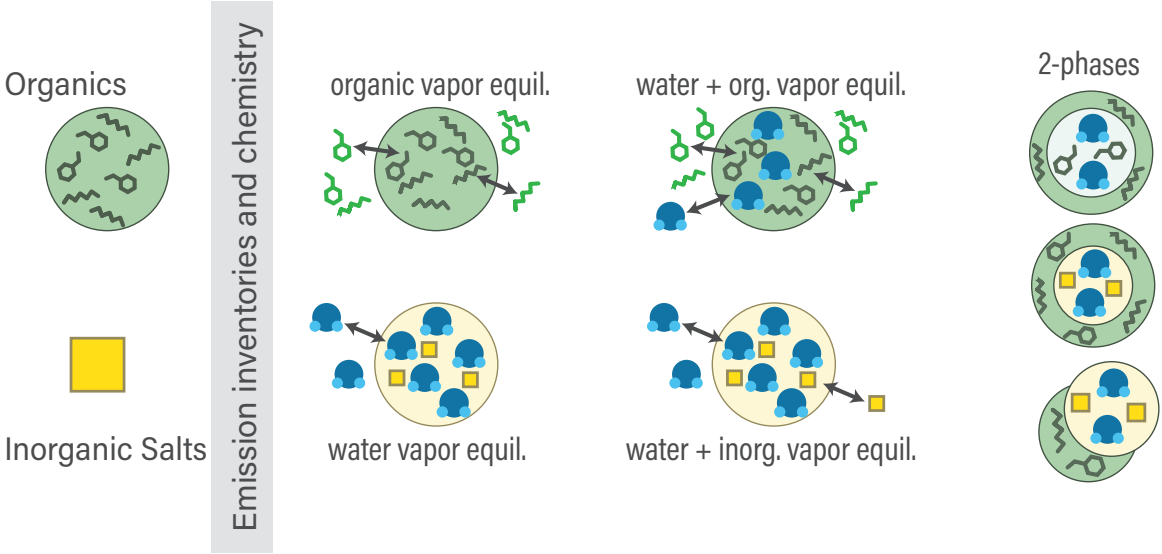
organic vapor equil.





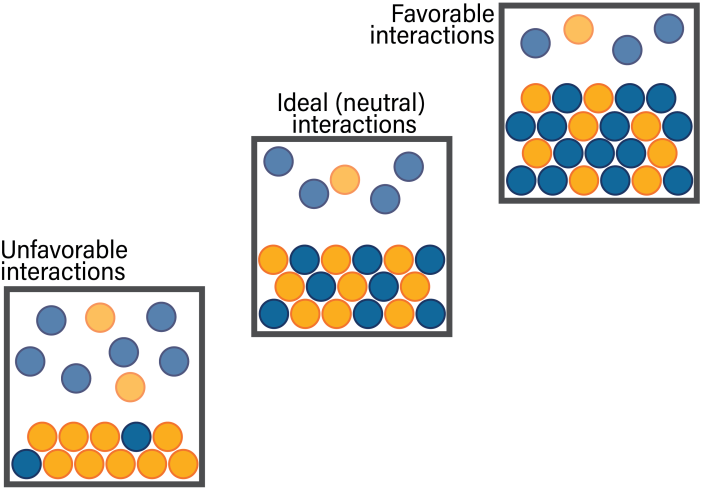
water vapor equil.

Modeling aerosol mass concentrations



Modified Raoult's law

- Mixing decreases the effective vapor pressure of each component.
- $p_j = p_j^{\mathrm{sat}} x_j \gamma_j = p_j^{\mathrm{sat}} a_j$
- The activity coefficient (γ_j) parameterizes the energetic cost (favorability) of mixing.



Modified Raoult's law

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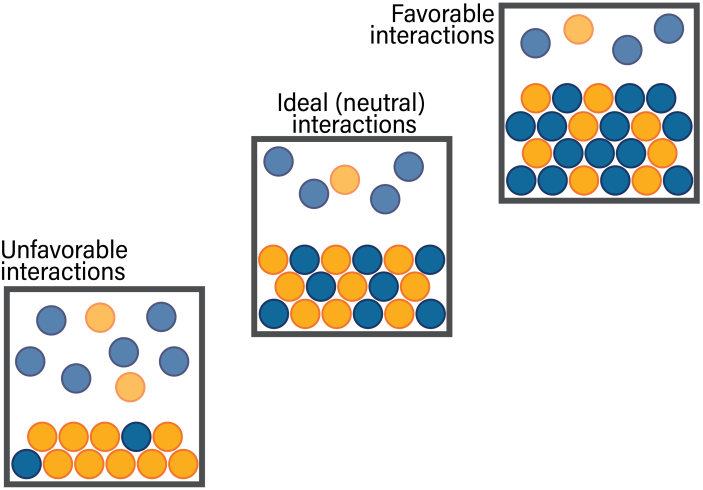
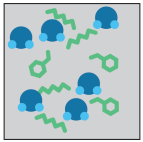
- $p_j = p_j^{\mathrm{sat}} x_j \gamma_j = p_j^{\mathrm{sat}} a_j$
- The activity coefficient (γ_j) parameterizes the energetic cost (favorability) of mixing.
- In terms of an effective volatility.
- $C_j^* = C_j^{\mathrm{sat}} C_{\Sigma_k}^{\Sigma_\pi} \frac{\gamma_j q_j^\pi}{M_j \sum_k \frac{C_k^\pi}{M_k}}$



BAT: A reduced complexity model

- AIOMFAC is used as a high fidelity reference.

Multi-component mixture



$\gamma_{org,1}$

$\gamma_{org,2}$

γ_{w}

Non-ideal interactions among all species

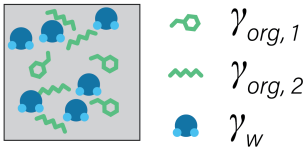
Gorkowski et al. (2019)

BAT: A reduced complexity model

- AIOMFAC is used as a high fidelity reference.
- A simpler Binary Activity Thermodynamics model is built using AIOMFAC generated data.
- `github.com/Gorkowski/Binary_Activity_Thermodynamics_Model`

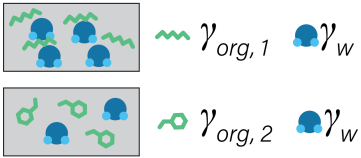
BAT
Binary Activity Thermodynamics

Multi-component mixture



Non-ideal interactions among all species

BAT approximation



Non-ideal interactions with water

Gorkowski et al. (2019)

BAT: Essential equations

- Following a power series expansion of molar excess Gibbs energy of mixing (G^E) (Redlich and Kister, 1948).
- Generalized by parameterizing the expansion coefficients (c_n).
- Independent variables
  mole fraction (x_{org}) or scaled Vol. (ϕ_{org}), O:C, & M_{org}

$$G^E/RT = \phi_{org}(1-\phi_{org})\left[c_1 + c_2(1-2\phi_{org})\right] \quad (1)$$

$$c_n = a_{n,1}\ \exp(a_{n,2} \times O:C) + a_{n,3}\ \exp\left(a_{n,4}\ \frac{M_w}{M_{org}}\right) \quad (2)$$

Gorkowski et al. (2019)

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- Generalized by parameterizing the expansion coefficients (c_n).
- Independent variables
  mole fraction (x_{org}) or scaled Vol. (ϕ_{org}), O:C, & M_{org}
- Dependent variables
  water activity ($a_w \times 100\,\% \approx RH$) and organic activity

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$$\ln(\gamma_w) = (G^E/RT) - x_{org}\frac{d(G^E/RT)}{dx_{org}} \quad (3)$$

$$a_w = \gamma_w(1-x_{org}) \quad (4)$$

Gorkowski et al. (2019)

Inverting the BAT model

For gas–particle partitioning we need γ_j and $x_{org,j}$; both are used in $C_j^* = C_j^{\mathrm{sat}}\, C_{\Sigma_k}^{\Sigma_\pi} \frac{\gamma_j\, q_j^\pi}{M_j \sum_k \frac{C_k^\pi}{M_k}}$.

3-D Models will have:

- O:C
- M_{org}
- RH $\approx a_w \times 100\,\%$

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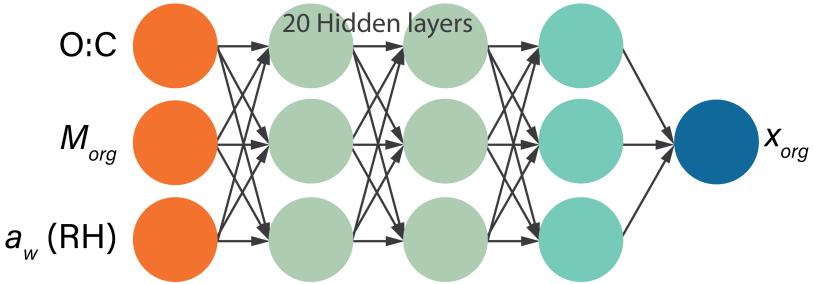
3-D Models will have:

- O:C
- M_{org}
- RH $\approx a_w \times 100\,\%$

- BAT model must be inverted to use a_w as an input.
- Iterative refinement is possible by varying $x_{org,j}$ to match a_w.
- Instead neural networks are used to directly predict $x_{org,j}$ to a high degree of accuracy.

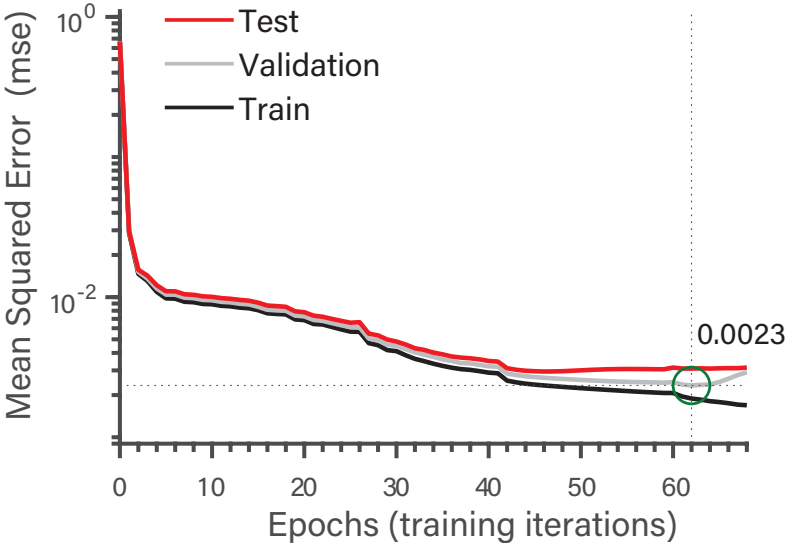
Building a Neural Network

- We used a deep belief network of artificial neurons.
- In effect this acts as a generalized curve fitting tool.
- A training database of random inputs and targeted outputs is generated with the BAT model.



Training of Neural Network

- BAT NN was trained on 9.8×10^6 data points.
- VBS NN was trained on 1.3×10^4 data points.
- Training, validation, and test split of 70/15/15.



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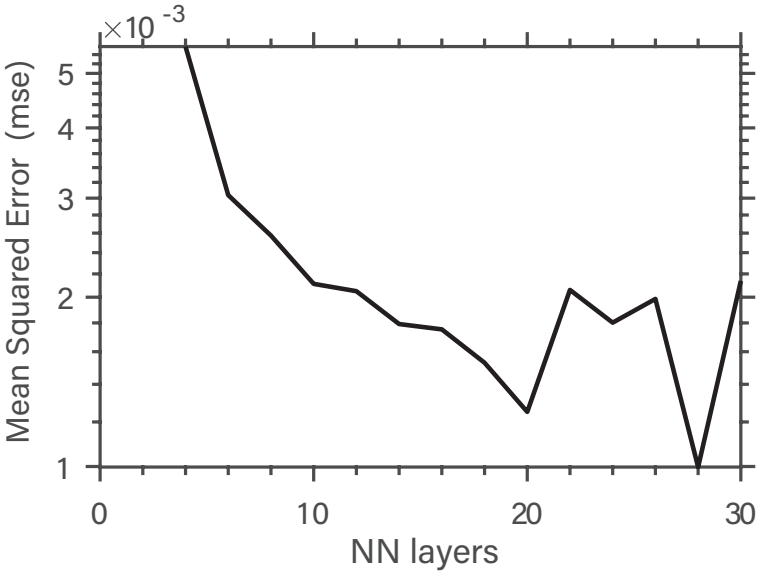
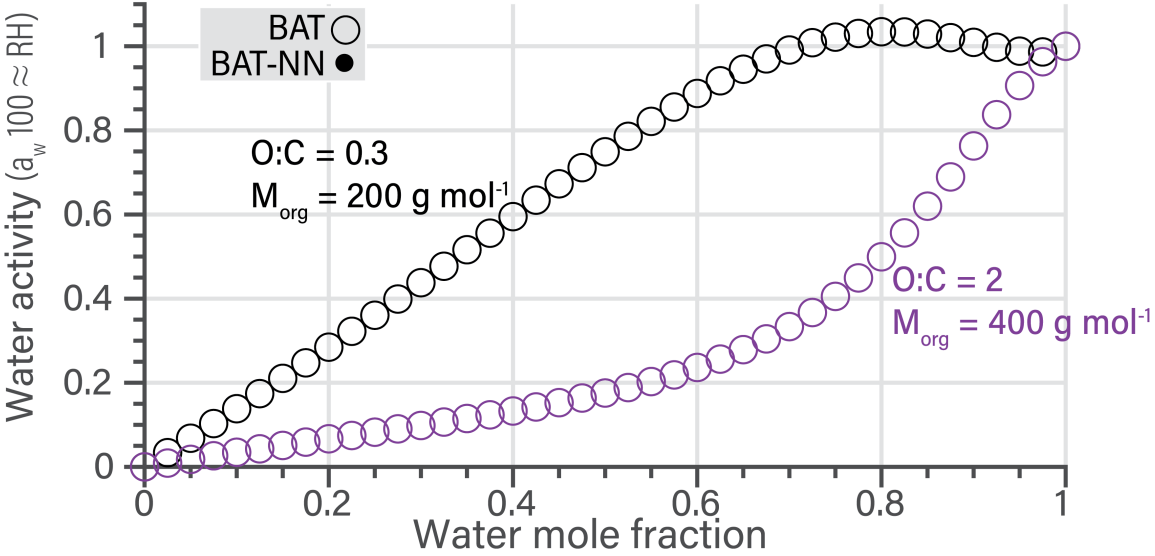


Neural Networks in the context of BAT



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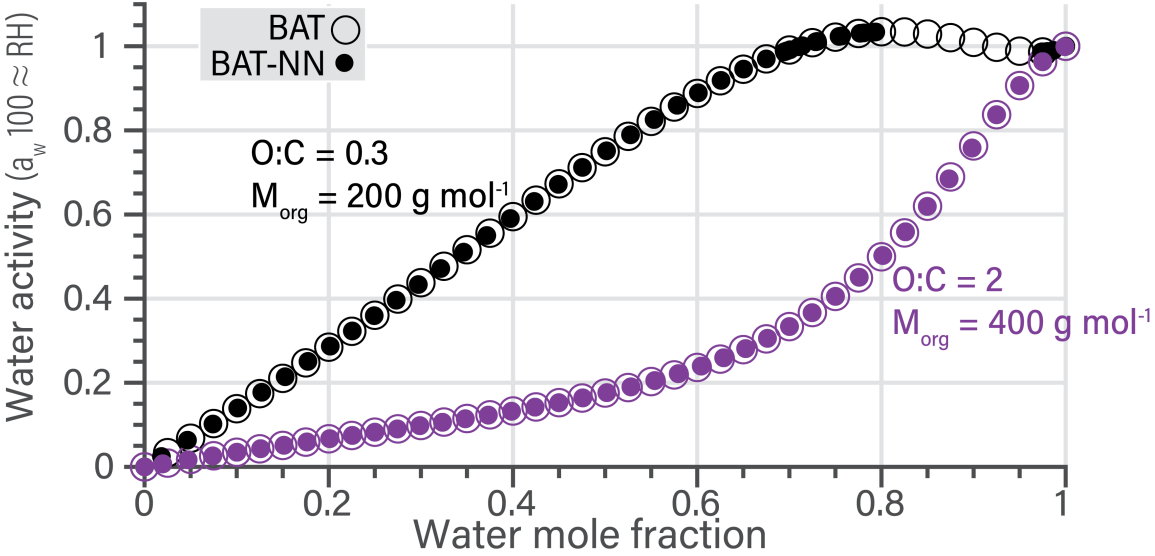
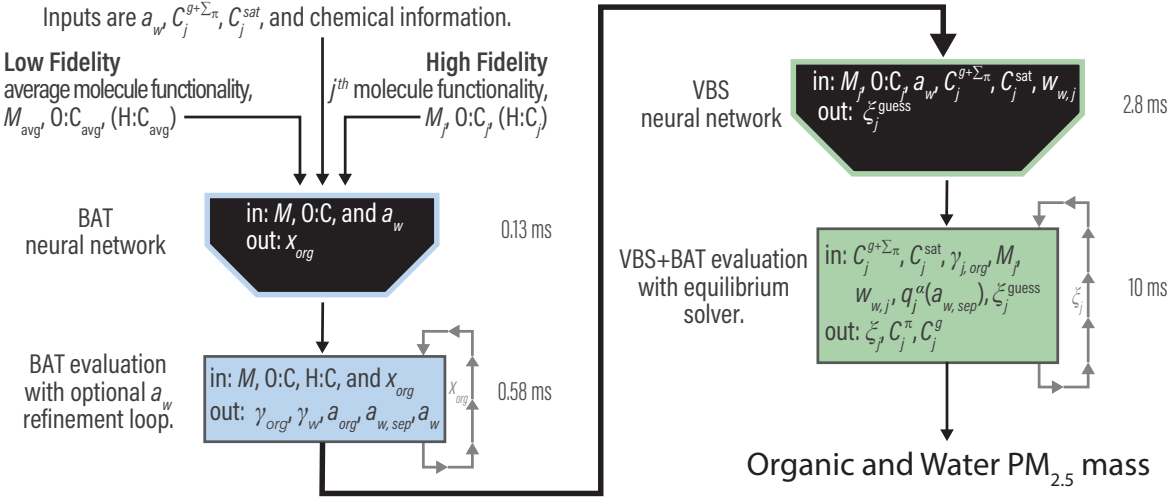
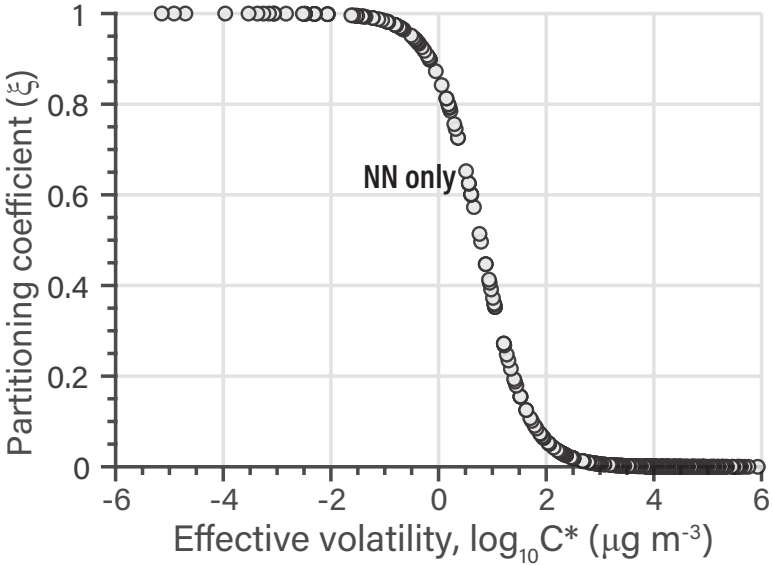


Targeting 3-D models (GEOS-Chem)



Isoprene SOA: NN underpredicts

- MCM simulation outputs.
- Isoprene and O_3 at 100 ppb.
- Simulation of 343 products after 12 hr.
- Used the EVAPORATION model for vapor pressure estimations.



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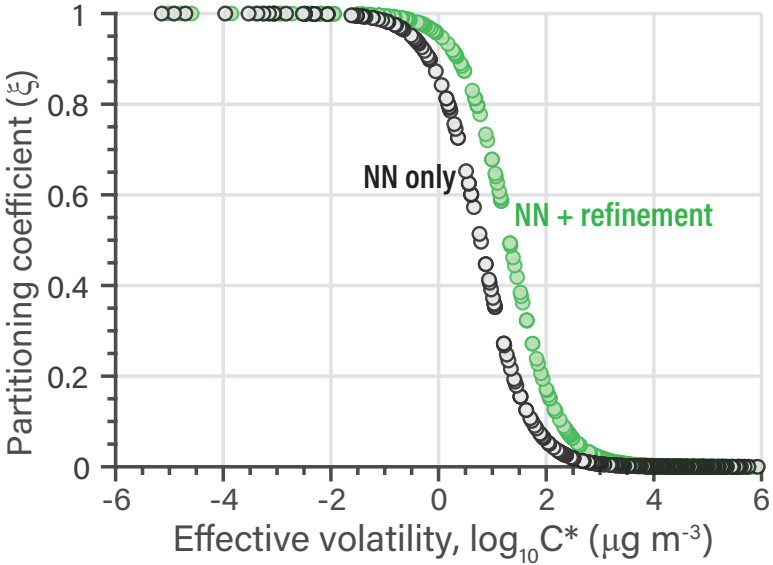

Isoprene SOA: PM mass agrees well

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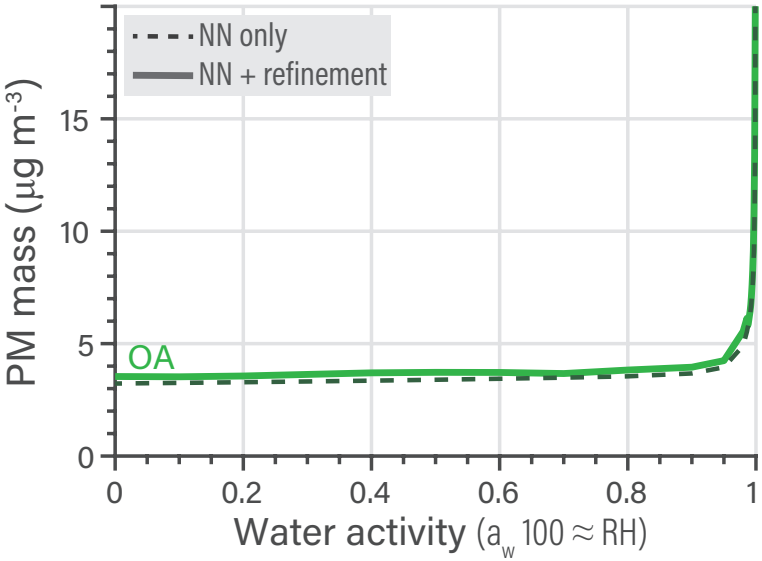

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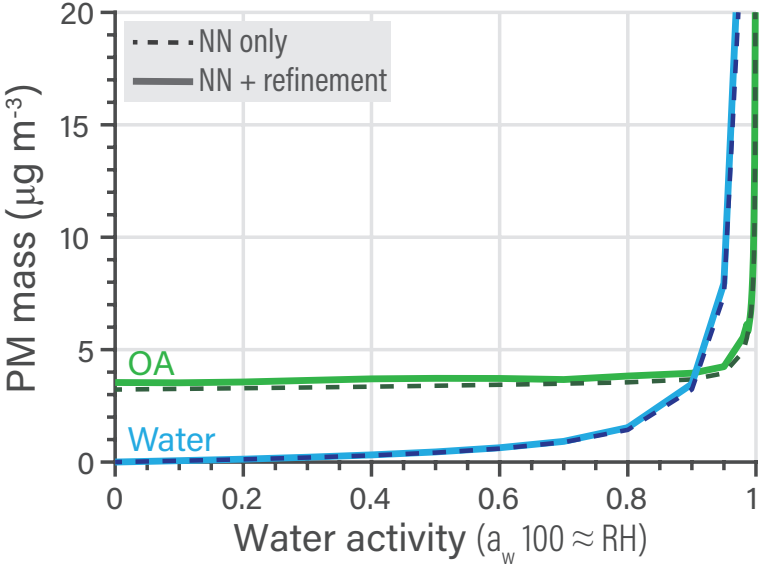

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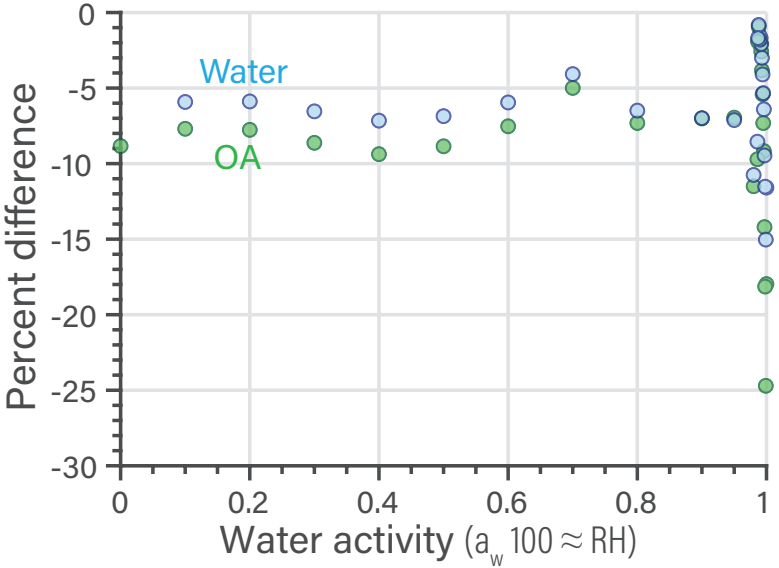

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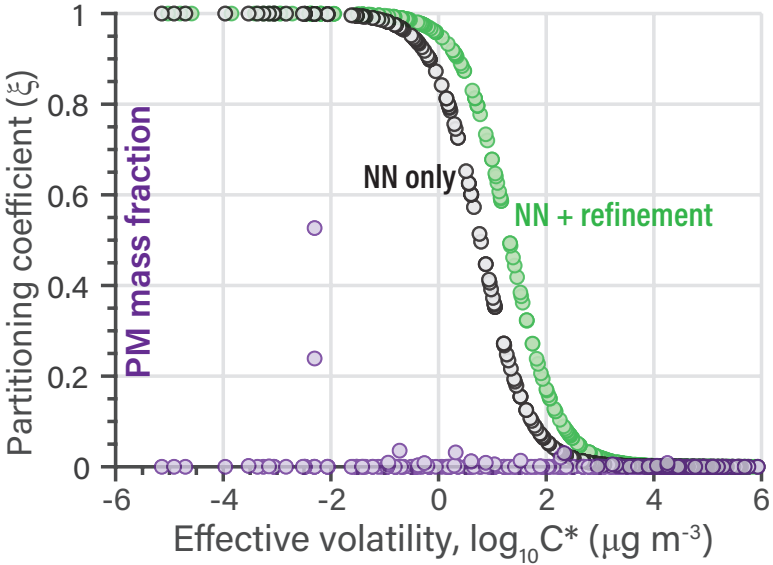
- The errors in partitioning coefficients are not directly translated to PM mass errors.



α-Pinene SOA: NN underpredicts

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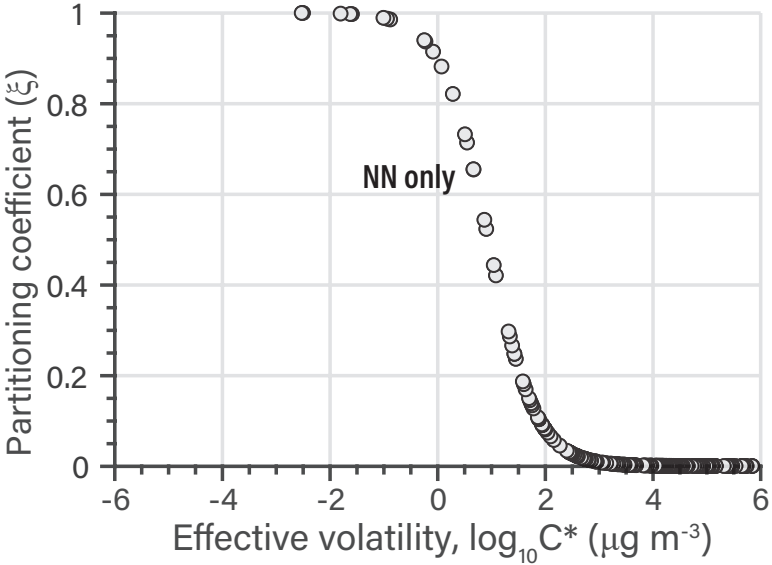

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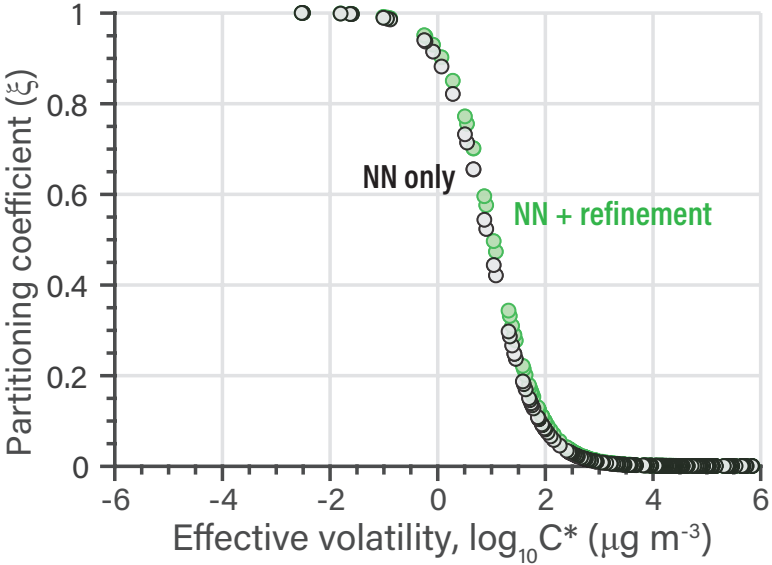
- NN+refinement was 2-5 times faster than iterative refinement only.



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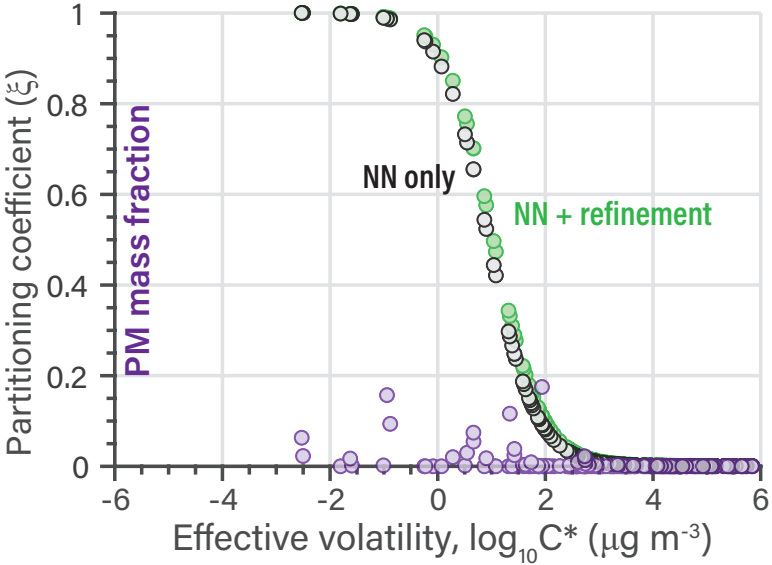

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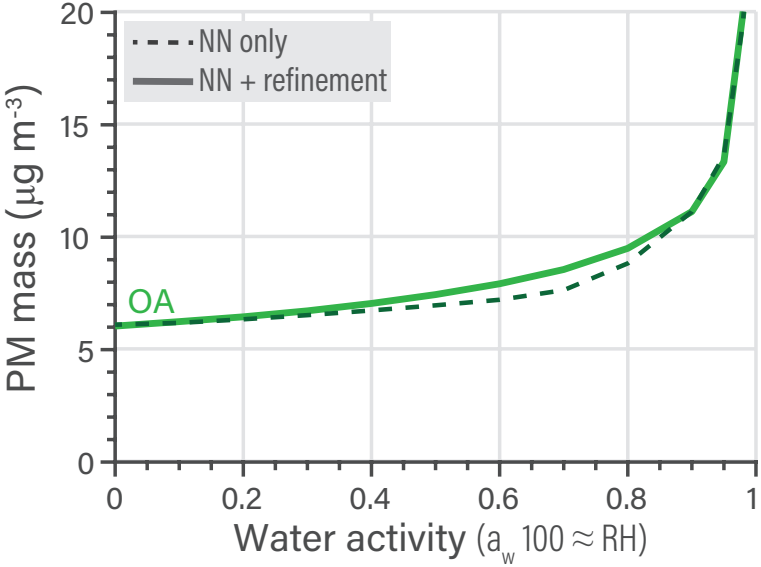

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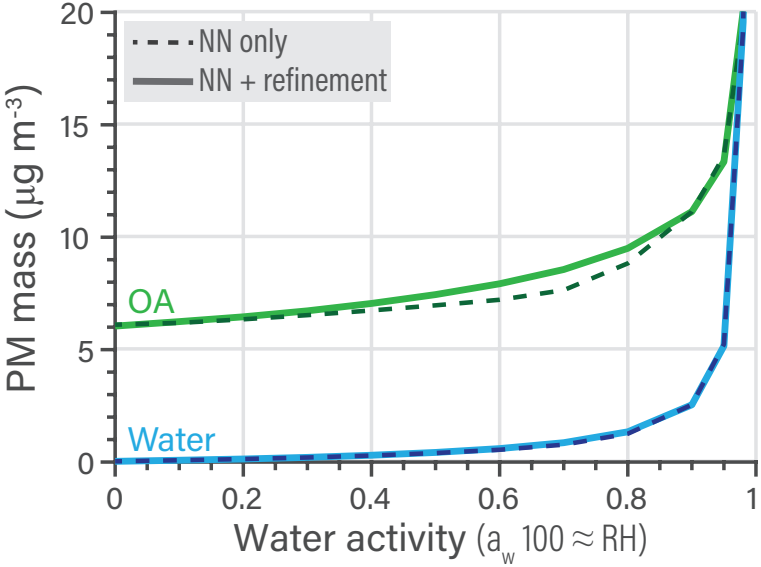

α-Pinene SOA: PM mass agrees well

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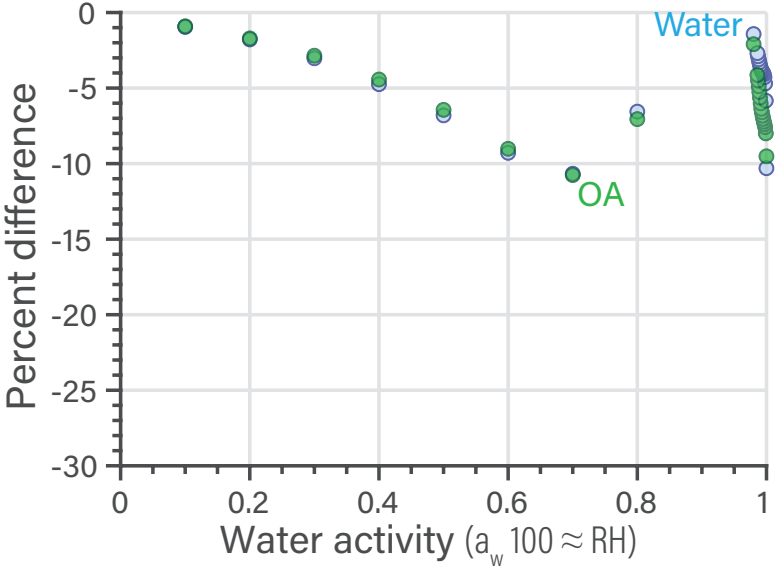
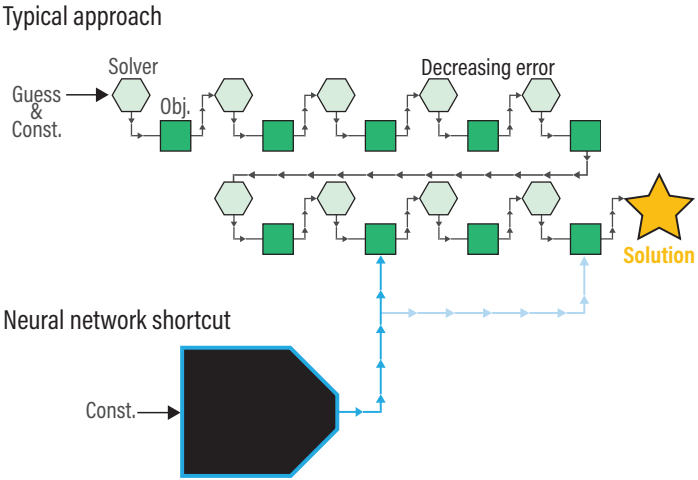
- The mass errors are $< 11\%$.



The neural network shortcut

- The combination of iterative solvers and NN can be accurate and efficient.



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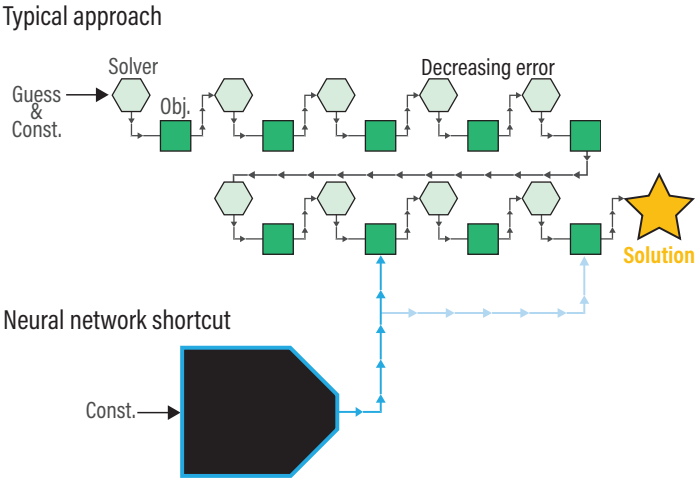
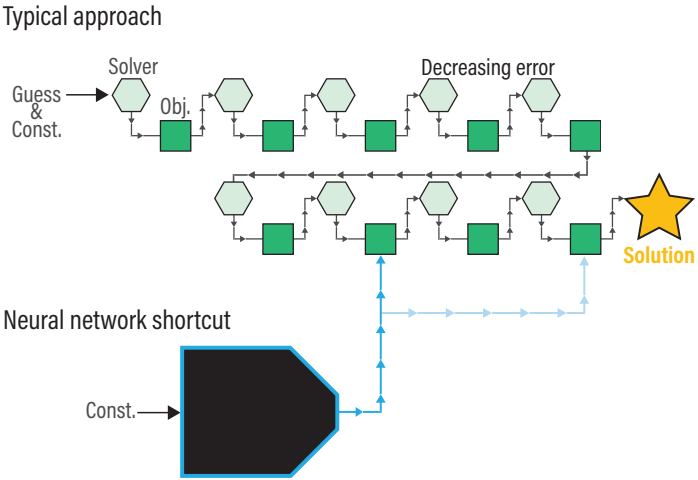
- Extensions of this methodology to other processes, is possible as long as the computation trade-off is favorable.

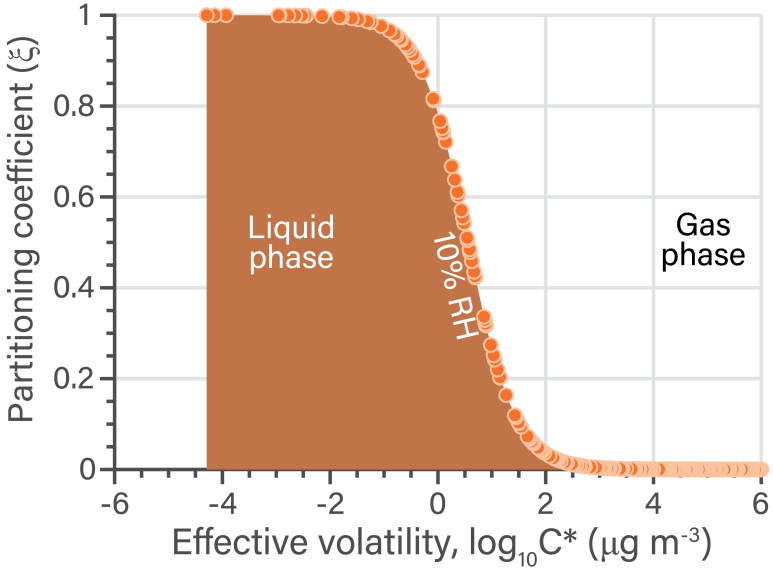


The neural network shortcut

- The combination of iterative solvers and NN can be accurate and efficient.
- Extensions of this methodology to other processes, is possible as long as the computation trade-off is favorable.
- Extensions to the BAT model will look at organic-organic and inorganic-organic equilibrium modeling.



Volatility is dependent on RH



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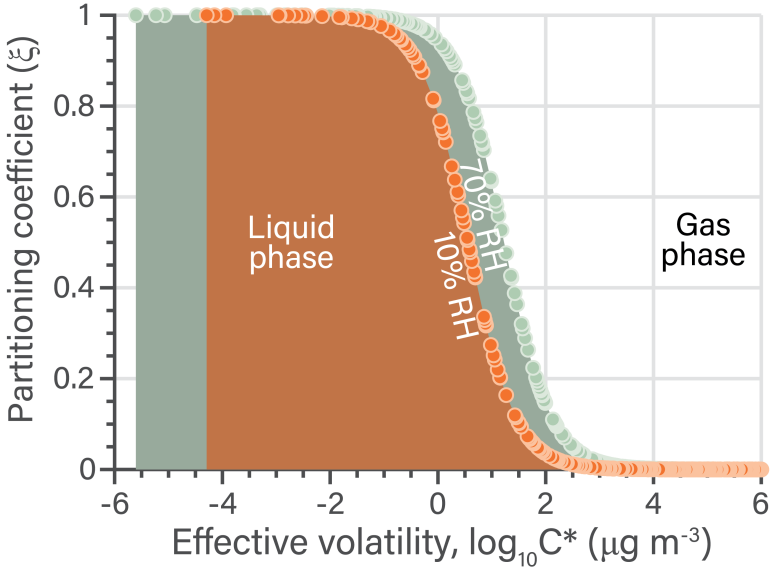

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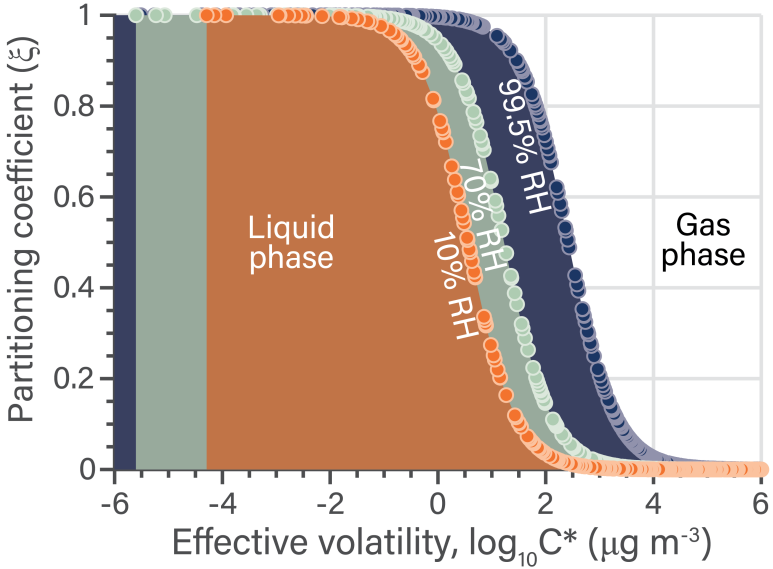
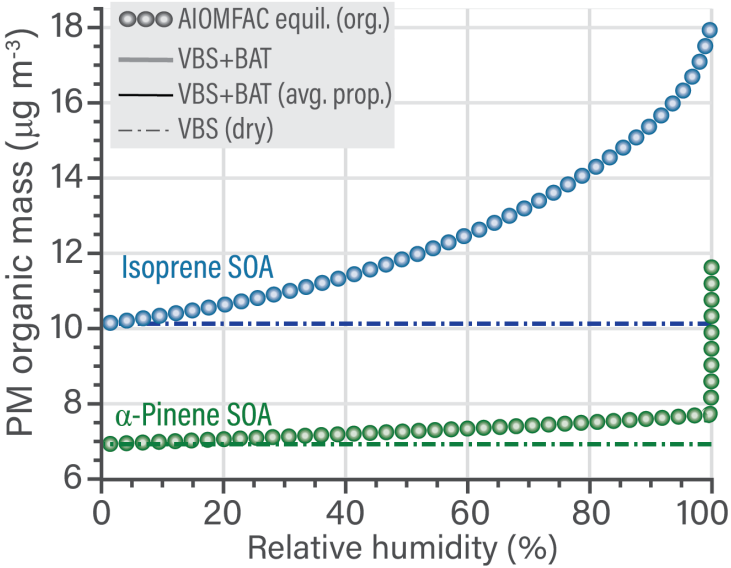


Organic mass shows good agreement



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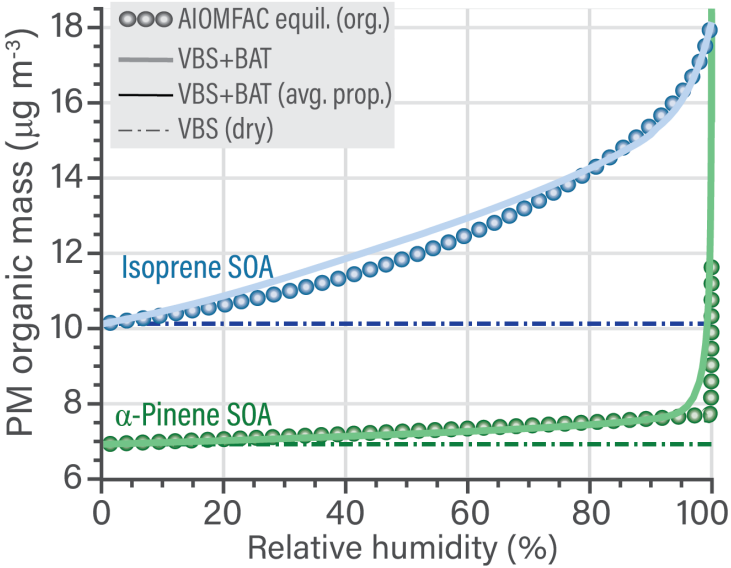

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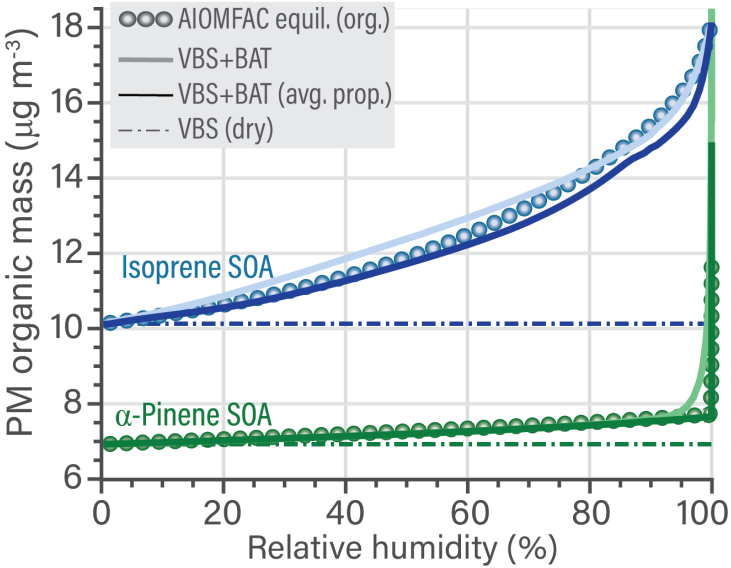

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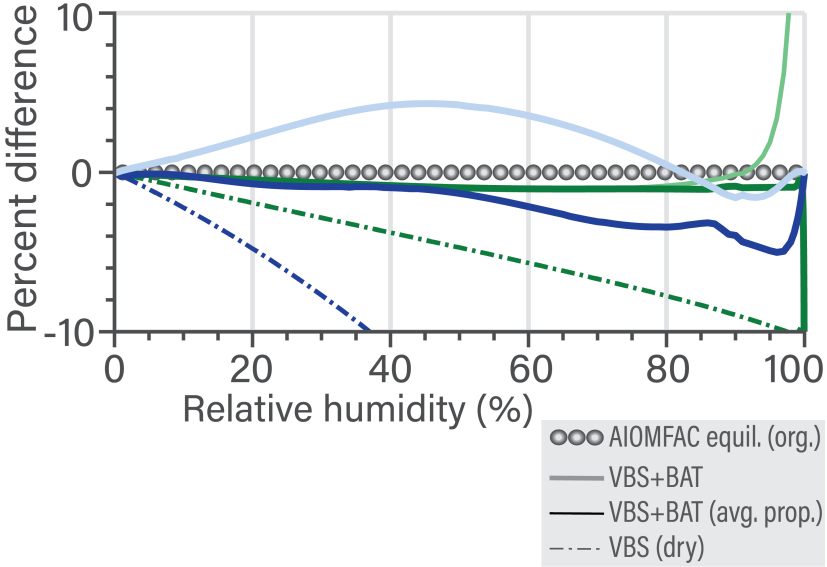
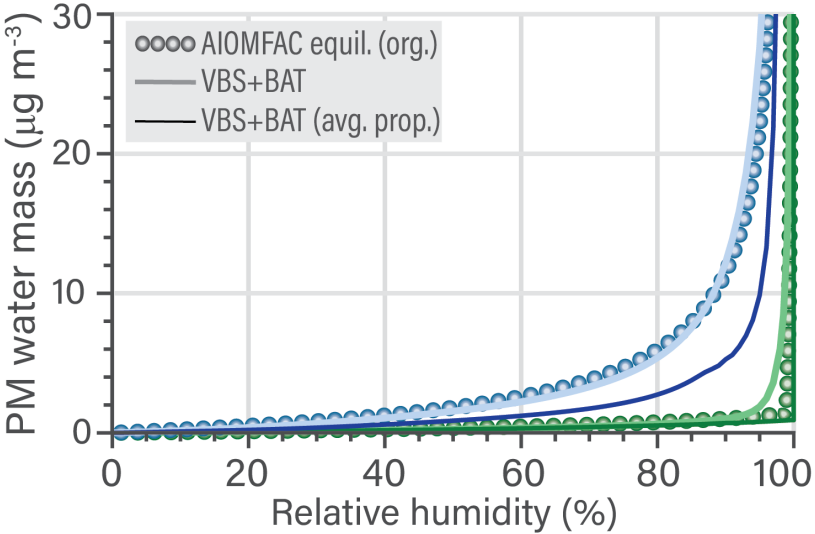
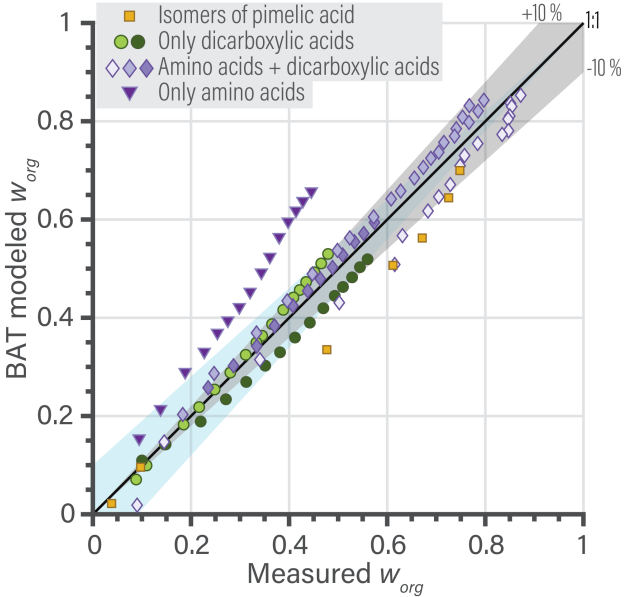


Water mass also agrees

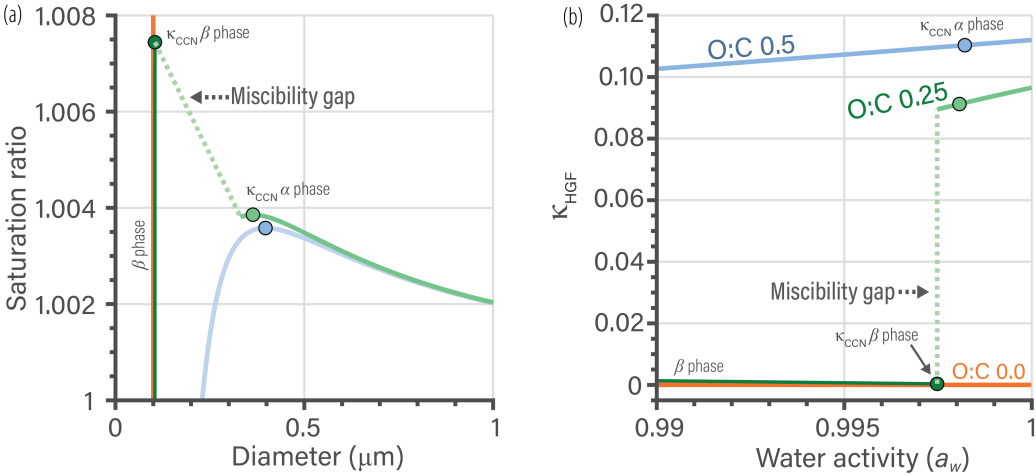


Measurement agreement of organic mass fractions



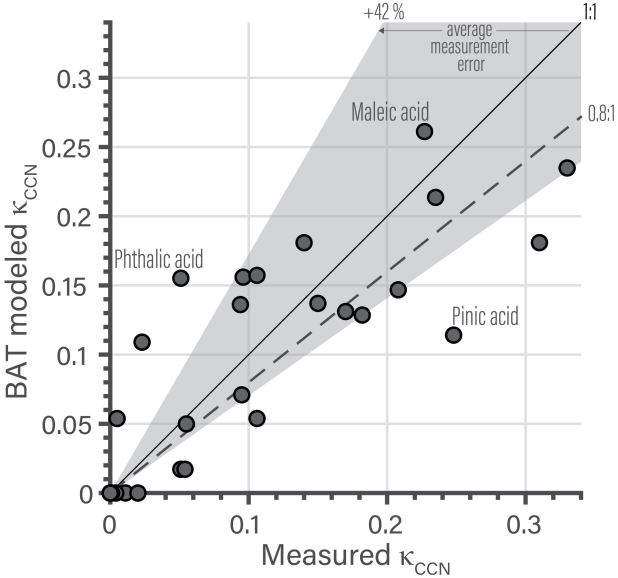
Side note on Köhler curves of CCN

$$\frac{1}{a_w} = 1 + \kappa_{HGF} \frac{V_{org,\,dry}}{V_w + V_{org} - V_{org,\,dry}}$$



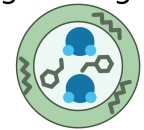
Measurement agreement: κ_{CCN}

- 30 comparison points
- Error of ± 0.055



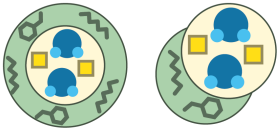
BAT sequels

Organic-organic interactions

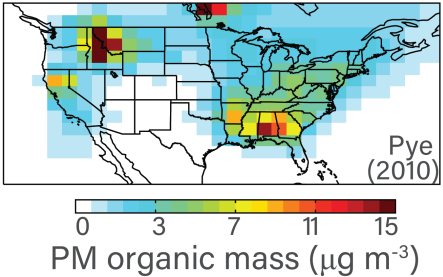


Inorganic-organic interactions

2-Phase systems



Camilo is pursuing the implementation into GEOS-Chem.



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