A method for testing the significance of novel aerosol processes

PyCHAM
CHemistry with Aerosol Microphysics in Python

An accelerated method for testing the significance of novel aerosol processes

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Novel aerosol processes

- Novel aerosol processes are formalised in mathematical form
- Evaluating new processes for significance in regional/global models often needs simplification due to three-dimensional (3D) model computational expense
- Accuracy may be lost, and substantial researcher time spent, in transferring the process to 3D models
- In Table 1 we present a method for accelerating the evaluation of processes in 3D models, whilst Figures 1-3 show associated results

Stage	Method	Example	Processing time
Formalisation	New process represented in mathematical form (this stage is the same as previous methods, but remaining stages accelerate)	Equations for reactions of peroxy radicals (RO_2) of highly oxidised molecules (HOMs) (HOM-RO_2) extended to existing photo-oxidation and autoxidation schemes (Jenkin et al. 1997, Roldin et al. 2019, Pichelstorfer et al. 2024)	Variable
0D Simulation	Implement process in a zero-dimensional (0D) model to estimate output(s) of interest w/ and w/o process across ambient conditions	Secondary organic particulate matter (SOPM) concentrations estimated w/ and w/o HOM-RO_2 reaction to give difference (ΔSOPM(w/-w/o)) using CHemistry with Aerosol Microphysics in Python (PyCHAM) model (O'Meara et al. 2021)	3 weeks
Parametrisation	Machine learning parametrises difference in output(s) w/ versus w/o process (as provided by 0D simulation)	Decision tree model created for difference in SOPM w/ and w/o HOM-RO_2 reaction as a function of potential parameters using Extreme Gradient Boosting (XGBoost) algorithm (Chen et al. 2016)	3 minutes
3D Application	Parametrisation applied to parameters estimated by 3D model for quantification of effect of omitting new process	Regional effect of omitting HOM-RO_2 interaction quantified by running the decision tree model with parameter values from the European Monitoring and Evaluation Programme (EMEP) model (Simpson et al. 2012) as input	1 day

Table 1: Outline of method, with acronyms defined in upper rows, column 'Example' describes the demonstration presented here, and 'Processing time' is computer processing time

Formalisation and 0D Simulation

- In our example, the base chemistry (i.e. w/o new process) used in 0D simulations was the photo-oxidation and autoxidation of α-pinene and benzene (references in Table 1)
- The new process in our example was the reaction between HOM-RO_2 (including cross-reactions between α-pinene and benzene RO_2), and the output of interest was the difference in SOPM mass concentration w/ vs. w/o HOM-RO_2 reaction (ΔSOPM(w/-w/o))
- Pairs of 0D simulations are run where a single pair has the same parameter values, but the new process under investigation is either turned on (w/ process) or off (w/o process), and multiple pairs cover the ambient range of parameter values, e.g. temperature
- 0D simulations are conducted in the manner of a continuous flow reactor, with results for our example in Fig. 1

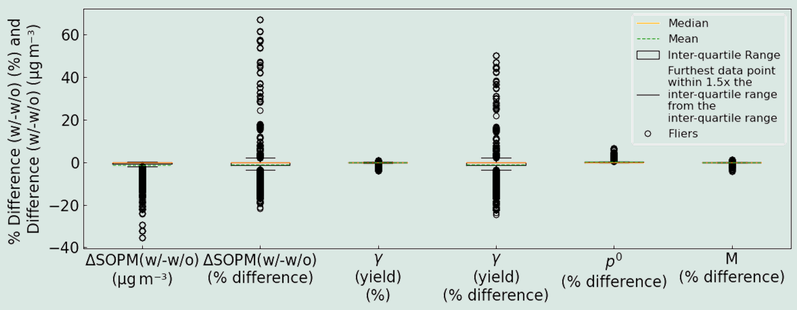

Figure 1: 0D simulation results for differences in SOPM mass, yield and the mass-weighted mean of condensable component vapour pressures and molar mass, where yield = SOPM formed (μg m^{-3})/(consumed α-pinene+benzene (μg m^{-3}))

Parametrisation and 3D Application

- 0D results are split 25:75 for testing:training the parametrisation from the machine learning algorithm
- E.g., decision tree model accuracy metrics are shown in Fig. 2, with a mean relative error of less than 10 % for relatively large values of ΔSOPM(w/-w/o)
- Parameter values from the 3D model are then applied to the parametrisation to quantify regional/global effect
- E.g., Fig. 3 indicates that omitting HOM-RO_2 reaction underestimates SOPM mass by around 5 % over large parts of Europe

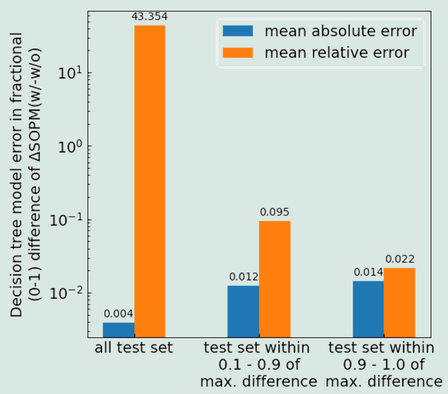

Figure 2: Accuracy of the decision tree model when compared against 0D model results

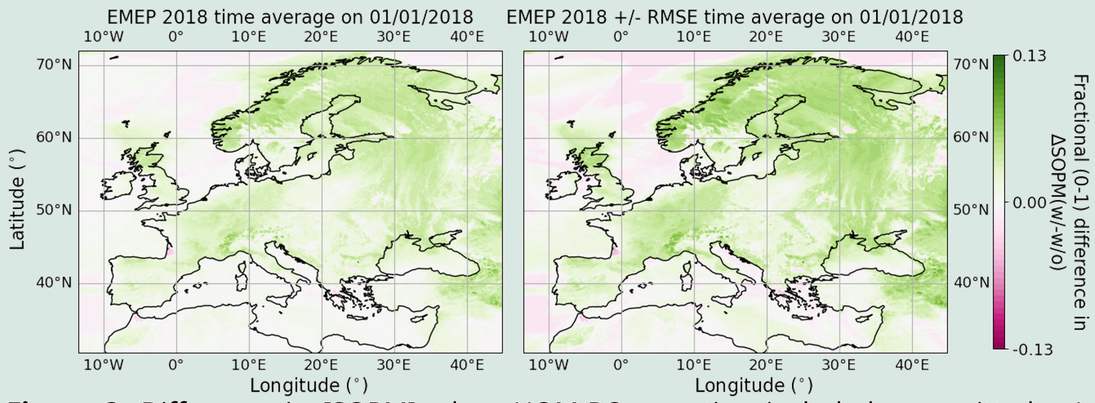

Figure 3: Difference in [SOPM] when HOM-RO_2 reaction included vs. omitted; +/- RMSE (root mean square error of the decision tree model) means added to positive differences and subtracted from negative differences (indicating effect of inaccuracy)

Conclusion

- A new method for evaluating the significance of processes at regional or global scale has been presented with the following characteristics: rapid transfer from detailed mathematical representation to 3D model, and acceptable accuracy of parametrisation
- Next step: how can we test the representativeness of the continuous flow reactor model setup for 3D model setups?

References: doi. Acknowledgements
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