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Toward Emulating an Explicit Organic Chemistry Mechanism With Random Forest Models

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Key Points:

- Random forests were trained to reproduce toluene and dodecane secondary organic aerosol formation calculated by an explicit organic chemistry mechanism
- The random forests performances are highly sensitive to the chemical regime
- There is an optimal number of predictors and their selection depends on the precursor

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Abstract Predicting secondary organic aerosol (SOA) formation relies either on extremely detailed, numerically expensive models accounting for the condensation of individual species or on extremely simplified, numerically affordable models parameterizing SOA formation for large-scale simulations. In this work, we explore the possibility of creating a random forest to reproduce the behavior of a detailed atmospheric organic chemistry model at a fraction of the numerical cost. A comprehensive data set was created based on thousands of individual detailed simulations, randomly initialized to account for the variety of atmospheric chemical environments. Recurrent random forests were trained to predict organic matter formation from dodecane and toluene precursors, and the partitioning between gas and particle phases. Validation tests show that the random forests perform well without any divergence over 10 days of simulations. The distribution of errors shows that the sampling of initial conditions for the training simulations needs to focus on chemical regimes where SOA production is the most sensitive. Sensitivity tests show that specializing multiple random forests for a specific chemical regime is not more efficient than training a single general random forest for the entire data set. The most important predictors are those providing information about the chemical regime, oxidants levels, and existing organic mass. The choice of predictors is crucial as using too many unimportant predictors reduces the performances of the random forests.

Plain Language Summary Organic compounds constitute a significant fraction of atmospheric particles and thus have an impact on health and climate. Predicting the contribution of organic compounds to atmospheric particles is extremely complex because of the very large number of different chemical species potentially condensing into the aerosol phase. Air quality and climate models usually rely on simplified, empirical approaches to predict organic aerosol mass concentrations, based on laboratory experiments. In this work, we apply a machine learning approach to construct a tool that behaves like the most detailed organic chemistry model, for a numerical cost affordable by air quality and climate models. Building upon this method, it will be possible to bring the complexities of organic chemistry to large-scale models.

1. Introduction

Secondary organic aerosol (SOA) constitutes a major fraction of atmospheric particles worldwide. It is composed of a multitude of organic compounds (e.g., Kourtchev et al., 2016). Our current understanding and modeling of SOA formation processes are highly uncertain (Pai et al., 2020) and involve representing the complex interplay between gas-phase oxidation and condensation of semi- and low-volatile organic species. SOA models need to include processes such as (a) the multi-step oxidation of the large variety of organic compounds emitted naturally and by human activities, (b) the condensation of semi-volatile species to the particle phase, and (c) the heterogeneous and in-particle reactivity of condensed species. This complexity can only be represented in models that explicitly account for aerosol physico-chemical processes. In these so-called explicit models, the aim is to represent the fate of each individual chemical species through individual reactions, which can number in the 10^9 range (e.g., Aumont et al., 2005). The Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere (GECKO-A, Aumont et al., 2005) is an example of such a model able to generate chemical mechanisms that explicitly describe the oxidation of organic compounds in the atmosphere, as well as their condensation into the particle phase (Camredon et al., 2007). It has previously been used to study SOA formation in various settings such as atmospheric chamber experiments (La et al., 2016), sensitivity studies (Aumont et al., 2012; Hodzic et al., 2015; Valorso et al., 2011) and urban plume modeling (Lee-Taylor et al., 2015; Mouchel-Vallon et al., 2020).

Because of their size, explicit mechanisms like those generated by GECKO-A cannot be used in 3D air quality models, which rely on empirical SOA parameterizations. The volatility basis set (VBS, Donahue et al., 2006) and its derivatives (e.g., Cappa & Wilson, 2012; Donahue et al., 2011) are the most prevalent representations of SOA chemistry in this field. In such models, simplifications are made to represent SOA formation by grouping organic species of similar properties into discretized bins, that is, volatility or oxidation state are the typically chosen properties for the VBS bins. This approach has been presented by Pankow et al. (2015) as the *anonymized* view of SOA modeling, as opposed to the *molecular* view of SOA modeling used by explicit models.

Previous attempts have been made to bring the molecular view of SOA modeling to 3D models. Li et al. (2015) included the near explicit Master Chemical Mechanism (MCM v3.2, Saunders et al., 2003; Jenkin et al., 2003), in the Community Multiscale Air Quality model (CMAQ, Foley et al., 2010). MCM is a *near* explicit mechanism, as some simplifications are made to simplify its development such as removing unlikely reaction channels and simplifying the oxidation of minor or unknown products. At the time MCM therefore used approximately 17,000 reactions involving approximately 6,000 species to represent the progressive oxidation of 142 primary hydrocarbons. Although the implementation of MCM in CMAQ was able to reproduce reasonably well the observed SOA surface concentrations over eastern US for a case study, this approach did not have further applications to our knowledge, and was limited by the considerable computational cost required to run regional scale simulations.

Lannuque et al. (2018) created VBS-GECKO, an empirical VBS parameterization where the stoichiometric coefficients were optimized to fit data produced from GECKO-A runs instead of being fitted to empirical data. Their method had the advantage of parameterizing the model over the multi-day simulated aging of SOA, which cannot be obtained from the shorter chamber studies used to derive traditional VBS parameterizations. Lannuque et al. (2020) ran VBS-GECKO in an air quality model (Menut et al., 2013) and showed that VBS-GECKO was producing more SOA in the summer over Europe compared to the traditional SOA parameterization that is based on laboratory data (Couvidat et al., 2012). Because, in essence, their resulting model was a linear combination of multiple VBS produced for different levels of pollution, it relied on the assumption that atmospheric chemistry behaves linearly between the selected chemical regimes. As a result, VBS-GECKO may have been applied outside of its application domain.

Here, we propose to use a machine learning (ML) approach to bring the molecular view to 3D chemistry-climate models across a range of chemical regimes representative of tropospheric conditions. ML techniques have been applied previously for air quality forecasts (Liao et al., 2020) demonstrating that it is possible to run a trained artificial intelligence in a 3D model. Keller and Evans (2019) used the GEOS-Chem chemical mechanism solver to train multiple random forests that were then able to emulate the chemical solver behavior in that same model for various pollutant, for a fraction of the computational cost of the default GEOS-Chem model. Kelp et al. (2020) improved on this method by using a unique neural network to predict 20 chemical species. They implemented it in GEOS-Chem (Kelp et al., 2022), achieving stable 1-year simulations for ozone prediction with less than 10% bias compared to the reference and reducing computational times by a factor of five. The motivation of these previous studies stems from reducing the costs of calculating chemistry, that is usually taking from 50% to 90% of the computational costs of running global chemistry models such as GEOS-Chem (Keller & Evans, 2019).

Schreck et al. (2022) recently presented a neural network approach to emulate the behavior of idealized GECKO-A simulations for the SOA formation following the oxidation of three individual precursors reacting with OH under varied environmental conditions. While this work showed the ability of neural networks to reproduce idealized oxidation situations, it showed their limitations when extrapolated to realistic simulations with diurnally varying conditions. The results indicate that this type of system needs to be trained with a data set representative of the conditions in which it will be applied.

In this paper, we train a random forest on a data set constructed with multiple GECKO-A simulations, with the primary aim of predicting SOA mass from the oxidation of toluene and dodecane for realistic atmospheric conditions over a range of chemical regimes covering daytime and nighttime oxidation by the main oxidants (OH, O_3, and NO_3). Our objective is to build an empirical SOA model that is able to reproduce the aerosol mass that a complex, explicit mechanism would predict, at a numerical cost that is comparable to that of reduced chemical mechanisms currently used in large-scale models.

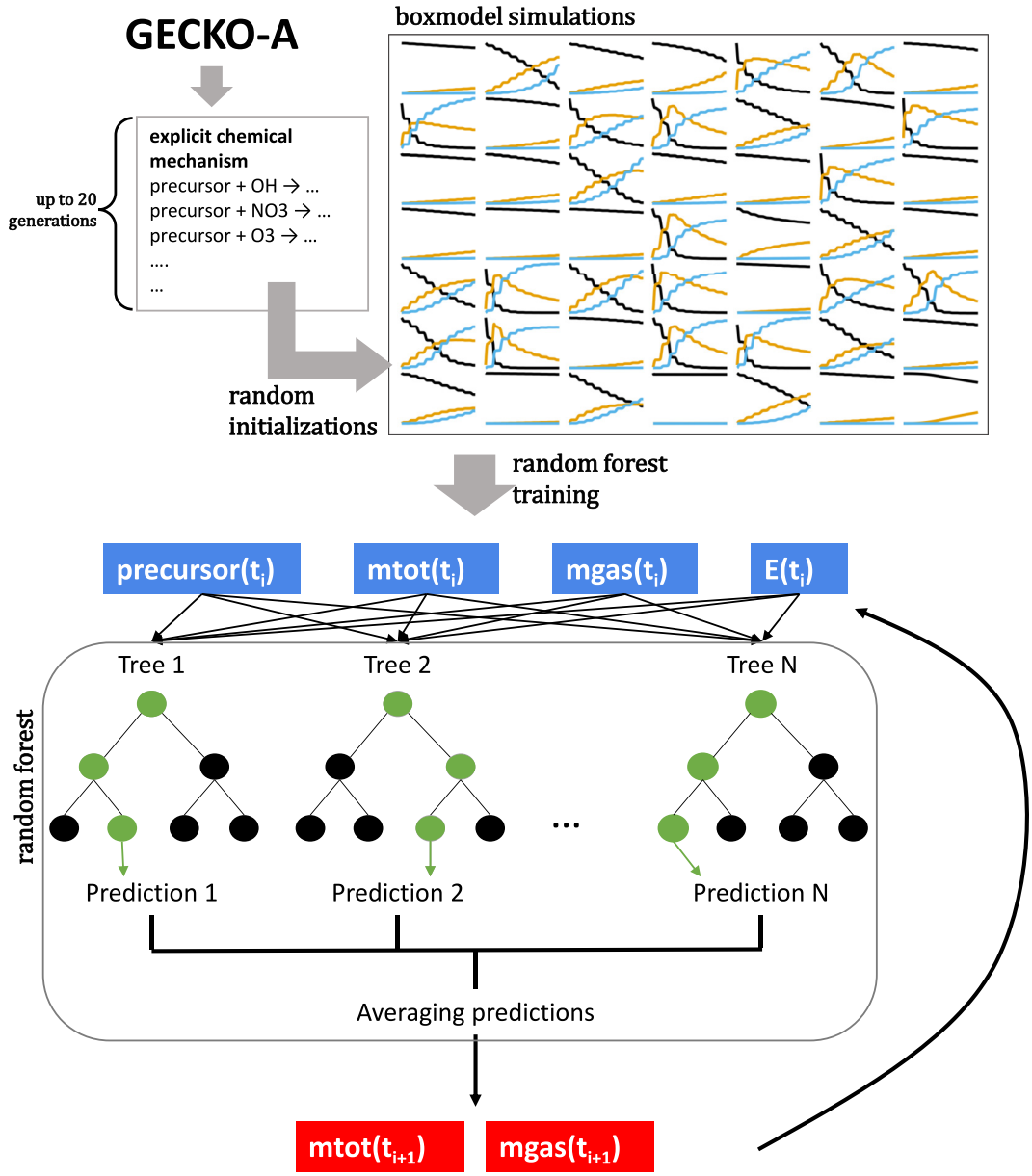

Figure 1. Schematic depiction of the data set construction based on Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere explicit modeling simulations (top) used to train the random forests depicted on the bottom. The variables meaning (mtot and mgas) are indicated in Table 2. $E(t_i)$ is lumping all environmental conditions (see Table 2) at time t_i.

2. Methods

2.1. Reference Organic Chemistry Mechanisms

GECKO-A (Aumont et al., 2005) is a software tool allowing the automatic generation of detailed multi-generational organic chemistry mechanisms (Figure 1). It is based on state-of-the-art knowledge of atmospheric organic chemistry and structure activity relationships (e.g., Atkinson, 1997; Raventos-Duran et al., 2010) to estimate unknown reaction kinetics and thermodynamics. It calculates the gas-particle partitioning of individual organic species based on estimates of their volatility (Valorso et al., 2011). In the present study, chemical mechanisms were generated for the oxidation by OH, NO_3, and O_3 of toluene (C_7H_8) and dodecane ($C_{12}H_{26}$), two compounds emitted by anthropogenic activities. Currently, GECKO-A only includes gas-phase oxidation and condensation of semi-volatile organic compounds. There are no heterogeneous processes and no aerosol phase processes (e.g.,

Table 1
Environmental and Chemistry Parameters Used for Generating the Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere Box-Model Data Set

Parameter	Range
Latitude (°)	−80–80
Temperature (K)	216–313
Preexisting aerosol seed (μg m^{-3})	0.03–340
Initial precursor (ppb)	0–16
Initial O_3 (ppb)	1–100
Relative humidity (%)	3–102
Atmospheric pressure (atm)	0.5–1.02
Initial NO_x (ppb)[a]	10^{-4}–42
Initial CO (ppb)	33–1,012
NO Emission (molec cm^{-2} s^{-1})	10^7–10^9

[a]Sampled in log space.

oligomerization) included in the model. The resulting toluene full oxidation mechanism contains 8,560 species involved in 47,349 chemical reactions, including 4,536 gas-aerosol equilibrium reactions. The oxidation mechanism for dodecane was completed to the fourth generation. This was done in order to reduce the generated mechanism to a manageable size for the purpose of this work. The resulting dodecane oxidation mechanism contains 75,745 species involved in 465,751 chemical reactions, including 20,968 gas-aerosol equilibrium reactions.

2.2. Data Set Construction

To create the data set used to train and evaluate the random forest model, we ran a large set of simulations for toluene and dodecane (see Figure 1). Each simulation is performed for 230 hr with a uniformly sampled set of randomly chosen initial conditions and external forcing (see Table 1). Temperature, relative humidity, and atmospheric pressure are selected in ranges typical of values found in the lower troposphere. This ensures that the SOA emulator will have the correct sensitivity to changes in these parameters through the effects of (a) temperature on reaction rates and SOA evaporation, (b) relative humidity on OH formation, and (c) pressure on third-body reaction rates. Initial concentrations of precursors, NO_x and CO are randomly picked to cover a wide range of chemical regimes. All model simulations start at 10:00 a.m. UTC and simulate a diurnal light cycle defined by the chosen latitude. The latitude is varied from 80°S to 80°N to ensure that the model does not fit to a specific diurnal cycle. The model time-step length is 5 min. After initialization, the precursor, NO_x, O_3, and CO freely react without constraints. The other external forcings (temperature, relative humidity, pressure, latitude, NO emissions, and seed) are maintained constant for the whole simulation. Holding these external forcings constant in the data set in not expected to bias the random forests results because the high number of different simulations still covers a wide range of environmental conditions. The simulated photochemistry leads to the multi-generational formation of semi- and low-volatile secondary organic compounds that can condense to form SOA.

Table 2
List of Predictors and Outcomes Used for Training the Random Forests

Predictors (units)	Outcomes (units) (prediction method)
Temperature (K)	Total organic mass (μg m^{-3}) (direct)
Water vapor concentration (H_2O) (molec/cm^3)	Organic gaseous fraction (dimensionless) (trend)
Pressure (atm)	
Solar zenith angle (deg)	
J_{NO_2} (s^{-1})	
NO (molec/cm^3)	
NO_2 (molec/cm^3)	
O_3 (molec/cm^3)	
OH (molec/cm^3)	
H_2O_2 (molec/cm^3)	
CH_2O (molec/cm^3)	
Aerosol seed mass (μg m^{-3})	
Total organic mass (mtot) (μg m^{-3})	
Organic gaseous fraction (mgas) (dimensionless)	
Organic aerosol fraction (maer) (dimensionless)	
Precursor (molec/cm^3)	

Note. The short names used in Figure 9 are mentioned in round brackets.

2.2.1. Outcomes

In this work, the aim is to predict the distribution of organic species between gas and aerosol phases. In order to build a flexible approach that will allow future developments such as adding additional phases (e.g., aqueous phase) and predicting organic matters properties (e.g., solubility for deposition of organic vapors and particles), the first chosen outcome is the total organic mass m_t (μg m^{-3}):

$$m_t = m_g + m_a \quad (1)$$

m_g and m_a are respectively the total gas- and particle-phase organic mass. The goal is to have only one outcome m_t in mass concentration units (μg m^{-3}) and predict the contributing phases to the total mass as fractions of this m_t. We arbitrarily chose to predict gaseous mass fraction γ ($m_g = \gamma \times m_t$). m_a can then be derived as $m_a = (1 - \gamma) \times m_t$.

Following the method of Keller and Evans (2019), we also established a variance criterion to decide whether the random forest should predict the value of the predictor or its trend. For stability and better performances, this variance criterion is used to identify stable and unstable outcomes. This classification is based on the standard deviation of the ratio between post- and pre-numerical time-step solve value. This ratio was calculated for each outcome and each time-step on the whole training data set. The standard deviation of these ratios was calculated and if the value of this standard deviation is below a threshold of 0.07, the outcome is classified as stable and its trend is predicted. Otherwise, the outcome is unstable and its direct value is predicted. For the two outcomes used in this work, we found that the value of the total mass outcome m_t is unstable and needs to be directly predicted, while the gas phase fraction γ is stable and its trend is predicted.

2.2.2. Predictors

We selected the predictors based on parameters relevant to SOA formation (see Table 2). The concentration of the precursor as well as the main daytime oxidants (OH, O_3) and the aerosol seed concentrations have been chosen for their key role in SOA formation. The pressure and temperature modulate the kinetics that control gas-phase oxidation. Temperature is also very important for the condensation of vapors. The solar zenith angle and the photolysis rate of NO_2 represent the influence of the diurnal cycle on gas kinetics.

At each time-step of the GECKO-A simulations, predictors are computed before chemistry is integrated, and outcomes are computed after the solver has finished. Four Thousand simulations ($\approx 10^6$ time-steps) where produced for each precursor, resulting in a total of 8,000 simulations ($\approx 2 \times 10^6$ time-steps).

The collection of all predictor and outcome values at every time-step therefore constitutes a data set representative of what the integration of an explicit chemical scheme would produce for a given predictor over one time-step.

2.3. Random Forest Regression

We use Python libraries *scikit-learn* (Pedregosa et al., 2011) to fit the random forests, and *dask* (Dask Development Team, 2016) to handle parallelization of the code on the NCAR CISL supercomputers (Computational and Information Systems Laboratory, 2017). The simulations were randomly split between training (80% of all simulations) and validation (20%) sets. The analysis shown in Section 3 was performed on the validation data set.

Keller and Evans (2019) trained individual random forests for each of their outcomes, considering all chemical tracers as predictors. They integrated the random forest models within GEOS-Chem at each model time-step. In contrast, our approach involved training individual random forests for each SOA precursor to predict all outcomes simultaneously (total organic mass and organic gaseous fraction). This means that each tree predicts a vector of values instead of predicting single values (see Figure 1). This approach has the advantage of simplifying the model training and evaluation, and its future implementation in 3D models. As the data set contains values spanning many orders of magnitude, it was successively log-transformed, power-transformed and normalized to map the data as close as possible to a Gaussian distribution. For simplicity, this sequence of transformations was applied to all variables, including those that are almost Gaussian. The training data set was shuffled prior to the regression procedure to avoid bias related to the random forest learning a specific diurnal cycle.

Hyperparameters for the random forest were tuned automatically during the random forest training with the *scikit-learn* library. The number of decision trees is the most important hyperparameter because it impacts both the numerical cost of running the random forest as well as the quality of the random forest. Random forest's

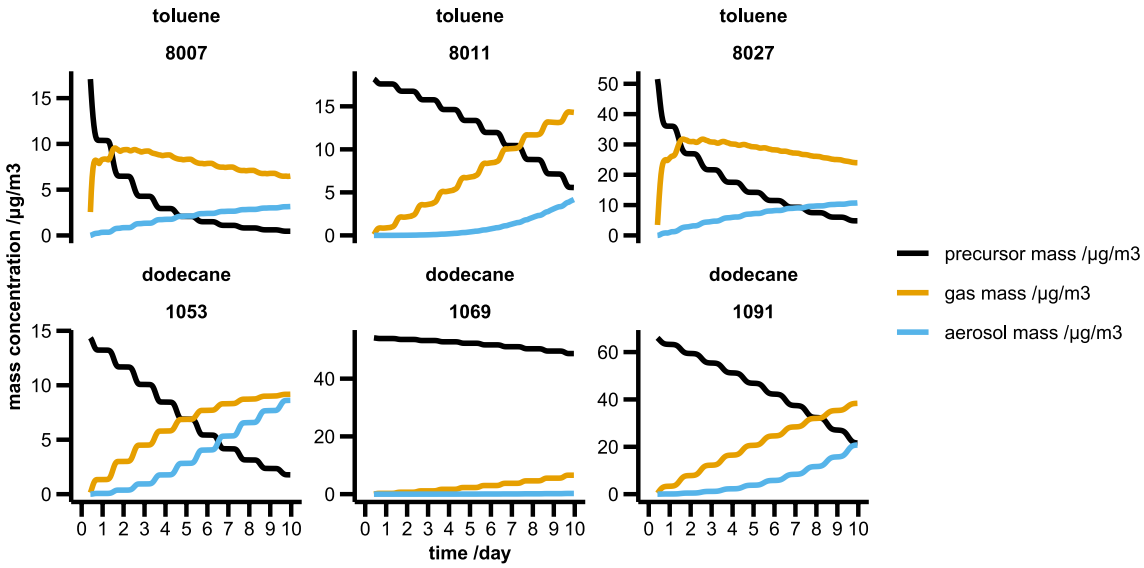

Figure 2. Time evolution of precursor oxidation, organic gas, and organic aerosol formation from toluene and dodecane oxidation for representative training simulations.

training configurations were tested with 10, 50, 75, and 100 decision trees. The random forest hyperparameters optimization consistently selected 50 trees, which is the same number of decision trees that were selected in Keller and Evans (2019).

3. Results

3.1. Training Data Set Characterization

Figure 2 depicts the typical time evolution of a few randomly selected simulations from the validation data set. As expected, the precursor is progressively oxidized during the 10 days of the simulation. The kinetics of this decay depends on the concentration of the precursors' oxidants: OH (daytime) and NO_3 (nighttime) for dodecane, OH, O_3 (daytime), and NO_3 (nighttime) for toluene. Given that concentrations of these oxidants depend on randomly selected initial and environmental conditions in each simulation, the decay kinetics vary for each simulation. The oxidation of the precursor leads to the progressive formation of gaseous organic compounds. Depending on the availability of oxidants, the formation of these secondary organic compounds can peak early in the simulation as in simulations 8,007 and 8,027 in Figure 2. On the other hand, the evolution displays a characteristic stepwise diurnal profile, with the organic mass increasing during daytime when photochemistry can take place. After the peak of the quicker oxidation simulations, the total organic mass decreases because the oxidation products are ultimately lost to the terminal CO_2 formation step. As their oxidation progresses, the secondary gaseous compounds become more oxidized and are able to condense onto the pre-existing aerosol seed, forming SOA. SOA formation therefore highly depends on the availability of oxidants. For instance, simulation 1,069 displays a typical case of a slow precursor decay causing the slow formation of secondary organic compounds with almost no SOA production.

As shown in Figure 2 on a few sample simulations, this work is aimed at reproducing a large variety of situations, with the SOA formation behavior that non-linearly depends on multiple parameters. Figure 3 depicts the distribution of SOA mass yield as a function of key parameters describing the chemical regimes controlling SOA formation.

The precursor controls the total amount of organic carbon that is available to form SOA. Both dodecane and toluene SOA yields are not constant as a function of the precursor concentration. This is a typical illustration of the non-linearity of SOA formation and atmospheric chemistry in general. As precursor mixing ratios increase, the precursor becomes a significant competitor for oxidants, slowing the formation of later generations organic compounds that are more likely to efficiently contribute to SOA. This hypothesis is confirmed by the fact that

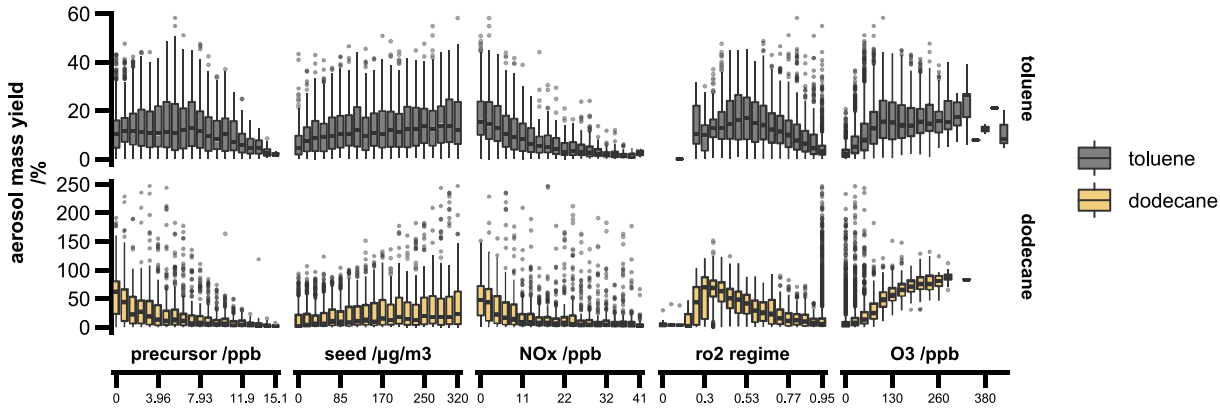

Figure 3. Dependence of average organic aerosol mass and aerosol mass yield as a function of the chemical environment.

the dodecane SOA yield decay starts for lower precursor mixing ratios, which is explained by the faster reaction rate of dodecane with the main oxidant OH compared to toluene ($k_{\text{dodecane + OH,298}} = 1.32 \times 10^{-11}$ cm^3/molec/s vs. $k_{\text{toluene +OH,298}} = 5.6 \times 10^{-12}$ cm^3/molec/s, Mellouki et al., 2021).

As the seed concentrations were selected over a wide range (see Table 1), the limiting effect of low pre-existing seeds can only be seen in the lowest chosen concentrations, below 40 µg m^{-3}. In the rest of the range of seed concentrations, SOA yield has identical distribution probability. In our simulations, as the nature of this seed is not accounted for, the fact that seed is present is enough to trigger SOA condensation and it is rarely limiting.

The NO_x mixing ratios control the formation of ozone and OH through the photolysis of NO_2 and the reaction of ozone with NO. However the relationship of NO_x with oxidants levels is not trivial and also depends on the concentrations of organic compounds. Here, the simulated higher SOA yields at lower NO_x levels could be explained by the role of NO_x on the oxidation of organic compounds. After the initial oxidation step forming a peroxy radical ($RH + OH \xrightarrow{+O_2} RO_2 + H_2O$), the peroxy radical can react with NO to form an alkoxy radical that can fragment, leading to more volatile compounds that are less likely to form SOA. If NO concentration is low enough, peroxy radicals are more likely to react with HO_2 and other peroxy radicals to form more oxidized species that are more likely to form SOA. This can explain the higher SOA yields at lower NO_x shown on Figure 3. This effect is better seen after defining the RO_2 regime β as:

$$\beta = \frac{k_{RO_2+NO} \times NO}{k_{RO_2+NO} \times NO + k_{RO_2+HO_2} \times HO_2} \quad (2)$$

where $k_{RO_2+NO} = 7.7 \times 10^{-12}$cm^{3}molec^{-1}s^{-1}, $k_{RO_2+HO_2} = 5.1 \times 10^{-12}$cm^{3}molec^{-1}s^{-1}. This ratio indicates which pathway is favored for RO_2 radicals: when $\beta = 1$, they only react with NO and when $\beta = 0$ they never react with NO. For dodecane, Figure 3 shows that from $\beta = 0.4$ to $\beta = 1$, the median value of the SOA yield decreases from 60% to less than 1%. Below $\beta = 0.4$, the SOA yield decreases down to less than 1% at $\beta = 0.15$. This low yield for low β values can be explained by low levels of oxidants limiting SOA production in very low NO_x situations. For toluene, this peak in SOA yield happens around $\beta = 0.5$ with a median value of 19%. It is less marked than for dodecane because the impact of the $RO_2 + NO$ reaction pathway on fragmentation is lower on cyclic and shorter molecules like toluene and its oxidation products (Aumont et al., 2013).

For both precursors, the SOA yield increases with ozone mixing ratios. Higher ozone mixing ratios are associated with higher OH concentrations, which can explain higher SOA yields.

3.2. Training Data Set Size Impact

For this sensitivity test, random forests were trained for each predictor, with a limit on the number of points taken from the data set to train the random forest (50,000, 100,000, 500,000, and 1,000,000 points). Two kinds of tests are presented for each validation simulation. First, the random forest was tested on each time-step individually by using the reference predictors as inputs and comparing the random forest output with the reference outcomes.

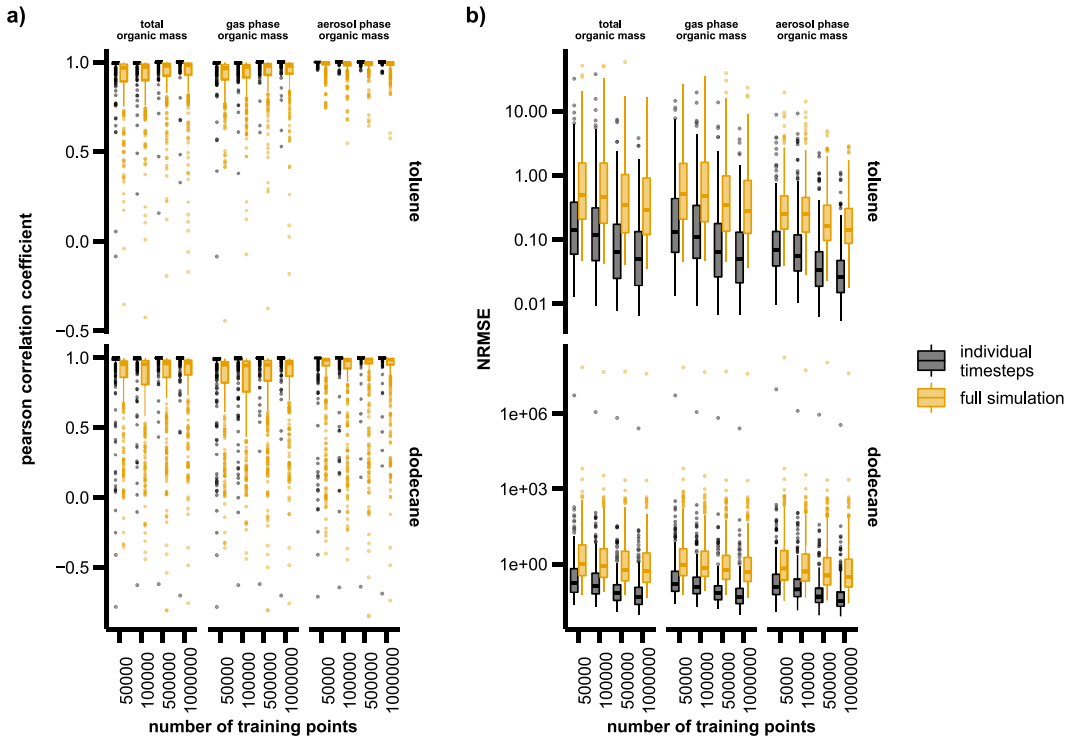

Figure 4. Boxplot distribution of (a) Pearson correlation coefficients and (b) normalized root mean square error (note the *y*-axis log-scale) for testing individual time-step predictions (black) and full simulation runs (yellow) for toluene and dodecane simulations. The number of points that were used to train each random forest are shown on the *x*-axis. The middle lines of the boxplots are the median, the top and bottom of the boxes denote the first and third quartiles and the whiskers extend to the 5th and 95th percentiles of the distribution.

To evaluate the performance of the random forests, the Pearson correlation coefficient (r) was used to depict the ability of the random forests to reproduce trends in SOA formation and destruction. The normalized root mean square error (NRMSE) was also used:

$$\text{NRMSE} = \frac{\sqrt{\frac{\sum_{i=1}^{N}(y_i - \hat{y}_i)^2}{N}}}{Q_3 - Q_1} \quad (3)$$

y_i and $\hat{y}_i$ are the ith GECKO-A reference and predicted aerosol mass respectively, N is the number of time-steps and $Q_3 - Q_1$ is the difference between the first and the third quartiles. NRMSE values illustrate the ability of the random forests to simulation SOA mass without bias.

The resulting Pearson correlation coefficients (r) and NRMSE distributions are shown on Figure 4 (black boxplots), as a function of the number of points used for training the random forest. With the exception of a few outliers, the random forest is very accurate to predict outcomes on a single time-step, even with only 50,000 training points. The Pearson correlation coefficient displays median values around $r = 0.99$ for both toluene and dodecane while the NRMSE displays median values below 0.1 for toluene and 0.15 for dodecane.

For the second test in Figure 4 (orange boxplots), as well as the rest of this paper, the random forest is constrained with the initial conditions and the environmental conditions from the reference simulation. At the end of each time-step, the predicted outcomes are used in the input predictors for the next time-step. In these validation tests, the random forest model accumulates errors with time. This is reflected in the median r values decreasing compared to individual time-steps tests to approximately $r = 0.98$ for toluene and $r = 0.96$ for dodecane. The performances are also more spread with the interquartile range for 100,000 points increasing from 0.002 to 0.05 for toluene and from 0.003 to 0.09 for dodecane. Conversely, for 100,000 training points, NRMSE values increase to approximately 0.3 for toluene and 0.5 for dodecane. The NRMSE interquartile range for 100,000

points increases from 0.09 to 0.3 for toluene and from 0.2 to 2 for dodecane. Increasing the number of points used to train the random forest slightly improve the r scores. For toluene, using 500,000 or 1,000,000 points provides similar performances while for dodecane, using 1,000,000 points still increases r and reduces the interquartile range. The median NRMSE decreases slightly when increasing the number of training points, from 0.3 to 0.2 for toluene and from 3 to 1 for dodecane. We could not test higher numbers of training points due to the limited size of the created data set, but it seems that above 500,000 training points, the gains are marginal at best for the considered precursors.

3.3. Sample Simulations Tests

To illustrate the behavior of the random forest model, Figure 5 displays the random forest results on the same sample simulations that were shown on Figure 2. The associated relative errors on predicted aerosol mass are shown on Figure 6. The random forest is able to reproduce the timeseries of gas and aerosol mass in all but one of the examples (simulation 1,053). For these simulations, the random forest can reproduce the typical stepwise daytime growth of organic mass of the slower oxidation simulations (simulation 1,069, 1,091, and 8,011), as well as reproducing the peaking growth of organic mass for faster oxidation simulations (8,007 and 8,027). For all simulations (except 1,053), the relative error tends to be the highest for the first 5 days of the simulations, converging toward errors lower than 10% for the last 5 days. Finally, for the worst random forest simulation in this sample (1,053), the model exhibits errors around ±100% after 2 days and cannot recover from the accumulated errors. The relative error remains between 50% and 100% but not producing any unrealistic mass concentrations.

It is clear that the computational resources required to run the random forest emulators are much lower than those needed for full GECKO-A model runs. However, it is difficult to perform a fair comparison because the GECKO-A box model is parallelized and coded in Fortran, while the random forests were tested with a simple, non parallelized Python code. Such a comparison between GECKO-A and neural networks was carried out by Schreck et al. (2022), showing that runtime was decreased by a factor of 300 for toluene, and a factor of 22,000 for dodecane simulations. These factors are increased by one or two additional orders of magnitude when the neural networks are run on GPUs, but the comparison is unfair to the CPU bound GECKO-A code. We however can still expect that the random forests trained in this work would exhibit similar performance improvements, but further investigations are needed.

3.4. Errors Distribution

In order to identify the type of situations where the random forest is not able to reproduce the explicit model behavior, we examine the distribution of the NRMSE defined in Equation 3. The validations random forest simulations were split in four categories, depending on their NRMSE. The distributions of environmental conditions according to this split are displayed on Figure 7.

First the existing seed concentration distribution does not vary with the quality of the random forest simulations. As was shown above on Figure 3, the aerosol mass yields dependence on seed concentration is low, which explains why the random forest performances are not sensitive to this variable. As long as some seed aerosol is available, the system is not sensitive to its mass concentrations.

For toluene, the lower quality random forest simulations (third and fourth NRMSE quartiles) are typically described with lower NO_x regimes: on average $\beta \approx 0.82$ for the first two NRMSE quartiles, compared to $\beta = 0.7$ and $\beta = 0.65$ for the last two NRMSE quartiles. The third and fourth NRMSE quartiles simulations also exhibit higher OH mixing ratios: $OH_{median}^{3^{rd}quartile} = 0.04$ ppt and $OH_{median}^{4^{th}quartile} = 0.07$ ppt compared to $OH_{median}^{1^{st}quartile} = 0.01$ ppt for the first quartile. The aerosol mass yields (≈12%–13%) are similar for all quartiles. The non-linear dependence of the SOA yield on the RO_2 regime (Figure 3) seems to be the determining factor for toluene simulations. Under-representing lower RO_2 regimes in the training data set therefore has a strong impact on the random forest performances. In our case, this under-representation is the likely the consequence of the simple random selection of the training simulations environmental conditions.

Dodecane lower quality simulations are heavily skewed toward simulations with high NO_x regimes ($\beta_{median}^{4^{th}quartile} = 1$ vs. $\beta_{median}^{1^{st}quartile} = 0.87$) and lower OH mixing ratio ($OH_{median}^{4^{th}quartile} = 8.8 \times 10^{-5}$ ppt vs. $OH_{median}^{1^{st}quartile} = 3.7 \times 10^{-3}$ ppt) and lower aerosol mass yields ($Y_{median}^{4^{th}quartile} = 0.68\%$ vs. $Y_{median}^{1^{st}quartile} = 8.1\%$). This behavior difference between

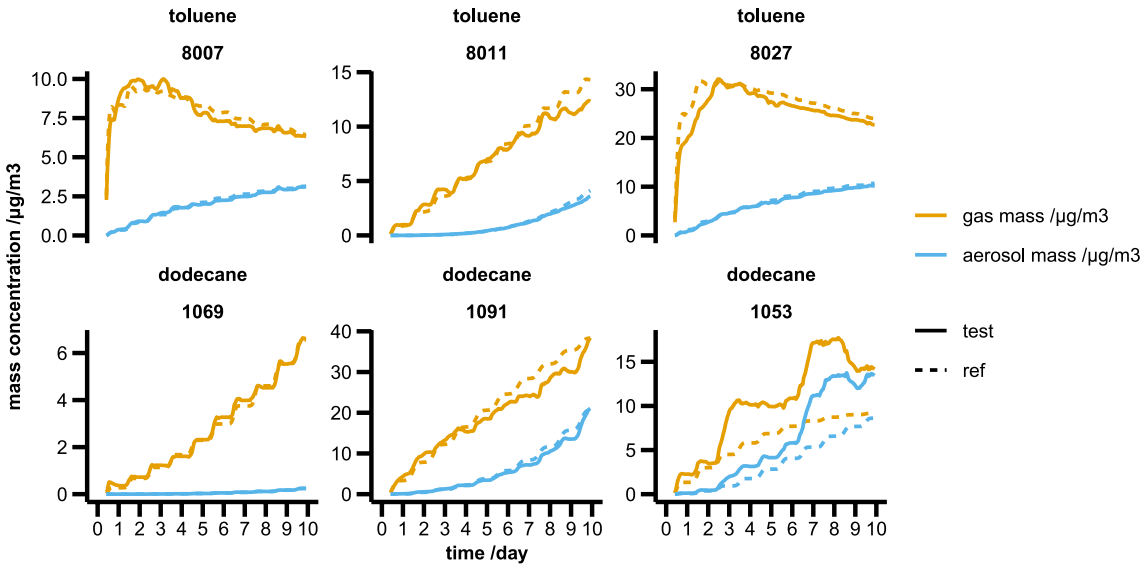

Figure 5. Time-series of three toluene and three dodecane sample experiments, comparing the Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere reference simulations (dashed lines) and the random forest simulations (continuous lines) for the predicted organic gas (black) and aerosol (orange) mass.

toluene and dodecane may be explained by the number of dodecane training simulations available for higher NO_x regimes, which include many outliers (1,236 outliers out of 2,568 point for the highest RO_2 regime bin on Figure 3) in terms of aerosol yield. The random forest is therefore not able to reproduce the complex behavior of dodecane SOA formation in this specific regime. In this case, the complexity of the dodecane oxidation for very high NO_x situations cannot be properly reproduced by the random forest with the given training data set.

A possible way to improve the ability of the system to reproduce the complex relationship between β and SOA formation is to create independent random forests specialized for specific RO_2 regimes. To test this hypothesis, the training data set was split in three separate sets according to the initial RO_2 regimes: a low NO_x set ($\beta < 0.3$, 100,280 training points for toluene, 72,200 points for dodecane), a mid NO_x set ($0.3 < \beta < 0.7$, 594,780 points for toluene, 268,640 points for dodecane) and a high NO_x set ($\beta > 0.7$, 363,170 points for toluene, 650,900 points for dodecane). Three specialized random forests were therefore trained on these three datasets for each precursor.

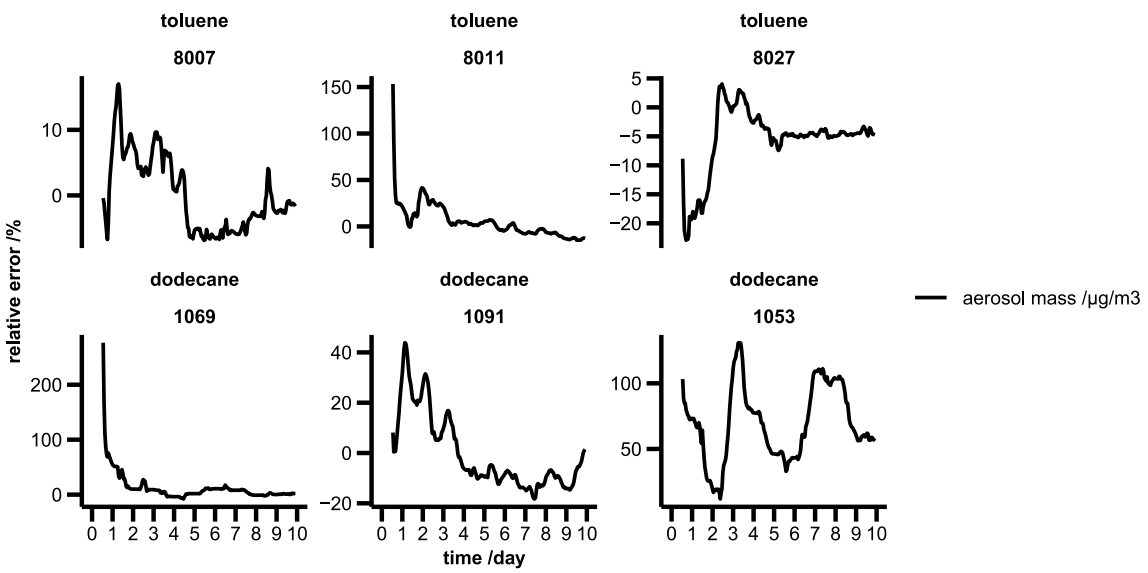

Figure 6. Time-series of the relative error on predicted aerosol mass for three toluene and three dodecane sample experiments shown on Figure 5.

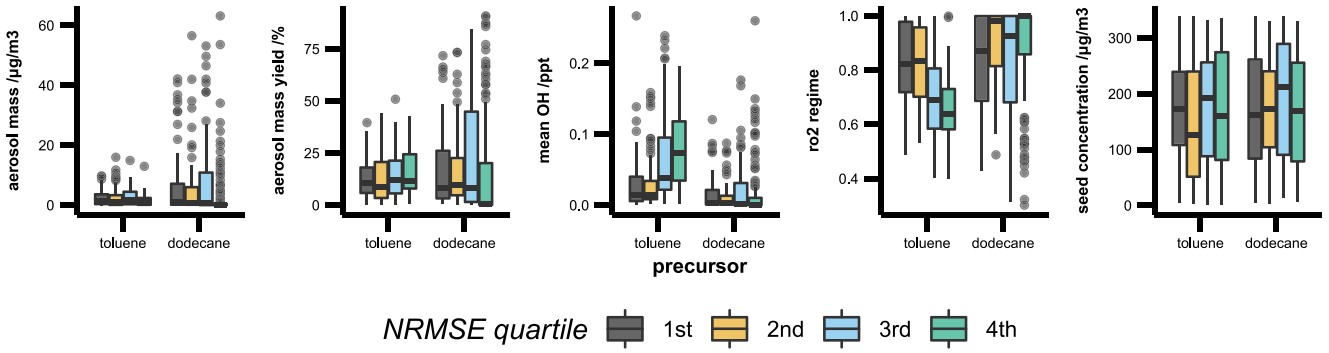

Figure 7. Boxplot distribution of validation simulations properties, according to their Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere simulation versus random forest normalized root mean square error quartile.

Figure 8 displays the distribution of Pearson correlation coefficients for the toluene and dodecane validation simulations for each specialized random forest compared to the original random forest trained with 1,000,000 points. For toluene simulations, all the specialized random forests display similar performances to the original random forest: for low NO_x, $r_{median} = 0.98$, for mid NO_x, $r_{median} = 0.99$ and for high NO_x, $r_{median} = 1.0$. Similarly for dodecane simulations, all specialized random forests exhibit performances similar to the original one, with median r values ranging from 0.98 to 0.99.

It is likely that the potential improvement caused by specializating the random forests over different RO_2 regimes is negatively compensated by performance reduction caused by a lower number of training points. Furthermore, in the dodecane case, specializing a random forest for high NO_x does not have a significant impact on the number of validation outliers in this regime.

3.5. Predictors Importance

After training the random forest, it is possible to estimate the relative importance of the chosen predictors. The feature importances in the *scikit-learn* python library, known as Gini importance or mean decrease impurity, are estimated following Breiman (2017): at a given node, the importance of the predictor selected as the threshold

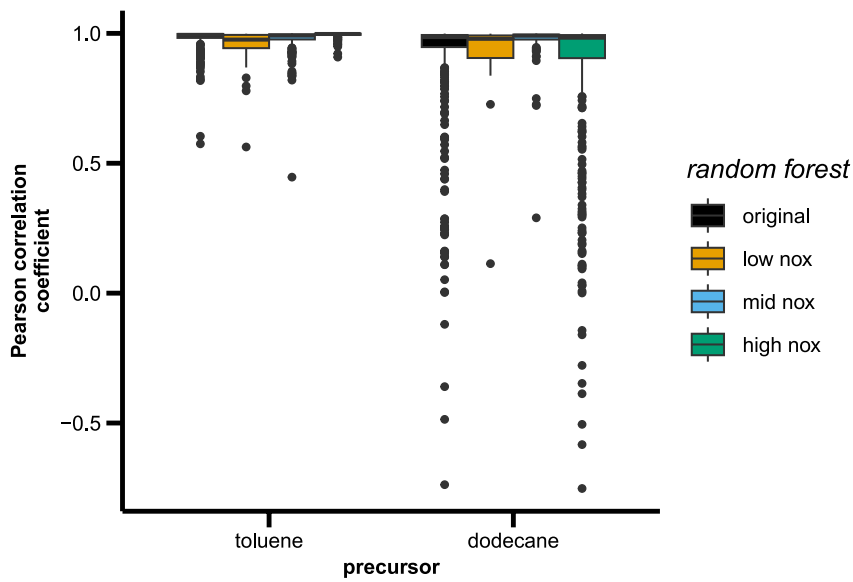

Figure 8. Boxplots of the Pearson correlation coefficients distribution for the toluene and dodecane validation simulations for the original random forests (black) and the three specialized random forests: low NO_x (orange), mid NO_x (blue), and high NO_x (green).

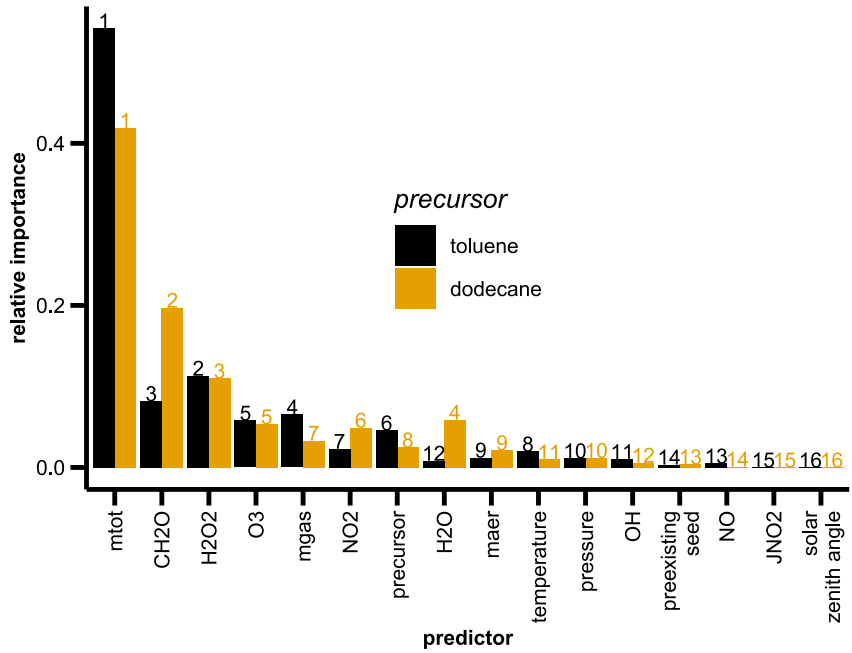

Figure 9. Predictors relative importance for the toluene (black) and dodecane (orange) random forests. The rank of each predictor is indicated at the top of the bars. The predictors names are detailed in Table 2.

criteria is defined as the total decrease in node impurity weighted by the proportion of samples reaching that node (which approximates the probability of reaching that node). This method gives more weight to predictors used in nodes higher up in the decision tree structure. The importance of each predictor is then averaged over all trees of the random forest. It is crucial to keep in mind that the correlation of predictors in the data set can bias the importance metrics of the model. When there is high correlation between predictors, it can be difficult for the model to determine which of the correlated variables are actually important for making predictions. As a result, the predictors importance metrics may be distributed more evenly across the correlated variables, leading to lower importance scores for all the correlated variables. This makes it more difficult to interpret the relative importance of each predictor and identify the most important predictors. Several techniques can be used such as recursive feature elimination (Gregorutti et al., 2017) to remove highly correlated predictors before training the model to obtain more accurate and informative variable importance metrics.

The resulting predictors importances are shown on Figure 9. For both toluene and dodecane, the most important predictor for organic mass is the total organic mass in the previous time-step. This finding is consistent with the fact that total organic mass is one of the predicted outcomes by the random forests. CH_2O and H_2O_2 are the second and third (resp. third and second) most important predictors for dodecane (resp. toluene). Since higher H_2O_2 concentrations are indicative of low NO_x situations, H_2O_2 can be considered as a proxy for the RO_2 regime. CH_2O is the only predictor related to secondary organic gaseous species and could be interpreted as a proxy for organic gases formation.

The water vapor concentration and organic gaseous fraction (m_g) are the fourth most important predictors for dodecane and toluene, respectively. Since the random forests are predicting the trend of m_g, it is logical that its previous step value is a significant predictor. The importance of water vapor is likely related to its role in OH production.

O_3 is the fifth most important predictor for both precursors. The information brought by the ozone predictor is related to general oxidants levels, the diurnal cycle as well as the RO_2 regime. NO_2 is the sixth most important predictor for dodecane while it is the precursor's concentration for toluene. These two predictors provide information about the diurnal cycle, the oxidants levels as well as the potential for secondary organic matter production. The seventh is the organic gaseous mass fraction for dodecane and NO_2 for toluene. The precursor's concentration and temperature are the eighth most important predictors for dodecane and toluene respectively. For both precursors, the organic aerosol fraction (m_a) is the ninth most important predictor. Because the gaseous mass fraction

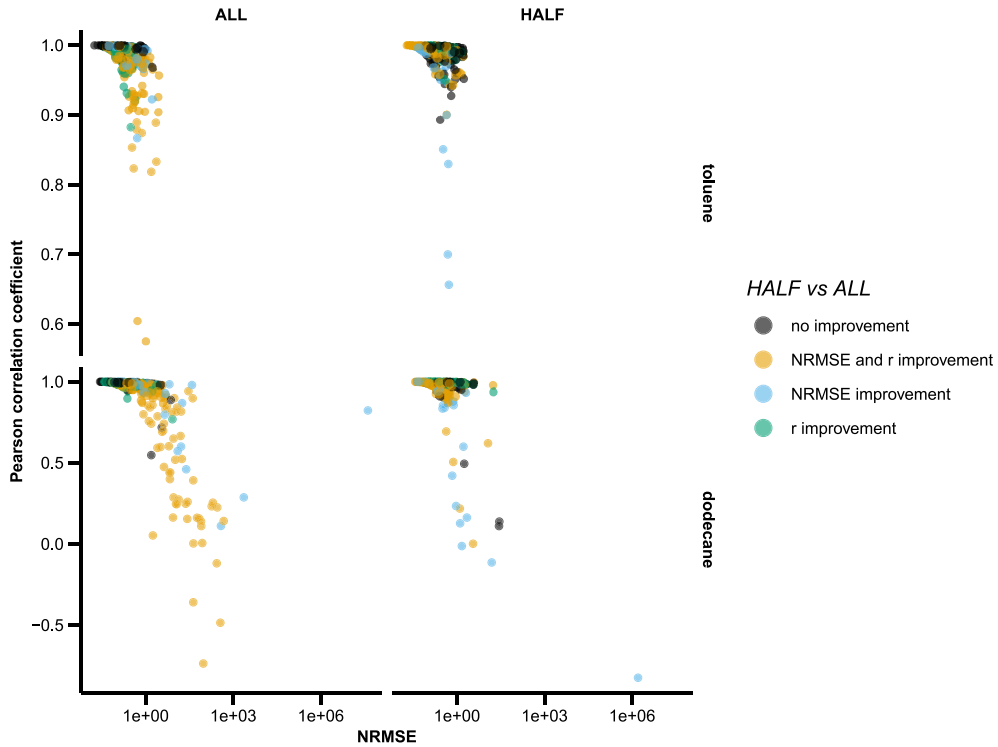

Figure 10. Scatterplot of the validation simulations' Pearson correlation coefficients (r) as a function of their normalized root mean square error (NRMSE) for toluene (top row) and dodecane (bottom row, note different y-axis scale), for the random forests trained with the 16 originally selected predictors (ALL, left column) and the random forests trained with only the eight most important predictors (HALF, right column). The colors depict whether r, NRMSE or both scores are improved when reducing the number of predictors.

is a more important predictor for both precursors, m_a only provides complementary information. The remaining predictors only have negligible contributions to the random forests. Since we've shown that SOA formation is not sensitive to pre-existing particle seed (see Figure 3) it is logical that this predictor is not important. Predictors directly related to the diurnal cycle (J_{NO_2} and solar zenith angle) are unimportant here, meaning that there is enough information provided by the time evolution of ozone and precursor concentrations to control for daytime versus nighttime organic matter production. Similarly, the precursor decay as well as ozone concentrations give enough information related to oxidant concentrations and RO_2 regime, explaining the weak importance of OH and NO predictors.

Since the contributions of the various predictors are dominated by only a few of them, we trained new random forests only using the 8 most important predictors for each precursor: m_t, CH_2O, H_2O_2, H_2O, O_3, NO_2, m_g and the precursor concentration for dodecane and m_t, H_2O_2, CH_2O, m_g, O_3, the precursor concentration, NO_2 and temperature for toluene. Figure 10 compares the Pearson correlation coefficients (r) and NRMSE scores calculated for each validation simulation (359 for each precursor) of the random forest trained with the 16 original predictors with the random forest trained with the eight most important identified predictors (see Figure 9).

Reducing the number of predictors for dodecane improves r for 239 (67%) validation simulations, with an average r increase of 0.12. The NRMSE decreased for 212 (59%) dodecane validation simulations, with an average NRMSE reduction of 59%. For 160 (45%) of the dodecane validation simulations, both r and NRMSE are improved. Reducing the number of predictors leads to an improvement of r for 212 (62%) toluene validation simulations, with an average r increase of 0.019. The NRMSE decreased for 190 (53%) toluene validation simulations, with an average NRMSE reduction of 40%. For 148 (41%) of the toluene validation simulations, both r and NRMSE are improved.

As shown in Figure 10, reducing the number of predictors is beneficial for the worst performing simulations, especially for dodecane. For both toluene and dodecane, the majority of validation simulations are improved

when halving the number of predictors. However, 7 out of the 8 selected predictors are shared by both random forests. The relative importance and ranks of the predictors differ between both random forests. There is therefore no guarantee that different precursors have the same optimal number of predictors and that they share the same predictors.

4. Conclusions

In this work we trained two random forests to predict organic mass production in both gas and aerosol phases resulting from toluene and dodecane oxidation. The random forests were trained on a data set created with the GECKO-A explicit organic chemistry box model. The data set contains a series of single box-model simulations covering a wide range of environmental conditions to ensure that the resulting random forests are able to reproduce the complex relations between organic aerosol production and the chemical environment. The resulting random forests show very good performances in predicting organic mass evolution in varied conditions when tested on a similar random set of box-model simulations. The distribution of errors in testing simulations highlights however the importance of carefully preparing the training data set. Our results suggest that random sampling over a range of possible environmental conditions is insufficient to build a robust training data set, and that is more important to properly sample a range of more complex chemical parameters such as the RO_2 regimes (β). We have shown that the range of β that needs focus depends on the precursors. For instance, the dodecane random forest has weaker performance for high β whereas the toluene random forest has lower performance for medium β values. However, creating multiple random forests each trained over a smaller range of β does not lead to more robust results than a single random forest. It seems more efficient to add additional training data points for the poorly performing RO_2 regimes.

The selection of predictors is also a crucial step. We have shown that it is possible to increase the random forest performance by reducing the number of predictors to the most important ones. However, there is no reason to think that these predictors have to be the same for different precursors, highlighting the care that must be taken in their selection. In this work, we selected the most important predictors by first training a random forests with a wide selection of predictors, and then training a new random forest with only the most important predictors identified in the first random forest.

In this work, we have therefore shown the feasibility of building random forests that behave like a detailed chemical mechanism for predicting secondary organic mass and its partitioning between gas and particle phases. There are still some limitations to overcome before the implementation of the random forest SOA emulator within a chemistry-climate model. First, even if the random forests are performing well, there are still some critical outliers at the validation stage (e.g., high NO_x dodecane). More work needs to be focused at removing these outliers because falling in one of these bad cases in a 3D model run would likely make the full simulation diverge. Second, the random forests were each trained to reproduce the oxidation of a single precursor. Additional studies are required to quantify whether it is important to represent the interactions of multiple primary hydrocarbons, their competition for oxidants, and the impact on the resulting SOA formation.

Data Availability Statement

The data set that was constructed to train and test the random forests and the Python code implementing the random forest training and testing have been uploaded on Zenodo (Mouchel-Vallon & Hodzic, 2022).

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