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## Precursor apportionment of atmospheric oxygenated organic molecules using a machine learning method†

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Gas-phase oxygenated organic molecules (OOMs) can contribute significantly to both atmospheric new particle growth and secondary organic aerosol formation. Precursor apportionment of atmospheric OOMs connects them with volatile organic compounds (VOCs). Since atmospheric OOMs are often highly functionalized products of multistep reactions, it is challenging to reveal the complete mapping relationships between OOMs and their precursors. In this study, we demonstrate that the machine learning method is useful in attributing atmospheric OOMs to their precursors using several chemical indicators, such as O/C ratio and H/C ratio. The model is trained and tested using data acquired in controlled laboratory experiments, covering the oxidation products of four main types of VOCs (isoprene, monoterpenes, aliphatics, and aromatics). Then, the model is used for analyzing atmospheric OOMs measured in both urban Beijing and a boreal forest environment in southern Finland. The results suggest that atmospheric OOMs in these two environments can be reasonably assigned to their precursors. Beijing is an anthropogenic VOC dominated environment with ~64% aromatic and aliphatic OOMs, and the other boreal forested area has ~76% monoterpene OOMs. This pilot study shows that machine learning can be a promising tool in atmospheric chemistry for connecting the dots.

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† Electronic supplementary information (ESI) available: Detailed information on the evaluation of the precursor apportionment model (Table S1); examples of the voting strategy (Table S2); examples of the expansion of aliphatic OOMs (Table S3); the Venn-plot of laboratory-generated OOMs used in this study (Fig. S1); the apportionment results of OOMs in urban Beijing using the workflow method (Fig. S2); the overall accuracy of the decision tree model using a laboratory dataset without the expansion of aliphatic OOMs (Fig. S3); the application results for the model trained with the dataset without the expansion of aliphatic OOMs (Fig. S4); and the performances of models consisting of different number of trees (Fig. S5). See DOI: <https://doi.org/10.1039/d2ea00128d>

### Environmental significance

The formation of new particles and secondary organic aerosols has potential effects on the earth's radiation balance and air quality, and atmospheric oxygenated organic molecules (OOMs) are currently acknowledged to significantly contribute to them. Volatile organic compounds (VOCs) are important precursors for OOMs. Since OOMs are often highly functionalized products of multistep reactions in the complex atmospheric environments, it is challenging to reveal the complete mapping relationships between OOMs and their precursors. In this study, we demonstrate that the machine learning method is useful in attributing OOMs to their precursors using several chemical indicators, such as O/C ratio and H/C ratio. Based on controlled oxidation experiments of four main types of VOCs (isoprene, monoterpenes, aliphatics, and aromatics), we trained the machine learning model to extract the internal mapping relationships of OOMs and their precursors. It showed unique advantages in precursor apportionment over current methods. We then used it for analyzing atmospheric OOMs measured in both urban Beijing and forested Hyttiälä. It can well identify the differences in OOMs between anthropogenic and biogenic dominated atmospheric environments. This pilot study shows that machine learning can be a promising tool in atmospheric chemistry for connecting the dots and is worth further exploring.

## 1 Introduction

Gas-phase oxygenated organic molecules (OOMs) can condense onto particles and usually play dominant roles in atmospheric



new particle growth and even in secondary organic aerosol formation,<sup>1–4</sup> which is known to affect the earth's radiation balance and air quality. OOMs are formed in the atmosphere *via* complex oxidation of volatile organic compounds (VOCs), in which autoxidation and multi-generation oxidation are usually involved.<sup>5,6</sup> Precursor apportionment of atmospheric OOMs is the cornerstone of understanding their origins, which helps in regulating the emissions of their precursors.

Both the complex functionalization processes and the differences between controlled laboratory conditions and real atmospheric environments make precursor apportionment of atmospheric OOMs very challenging. The VOC precursors of OOMs can generally be divided into anthropogenic volatile organic compounds (*e.g.*, aromatics and aliphatics) and biogenic volatile organic compounds (*e.g.*, monoterpenes and isoprene).<sup>7</sup> Positive matrix factorization (PMF)<sup>8</sup> is a widely used source apportionment analysis method,<sup>9,10</sup> which classifies numerous species into several factors based on their similarities in temporal variations. However, PMF could not sufficiently attribute atmospheric OOMs to their precursors.<sup>9,11</sup> It is partially because the concentration of OOMs is not only affected by their precursors but also strongly affected by the oxidation processes under the given atmospheric conditions. Atmospheric OOMs from different precursors may share similar time series if they are oxidized by the same oxidants. For example, OOMs generated from the photo-oxidation of aromatics and monoterpenes cannot be separated by the PMF analysis.<sup>3</sup> Recently, Nie *et al.*<sup>3</sup> developed a workflow method, which performs precursor apportionment of atmospheric OOMs based on the up-to-date knowledge of the characteristics of the products from VOC oxidation processes. In that method, however, the identification of OOMs from monoterpene oxidation (monoterpene OOMs) still relies on PMF. This is known to underestimate the concentration of monoterpene OOMs because monoterpene oxidized by the OH radical, which exists under the given conditions, cannot easily be retrieved by PMF.<sup>3,12</sup> Controlled laboratory experiments provide important information on OOMs produced from different precursors. However, the chemical settings of controlled laboratory experiments cannot fully reflect the complexity of the real atmosphere, which makes it difficult to interpret atmospheric OOMs solely based on laboratory conditions.

Machine learning methods possess the advantage of mining the relationships among complex data. For instance, the decision tree model, one of the classical machine learning methods, has been successfully applied in predicting amino acid sequencing and in proteomics research due to its high interpretability and tolerance of data scale.<sup>13,14</sup> With the development and application of high-resolution mass spectrometry in the field of atmospheric chemistry, large datasets of atmospheric OOMs at the molecular level are obtained with high time resolution for long-term periods.<sup>4,15</sup> Thus, finding precursors for atmospheric OOMs appears to be a natural playground for machine learning methods. They have the potential to mine large datasets from controlled laboratory experiments with no reliance on the variation of OOM concentration and further

attribute OOMs measured in ambient atmosphere to their likely precursors.

In this work, we use the decision tree model to test the feasibility of machine learning for precursor apportionment of atmospheric OOMs. This model is trained and tested using the datasets from controlled laboratory experiments using various VOCs as OOM precursors. It helps build a mapping relationship between OOMs and their precursors, including isoprene, monoterpenes, aliphatics, and aromatics. Finally, we apply the model to atmospheric datasets obtained in both urban Beijing and a remote forest environment of Hyytiälä.

## 2 Methods

### 2.1 Model training strategy

Decision tree was selected for precursor apportionment of OOMs, performed with the *Statistics and Machine Learning Toolbox* in MATLAB. It is a supervised machine learning method, which requires training data that are *pre-labeled* with known precursors (from controlled laboratory experiments). According to the standard training process of machine learning models, we randomly divided the pre-labeled data into the training dataset and the testing dataset, accounting for 70% and 30% of the data, respectively. As shown in Fig. 1, the training dataset was used to obtain the decision tree, and the testing dataset was used to evaluate the accuracy of the trained model. Cross-validation was also performed which is described in the ESI.†

In order to reduce the uncertainty caused by one single training model, we repeated the above process ten times to get ten independent decision trees. The outputs of all the ten decision trees vote together for the final apportionment result. Specifically, precursors with votes more than the upper limit, *i.e.*, the sum of the mean and standard deviation of the whole votes, will be retained, or, if all the precursors received votes no more than the upper limit, precursors with votes more than the lower limit will be retained, *i.e.*, the difference of the mean and standard deviation of the whole votes. There are examples in the ESI† to illustrate this rule.

This voting strategy also helps to reduce the uncertainty caused by the overlapping formulae of OOMs generated from different precursors. According to the laboratory experiments, there are overlaps between OOMs oxidized from those precursors (Fig. S1†). Therefore, in the pre-labeled dataset, the same descriptors of an OOM molecule may correspond to more than one label of precursors. The ten decision trees would take into account the overlaps and give a combination of the most likely answers. Table S2† shows two examples of the determination of overlapping molecules. The optimization process for the number of trees is described in the ESI.†

### 2.2 Pre-labeled dataset and atmospheric dataset

Pre-labeled data are obtained from single-precursor oxidation experiments performed under laboratory-controlled conditions. Precursors are marked with four labels, *i.e.*, isoprene, monoterpenes, aliphatics, and aromatics. Isoprene and





**Fig. 1** Schematic of the decision tree in the precursor apportionment of atmospheric oxygenated organic molecules. In the confusion matrix, TP, FP, or TN is a description of the labels and predictions, in which T is for true, F is for false, P is for positive, and N is for negative. The above table takes aromatics as an example. The first letter is for reality and the latter is for prediction. Thus, when aromatics are predicted as aromatics, the result is TP; when aromatics are predicted as aliphatics and monoterpenes, the result is TN; when aliphatics and monoterpenes are predicted as aromatics, the result is FP.

monoterpenes such as  $\alpha$ -pinene and limonene are representative biogenic precursors with high formation yields of OOMs. From previous monoterpene oxidation experiments, 872 products of monoterpene OOMs were reported (Table 1).<sup>16,17</sup> As the laboratory report for comprehensive isoprene OOMs is not available, we obtained 30 isoprene OOMs from a previous field measurement.<sup>18</sup> Aromatics are representative anthropogenic precursors and are considered to make a dominant contribution to the formation of OOMs in an urban atmospheric environment. According to previous studies of aromatic oxidation experiments, 485 oxidation products were used in the pre-labeled dataset.<sup>6,19,20</sup>

Aliphatics, including some endocyclic alkenes and straight-chain model compounds, also give considerable yields of OOMs which are present in relatively high atmospheric concentrations in an urban environment. Due to the limited laboratory experiments on aliphatics, only 63 oxidation products of aliphatics are available which are mainly C6 and C10

substances.<sup>21</sup> In order to reduce the possible biases due to the imbalanced number of input compounds for different precursors, we tested the case that artificially increases the number of aliphatic compounds by adding unstudied but likely existing ones that are homologous to the studied aliphatic compounds. The number of oxidation products of aliphatics was extended to 346. Details about the expansion of aliphatic OOMs and comparisons to the results without the expansion are provided in the ESI.† It should be noted that this cannot be done for isoprene OOMs, because isoprene itself has no homologous compounds.

As shown in Table 1, there are eight descriptors of oxidation products based on their chemical formula. The first four are the original information of elemental composition, *i.e.*, the number of C, H, O, and N. The other four are the processed information of the molecules: double bond equivalent (DBE) and H/C ratio which reflect the carbon saturation state of OOMs, and O/C ratio and OSc which reflect their carbon oxidation state. As

**Table 1** Summary of the oxidation products of monoterpenes,<sup>16,17</sup> aliphatics,<sup>21,31</sup> aromatics,<sup>6,19,20</sup> and isoprene<sup>18</sup> under controlled laboratory conditions

Precursors	Products	Descriptors <sup>a,b,c,d,f</sup>							
		nC	nH	nO	nN	DBE	H/C	O/C	OSc
<b>Monoterpenes</b>									
$\alpha$ -Pinene, limonene	872	12.7	19.3	9.3	0.4	3.9	1.5	0.9	0.2
<b>Aliphatics</b>									
<i>t</i> -Decalin, decalin, cyclohexane, <i>n</i> -decane, generated <sup>e</sup>	346	11.4	20.5	5.8	0.2	2.0	1.8	0.6	-0.6
<b>Aromatics</b>									
Benzene, toluene, ethylbenzene, xylene, mesitylene	485	9.7	11.8	8.8	0.2	4.7	1.2	1.1	0.9
<b>Isoprene</b>									
	30	4.7	8.7	4.9	0.9	0.9	1.9	1.4	1.02

<sup>a</sup> nC, nH, nO, and nN: the number of carbon, hydrogen, oxygen, and nitrogen atoms. <sup>b</sup> DBE: double bond equivalent, which is calculated as  $(2nC + 2nH - nN)/2$ . <sup>c</sup> H/C and O/C: the ratio of nH over nC, and nO over nC. <sup>d</sup> OSc: carbon oxidation state, which is calculated as  $2O/C - H/C$ .<sup>32</sup> <sup>e</sup> 63 reported aliphatic OOMs and 283 generated substances (adding an integer number of CH<sub>2</sub> groups to the reported aliphatic OOMs).<sup>f</sup> Values of nC, nH, nO, nN, DBE, H/C, O/C, and OSc in the table are averaged from each product with equal weight.



the VOC precursors are different in carbon number, DBE, and functional groups, their oxidation pathways and the generated OOMs are significantly different in these characteristics. For example, monoterpene OOMs and aromatic OOMs generally have higher DBE than aliphatic OOMs based on the current knowledge of their oxidation reactions.<sup>3</sup> We found that the minimum number of features is 3–4 including H/C and O/C. Increasing the number of features would slightly improve the performance of the models. So all the 8 features are selected.

The atmospheric OOM dataset was acquired from the measurements in Beijing (324 OOM species) and Hyytiälä (328 OOM species). The former was conducted at the BUCT-AHL site during 2018.12.26–2019.1.26, which is ~500 meters west of the Third Ring Road in Beijing with heavy traffic loading and surrounded by residential and commercial areas.<sup>4,22</sup> The latter was made at the SMEAR II station during 2018.3.9–2018.3.31.<sup>9</sup> Details of these two sites can be found in previous studies. Note that the atmospheric OOM dataset does not contain nitrated phenols since its concentration is significantly high in Beijing (~75%). In order to reduce the impact of extremely high signals,<sup>3</sup> we excluded nitrated phenols in this analysis.

### 2.3 Evaluation methods

Validation of the trained model was performed with the testing laboratory dataset. For the testing laboratory dataset, a confusion matrix was used to evaluate the consistency between the predicted results and their original labels (Fig. 1). Taking

aromatics as an example, *precision* is used to characterize how many compounds that were predicted as aromatic OOMs are truly from aromatics, and *recall* is used to characterize how many aromatic oxidation products are predicted as aromatic OOMs. The overall accuracy of the decision tree is evaluated by *F1-score* (ranging from 0 to 1), which is calculated from *precision* and *recall*. Details of the evaluation metrics are given in the ESI.† Generally, we will train a series of decision trees with different number of bifurcations and then choose the most appropriate solution based on the variation of the above three indexes.

$$F1 \text{ score} = \frac{2 \times \text{precision} \times \text{recall}}{\text{precision} + \text{recall}} \quad (1)$$

## 3 Results and discussion

### 3.1 Model testing with laboratory data

We first test the performance of the model with the resting laboratory data, and the testing diagnostics show that the decision tree method can effectively attribute OOMs to their likely precursors. The decision tree with bifurcations of 10 was selected as the optimal solution. The overall accuracy (*F1-score*) of the precursor apportionment using the decision tree model is 0.75, 0.73, 0.67, and 0.47 for OOMs oxidized from monoterpenes, aliphatics, aromatics, and isoprene, respectively. The



Fig. 2 The overall accuracy of the decision tree model in the precursor apportionment of atmospheric oxygenated organic molecules using laboratory testing data (a) is for *F1-score*, (b) is for *precision*, and (c) is for *recall*. The dotted lines are the mean value of repeated tests and the shaded area illustrates the standard deviation.







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