Effect of chemical aging of monoterpane products on biogenic secondary organic aerosol concentrations

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HIGHLIGHTS

• Developed parameterization for monoterpane aging chemistry in VBS.
• Modest increases (17–21%) predicted for biogenic SOA during summer in Eastern US.
• Sensitivity tests suggest that the effect may be even smaller.
• Modest effect on the performance of PMCAMx during 2001 and 2013 (SOAS).

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ABSTRACT

Capturing the effects of the complex processes of chemical aging of biogenic secondary organic aerosol (SOA) in chemical transport models (CTMs) has been challenging. Recent laboratory results from atmospheric simulation chambers are used in this study to develop a parameterization for biogenic SOA formation as a result of aging for use in CTMs using the Volatility Basis Set framework. This parameterization was implemented in PMCAMx which was then applied over the eastern United States to simulate summertime conditions. Using the base case parameterization of monoterpane SOA chemical aging resulted in modest increases (17–21%) in predicted domain average biogenic SOA. An alternative parameterization was developed fitting the same laboratory results, but assuming higher volatility products of chemical aging reactions. Use of this parameterization resulted in small increases (1–4%) of the predicted biogenic SOA. The PMCAMx predictions in sites where most of the SOA was biogenic were evaluated against measurements from the EPA IMPROVE network in July 2001 and during the Southern Oxidant and Aerosol Study in June 2013. The differences in the model performance were modest and mixed, a result consistent with the relatively small effect of the proposed parameterization on total organic aerosol levels.

1. Introduction

Anthropogenic activities and biogenic sources emit particulate matter (PM) and particle precursors into the atmosphere. Atmospheric PM has a significant impact on human cardiovascular health (Pope et al., 2009), as well as complex effects on climate (Climate Change 2007, 2007). Organic compounds comprise a major portion of submicron aerosol mass (Zhang et al., 2007). Particle phase organic compounds that are emitted directly into the atmosphere are classified as primary organic aerosol (POA). Anthropogenic POA sources include industrial emissions, motor vehicle emissions, as well as other combustion-related emissions. Biogenic POA sources include plant emissions, such as pollen. Organic species can also end up in the particle phase through gas-phase chemical reactions of volatile (VOCs), intermediate volatility (IVOCs), and semivolatile organic compounds (SVOCs) with oxidants in the atmosphere and consequent condensation of lower volatility products (Robinson et al., 2007). Atmospheric aerosol formed through this mechanism is classified as secondary organic aerosol (SOA). Anthropogenic SOA precursors include aromatic and aliphatic compounds from fuel combustion and evaporation processes. Biogenic SOA precursors include isoprene, monoterpenes and sesquiterpenes. The total emissions of biogenic SOA precursors significantly exceed anthropogenic emissions.

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emissions on a global scale (Kanakidou et al., 2005), therefore a detailed understanding of biogenic SOA formation is critical for our understanding of the role of atmospheric particles on both air quality and climate change.

Biogenic SOA is formed by the reactions of biogenic VOCs with oxidants such as ozone (O$_3$), the hydroxyl radical (OH), and the nitrate radical (NO$_3$). This process results in some products that are less volatile than the precursor due to the addition of oxygenated functional groups. Multiple generations of oxidation take place over a timescale of days in the atmosphere. This further oxidation of first-generation products and the corresponding SOA that forms as a result of later generations of reactions is known as chemical aging (Donahue et al., 2006). Previous studies have explored various chemical aging mechanisms including homogeneous oxidation in the gas phase (Donahue et al., 2012), oligomerization of oxidation products (Kalberer et al., 2006), heterogeneous reactions of oxidants with aerosol (George et al., 2008), and photolysis (Henry and Donahue, 2012). Homogeneous OH reactions are generally much faster than heterogeneous reactions due to diffusion limitations of the latter (Lambe et al., 2009). For first generation reactions, volatile precursors typically have SOA mass yields of less than 20% (Kroll and Seinfeld, 2008). A lot of the organic compounds remaining in the gas phase are either semivolatile or have intermediate volatility (Presto and Donahue, 2006). These compounds can react further in the gas phase forming additional SOA. These later generations of chemical reactions represent a possible missing link between field measurements and model predictions.

Most chemical transport models (CTMs) rely upon SOA formation mechanisms that are based on smog-chamber experiments that last for a few hours. However, because submicron particulate matter has a lifetime on the order of days, these parameterizations likely do not accurately represent the complete behavior of organic aerosol in the atmosphere. Aging parameterizations have been developed to describe the reactions of semivolatile and intermediate volatility primary emissions (Robinson et al., 2007). These aging schemes do produce improved results in densely populated urban areas such as Mexico City (Tsimpidi et al., 2011). However, models assuming aggressive chemical aging of biogenic SOA tend to significantly over-predict OA in areas where biogenic VOCs dominate emissions such as the southeastern United States (Lane et al., 2008a), but also in urban areas during relatively clean periods (Zhang et al., 2013). These conflicting results suggest the importance of biogenic SOA aging as a potential reason for discrepancies between field observations and model results.

Wang et al. (2018) and Wang (2018) performed experiments to determine SOA mass yields as a result of chemical aging of α-pinene SOA. The products of ozonolysis of α-pinene were then allowed to react further with OH over an equivalent of multiple days of typical atmospheric conditions. This process was shown to result in a 20–40% increase in SOA formed for the conditions of the corresponding experiments. These studies also explored differences in aged SOA formation of α-pinene ozonolysis products under both low-NO$_x$ and high-NO$_x$ conditions. The additional SOA observed as a result of chemical aging was significant but far less than what has been assumed in some previous modeling studies, where a two to three-fold increase was assumed (Lane et al., 2008a).

In this study, results from Wang et al. (Wang et al., 2018; Wang, 2018) are used to develop new parameterizations for the aging of biogenic SOA for use in CTMs. Two parameterizations are developed, both reproducing the experimental results. These could serve to characterize the uncertainty of the predictions of the aging scheme proposed. These parameterizations are implemented in the three-dimensional CTM PMCAMx and are used to quantify the effect of the chemical aging of monoterpene products on the predicted biogenic SOA in the Eastern US during two summer periods.

2. Model description and application

The Particulate Matter Comprehensive Air Quality Model with Extensions (PMCAMx) (Tsimpidi et al., 2011; Gaydos et al., 2007; Karydis et al., 2010; Tsipmidipitali et al., 2011; Gaydos et al., 2007; Karydis et al., 2010; Murphy and Pandis, 2010) was used to evaluate the parameterizations developed in this study. This CTM uses the framework of CAMx (ENVIRON, 2006) to simulate advection and diffusion, wet and dry deposition, as well as gas and aqueous phase chemistry. An aerosol sectional approach is implemented with 10 size bins ranging from 40 nm to 40 μm in diameter. Various inorganic aerosol species are modeled including sulfate, nitrate, ammonium, chloride, sodium, water, and crustal species. Inorganic aerosol partitioning is simulated using the ISORROPIA aerosol thermodynamic model (Nenes et al., 1998). Carbonaceous aerosol components modeled include elemental carbon as well as a series of surrogate primary and secondary organic components. The SAPRC (Carter, 1999) chemical mechanism is used to simulate atmospheric chemistry. The version of SAPRC used contains 237 reactions of 73 gas phase species and 18 radical species. 37 surrogate organic aerosol species have been added to the mechanism for the volatility basis set (VBS) implementation.

Volatility of organic aerosol is modeled using the VBS (Donahue et al., 2006) and SOA formation using NO$_x$-dependent yields (Lane et al., 2008b). POA species are described using 10 volatility bins ranging in saturation concentration from 10$^{-3}$ to 10$^{-6}$ μg m$^{-3}$ at 298 K. SOA species are described using 4 volatility bins ranging in saturation concentration from 1 to 10$^{-3}$ μg m$^{-3}$ at 298 K (Murphy et al., 2009), and are split into SOA from anthropogenic sources (aSOA) and SOA from biogenic sources (bSOA).

PMCAMx was applied over the Eastern United States for two simulation periods: July 12 – July 28, 2001 and June 8–24, 2013. The first is the EPA Supersites summer intensive period that has been studied extensively. The second is part of the Southern Oxidant and Aerosol Study (SOAS). The modeling domain for the 2001 simulation is a 3492 × 3240 km grid comprised of 36 × 36 km cells with 14 vertical layers that have a ceiling of 6 km. Emissions for the 2001 simulation were derived from the 1999 National Emissions Inventory (NEI) version 3 (Office of Air Quality Pla, 2002). Biogenic emissions were estimated using the Biogenic Emissions Inventory System (BEIS) version 3.13. Sesquiterpene emissions were assumed to be 30% of the monoterpene emissions (Helming et al., 2006). Meteorological fields for this time period were produced using MM5 (Georg et al., 1994).

The modeling domain for the 2013 simulation is a 3348 × 2880 km grid comprised of 12 × 12 km cells with 14 vertical layers that have a ceiling of 6 km. Emissions for the 2013 simulation were processed using the Sparse Matrix Operator Kernel for Emissions (SMOKE) modeling system (Houyoux and Vukovich, 1999). Point and area source emissions were based on the 2011 NEI (version 1) and biogenic emissions were estimated using BEIS version 3.61. Meteorological fields were produced using the Weather Research and Forecasting (WRF) model version 3.6.

In PMCAMx, SOA is formed through a series of steps. In the case of bSOA three gas phase SOA precursors (isoprene, monoterpenes, and sesquiterpenes) each undergo reactions with oxidants including hydroxyl radical (OH), ozone (O$_3$), nitrate radical (NO$_3$), and oxygen atoms (O). The monoterpenes and sesquiterpenes surrogates are lumped species that represent the wide range of compounds that comprise them by a weighted average of their physical and chemical properties (ENVIRON, 2006). These reactions reduce the concentration of each gas phase SOA of varying volatility as described by the VBS. A NO$_x$ level dependency is applied to the yield of each reaction for SOA in each volatility bin. The overall yield is given as

$$\alpha_i = \alpha_{i,\text{high}}B + \alpha_{i,\text{low}}(1 - B)$$

(1)

where $\alpha_{i,\text{high}}$ is the high-NO$_x$ yield, $\alpha_{i,\text{low}}$ is the low-NO$_x$ yield, and B is the branching ratio, which is a parameter determined by the fraction of
organo-peroxy radicals that react with NO (Lane et al., 2008). This branching ratio is given by

\[
B = \frac{\text{(rate of } \text{RO}_2 + \text{NO})}{\text{(rate of } \text{RO}_1 + \text{NO}) + (\text{rate of } \text{RO}_2 + \text{RO}_1) + (\text{rate of } \text{RO}_1 + \text{HO}_2)}
\]  

(2)

This branching ratio is allowed to vary in space and time inside the modeling domain. A new value is calculated at each time step and within each grid cell based on the rates of the reactions of organo-peroxy radical species with both NO as well as with other peroxy radical species. These rates used for calculating the branching ratio are also continuously calculated using chemical reaction parameters set in the model as well the relevant radical concentrations in each grid cell at each time step. The rates of approximately 20 reactions are used in PMCAMx based on the SAPRC chemistry for the continuous calculation of the branching ratio \( B \). A branching ratio of unity means that all organo-peroxy radicals react with NO and indicates high-NO\(_x\) conditions. A branching ratio of zero means that all organo-peroxy radicals react with other peroxy radicals and indicates a low-NO\(_x\) environment (Lane et al., 2008). A partitioning algorithm assuming pseudo-ideal equilibrium is then used to determine the mass of bSOA product that is transferred to the particle phase.

3. Parameterization of NO\(_x\)-Dependent chemical aging of SOA

Chemical aging in PMCAMx is modeled by transferring mass between volatility bins of the SOA species. Previous attempts at modeling aging of biogenic SOA have utilized a sequential, one bin downward shift in volatility (Qin et al., 2018), and in some cases included an increase in mass due to increased oxygen content as a result of aging processes (Lane et al., 2008a). These models have led to overestimation of OA. We tried a number of parameterization schemes of variable complexity to fit the measurements of Wang et al. (Wang et al., 2018; Wang, 2018) Given that even the simplest ones consisting of a single reaction could reproduce the observations, we have selected such simple schemes. In all cases, the parameterization of Lane et al. (2008b) is used for the production of the first generation SOA from monoterpenes. The new aging scheme outlined here is designed to specifically account for the later generations of chemical aging reactions of monoterpenes. Effects of other biogenic compounds (e.g. isoprene and its oxidation products) are not explicitly explored in this analysis, although this is a potentially important topic of future research.

The implementation of aging chemistry here assumes that the later generations of reactions are dominated by oxidation by OH. \( \text{O}_3 \) is not expected to have a major role in later generations of SOA formation after the first generation exhausts double bonds in precursor molecules that facilitate reaction with \( \text{O}_3 \). While \( \text{O}_3 \) could participate in the later generation of reactions, little is known about these specific processes and they are therefore not included in the model chemical reactions. However, the first generation of reactions of monoterpenes with \( \text{NO}_3 \) and the corresponding SOA production are included in the model based on the Lane et al. (2008b) parameterizations.

The base scheme assumes that the surrogate VBS component with the highest volatility, SOA(\( C^* = 10^3 \) \( \mu \text{g m}^{-3} \)), reacts with OH to produce material with the lowest volatility, SOA(\( C^* = 10^0 \) \( \mu \text{g m}^{-3} \)) with a yield \( a_0 \):

\[
\text{SOA}(C^* = 10^3) + \text{OH} \rightarrow a_0\text{SOA}(C^* = 10^0) + \text{volatile products}
\]

(3)

The yield \( a_0 \) was used as a fitting parameter and a value of 0.14 was found for the low-\( \text{NO}_x \) experiments and 0.08 for the high-\( \text{NO}_x \) ones. The predicted yields from this scheme are compared to the final yields (after chemical aging) of the Wang et al. (Wang et al., 2018; Wang, 2018) experiments in Fig. 1. The fractional error was found to be 17% for the low-\( \text{NO}_x \) experiments and 23% for the high-\( \text{NO}_x \) ones. The rate constant for these chemical aging reactions is assumed to be equal to \( 1 \times 10^{-11} \text{ cm}^2 \text{molec}^{-1} \text{s}^{-1} \). The \( \text{NO}_x \)-dependent yields are calculated using Eqs. (1) and (2). The above base case parameterization assumes that chemical aging processes result in the most extreme change in average volatility for the specific VBS scheme.

An additional parameterization was developed to assess the sensitivity of model results to the assumed volatility of the products of chemical aging reactions. In this parameterization, it is assumed that aged bSOA mass only moves from the highest volatility bin to the second highest volatility bin (i.e. \( C^* = 10^3 \) \( \mu \text{g m}^{-3} \) to \( C^* = 10^2 \) \( \mu \text{g m}^{-3} \)). This is represented by the reaction:

\[
\text{SOA}(C^* = 10^3) + \text{OH} \rightarrow a_0\text{SOA}(C^* = 10^0) + \text{volatile products}
\]

(4)

This version of the aging parameterization represents the opposite extreme case from the base case parameterization and will be referred to as the “sensitivity” parameterization. The optimum values of the mass yield \( a_0 \) were 0.37 for the low-\( \text{NO}_x \) case and 0.24 for the high-\( \text{NO}_x \) one. The rate constant for these chemical aging reactions is also assumed to be equal to \( 1 \times 10^{-11} \text{ cm}^2 \text{molec}^{-1} \text{s}^{-1} \). This parameterization was also able to fit the experimental measurements with fractional errors ranging from 14% for the high-\( \text{NO}_x \) measurements to 19% for the low-\( \text{NO}_x \) ones (Fig. 1), meaning its performance is as good as or slightly better than the base-case parameterization. In both versions of the aging parameterization, it is assumed that any mass lost from the highest volatility bin that is not transferred to the corresponding lower volatility bin becomes products of even higher volatility (fragmentation) that will not exist in the particle phase. Both parameterizations were implemented in PMCAMx to represent monoterpenes (using the results of \( \alpha \)-pine) and the sensitivity of OA predictions to the volatility of chemical aging SOA products is explored later sections. For the rest of the biogenic SOA components (isoprene and sesquiterpene derived SOA) the effect of the chemical aging reactions was assumed to be negligible (Baek et al., 2011).

4. Predicted biogenic secondary organic aerosol

Three simulations were performed for the 2001 and 2013 summer periods: one simulation with no aging of bSOA, one with low volatility bSOA products (base case), as well as one with high volatility bSOA products (sensitivity case). The predicted bSOA of each simulation is
examined in this section.

Fig. 2 shows the average predicted bSOA concentration for each simulation performed for the July 2001 period. The highest concentration of bSOA was predicted in the southeast in every simulation. The domain average bSOA concentration was 0.64 μg m⁻³, 0.78 μg m⁻³, and 0.65 μg m⁻³ for the no aging simulation, base simulation, and sensitivity simulation, respectively. In the southeastern US the average bSOA concentration was 1.93 μg m⁻³ (no aging), 2.33 μg m⁻³ (base case), and 1.97 μg m⁻³ (sensitivity case). The maximum concentration of bSOA in the southeastern US was 3.37 (no aging), 3.90 (base case), and 3.42 (sensitivity case). For all averages from the 2001 period, the sensitivity test resulted in increases of 1–2%. This small increase is due to the fact that the aging products remain in the gas phase in most areas and periods. On the other hand, the average concentrations increase 20–21% for the base case simulation as most of the aging products are in the particulate phase.

The highest percent increases of bSOA using the base case parameterization, up to 50%, are predicted downwind of source areas, in locations with relatively low predicted bSOA levels (Fig. 3). Significant increases of 20–30% are also seen in areas with high predicted bSOA levels, such as the southeast US. Using the sensitivity parameterization results in much lower percent increases across the domain. The largest effect on bSOA was a 4–5% increase that was predicted in low bSOA areas. Negligible to small increases of 0–2% were predicted in the southeast US.

Fig. 4 shows the average predicted bSOA concentration for each simulation performed for the June 2013 scenario. The domain average bSOA concentration was predicted to be 0.52 μg m⁻³, 0.64 μg m⁻³, and 0.55 μg m⁻³ for the no aging simulation, base simulation, and sensitivity simulation, respectively. In the southeastern US the average predicted bSOA concentration was 1.57 μg m⁻³ (no aging), 1.89 μg m⁻³ (base case), and 1.60 μg m⁻³ (sensitivity case). The maximum concentration of bSOA in the southeastern US was 2.71 (no aging), 3.22 (base case), and 2.86 (sensitivity case) in west-central Alabama. For all averages from the 2013 period, the sensitivity test resulted in increases of 2–6%. For the base parameterization, average concentrations increased by 20–23%.

In this case, once more, the sensitivity parameterization produced small changes in terms of bSOA concentrations in areas where large amounts of bSOA are expected (Fig. 5). With the base parameterization, increases in predicted bSOA of up to 50% were seen in low bSOA areas. Similar to the 2001-time period, increases in bSOA in the southeast US range from around 15–30%.

Further model evaluation will only use results from the no aging simulation and base case aging simulation, as the predicted change in

Fig. 2. Predicted average total bSOA concentration in μg m⁻³ for the (a) no aging simulation, (b) base simulation, and (c) sensitivity simulation for July 2001.

Fig. 3. Percentage increase in predicted bSOA due to chemical aging for: (a) base case and (b) sensitivity test for July 2001.
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bSOA using the sensitivity simulation was proven to be very small.

5. Comparison to field observations

To evaluate the model, organic aerosol (OA) predictions from the no aging simulation and base case aging simulation were compared to field observations in the Southeastern US where the higher bSOA concentrations are predicted and where the absolute effect of the chemical aging parameterization is higher. The metrics used for the evaluation are the fractional bias (FBIAS) and fractional error (FERROR).

\[
FBIAS = \frac{1}{n} \sum_{i=1}^{n} \left( \frac{P_i - O_i}{P_i + O_i} \right)
\]

\[
FERROR = \frac{1}{n} \sum_{i=1}^{n} \left( \frac{|P_i - O_i|}{P_i + O_i} \right)
\]

where \( n \) is the number of available observations, \( P_i \) is the predicted value corresponding to observation \( O_i \). The PMCAMx OA predictions are compared with field observations in IMPROVE sites for the July 2001 and June 2013 simulation periods in Fig. 6 and Fig. 7, respectively. These represent daily average measurements collected every three days. The measurements in the IMPROVE sites are expected to have a smaller influence from anthropogenic emissions. Table 1 summarizes the FBIAS and FERROR between predictions and field observations for the July 2001 and June 2013 periods using results from the no aging simulation and base case aging simulation.

During July 2001 the PMCAMx FBIAS for OA increased as a result of chemical aging. This is an expected result due to the increased bSOA production observed with the base case simulation. No change in FBIAS was observed for this time period. Hourly OA measurements taken during the Southern Oxidant and Aerosol Study in Centreville, AL and Research Triangle Park, NC (Carlton et al., 2013) were used to evaluate model performance in the June 2013 period. In this case, FBIAS also

Fig. 4. Predicted average total bSOA concentration in \( \mu g \ m^{-3} \) for the (a) no aging simulation, (b) base simulation, and (c) sensitivity simulation for June 2013.

Fig. 5. Percentage increase in predicted bSOA due to chemical aging for: (a) base case and (b) sensitivity test for June 2013.
while the model generally underpredicts the observed OA values, collected from the Centreville, AL site during the SOAS campaign in summertime OA in the southeastern United States, using samples ported that SOA from monoterpenes is an important contributor to the increase in bSOA also reduced FERROR during this time period, increased due to the increased production of low volatility bSOA species. The increase in bSOA also reduced FERROR during this time period, indicating a modest improvement in performance. Time series for the different parameterizations tested here are shown in Fig. 8 alongside daily OA observations at the Centreville, AL. Zhang et al. (2018) reported that SOA from monoterpenes is an important contributor to summertime OA in the southeastern United States, using samples collected from the Centreville, AL site during the SOAS campaign in 2013. While the model generally underpredicts the observed OA values, implementing the aging schemes based on monoterpenes brings the model closer to reproducing these values. Remaining discrepancies could be due to our simplified treatment of SOA production during the oxidation of isoprene, the contributions of sesquiterpenes, uncertainties in both anthropogenic and biogenic emissions, meteorology, etc.

6. Conclusions

Recent laboratory results were used to develop a parameterization for bSOA production as a result of aging for use in CTMs using the VBS framework for SOA formation. Using the base case parameterization, increases in predicted domain average bSOA of 0.14 μg m\(^{-3}\) (22%) and 0.12 μg m\(^{-3}\) (20%) were observed for the 2001 and 2013 time periods, respectively. This also corresponded with an increase in bSOA as a fraction of total OA of 0.04 (11–22%) in both simulation periods. The average bSOA in the Southeastern United States increased by 0.40 μg m\(^{-3}\) (21%) and 0.32 μg m\(^{-3}\) (20%) in 2001 and 2013, respectively. The maximum predicted bSOA in the Southeastern United States increased by 0.53 μg m\(^{-3}\) (16%) and 0.51 μg m\(^{-3}\) (19%) in 2001 and 2013, respectively.

A sensitivity test was performed in order to determine the effect of chemical aging product volatility on predicted bSOA. It was determined that using an aging scheme that results in products of only high volatility does not lead to enough particle phase bSOA to have any meaningful impact on OA predictions.

Comparison to field measurements for 2001 results showed an increase in fractional bias and negligible change in fractional error. Because the bias was already positive without any aging, implementing the base aging parameterization resulted in an increased fractional bias. Comparison to the 2013 measurements showed modest improvements in both fractional bias and fractional error, as the base case simulation underpredicts OA in this time period.

Using the base case parameterization for biogenic SOA presented in this work, we are able to implement an aging scheme supported by laboratory data that improves model performance in the June 2013 simulation period and produces small changes in performance for the July 2001 simulation period. This new chemical aging scheme allows for an increase in predicted OA concentrations which is consistent with laboratory observations without dramatically overpredicting OA in areas where biogenic SOA is a dominant contributor to total OA concentrations, which has been a persistent issue with chemical transport models in the past. The base case chemical aging parameterization proposed here can be implemented in any CTM using the VBS scheme for the description of secondary OA with a negligible computational cost.

Efforts to parameterize the SOA production from monoterpenes (including the present work) focus on the daytime oxidation and implicitly assume that the nighttime chemistry has a similar behavior. So, for example the branching ratio (Equation (2)) is calculated in the same way during both day and night. The simulation of nighttime production of SOA from monoterpenes is clearly an issue that deserves additional attention and could explain some of the discrepancies between model predictions and observations in continental areas during periods with high monoterpane emissions.

### Table 1

Comparison of predicted OA concentrations to measurements performed at IMPROVE network sites in the Southeastern United States during July 2001 and for the SOAS campaign in June 2013.

<table>
<thead>
<tr>
<th>Simulation Period</th>
<th>No Aging FERROR</th>
<th>Sensitivity FERROR</th>
<th>Base FERROR</th>
<th>No Aging FBIAS</th>
<th>Sensitivity FBIAS</th>
<th>Base FBIAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>July 2001</td>
<td>0.41</td>
<td>0.40</td>
<td>0.40</td>
<td>0.07</td>
<td>0.09</td>
<td>0.13</td>
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<tr>
<td>June 2013</td>
<td>0.39</td>
<td>0.37</td>
<td>0.34</td>
<td>-0.31</td>
<td>-0.27</td>
<td>-0.21</td>
</tr>
</tbody>
</table>
CRediT authorship contribution statement

B. T. Dinkelacker: Software, Formal analysis, Writing – original draft. Spyros N. Pandis: Conceptualization, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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