TITLE

Predicting the evolution of secondary organic aerosol (SOA) size distributions due to limonene ozonolysis in indoor environments

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ABSTRACT

Aerosol diameter is important in determining its fate and transport. Indoors, the ozonolysis of terpenoids, such as limonene, yields secondary organic aerosol (SOA), which can range orders of magnitude in size. The generated SOA size and mass distributions are affected by various processes, including nucleation, partitioning, coagulation, surface deposition, and air exchange; of these processes, only air exchange is not size resolved. Herein, a model was developed to predict the transient evolution of the SOA size distribution indoors due to limonene ozonolysis. The model simulates partitioning without requiring explicit knowledge of the chemical mechanism, because it instead combines the theory of the aerosol mass fraction (AMF = mass SOA formed / mass terpene reacted) with Fick's first law of diffusion to predict the change in aerosol volume from SOA formation. We used experimental results for limonene ozonolysis from our previous work to evaluate and tune the model, and the model performed well according to standard indoor air quality model assessment methods. Its framework can be extended to other terpenoids, and its current form can be used to predict the implications of size resolved SOA formation due to ozonolysis of limonene, the most common indoor terpene. Further, a sensitivity analysis showed that air exchange could be one of the most influential mechanisms to control SOA indoors.

Keywords: Indoor chemistry; terpenes; ozone; aerosol mass fraction (AMF); SOA yields; partitioning

Highlights:

- Secondary organic aerosol (SOA) formation can be a strong particle source indoors
- Formed SOA size distribution influences the SOA removal and inhalation rates
- Model developed to predict SOA size distribution evolution using aerosol mass fraction (AMF)
- Size distributions well predicted for limonene ozonolysis according to model assessment metrics
- Air exchange rate had the most influence on size distribution of explored input variables

1. INTRODUCTION

Studies have demonstrated correlations between exposure to ambient particles and adverse im-

pacts on human health [1]. For instance, increases in the concentrations of particulate matter (PM) can increase mortality [2-12] or incidences of cardiovascular and respiratory diseases [13-15]. Also, exposure to ultrafine particles can damage DNA or other macromolecules [16]. There is also convincing evidence for adverse effects being associated with organic aerosols (OA) specifically [17]. Indoor OA is generated from occupant activities such as cooking [18-20] or smoking [21, 22], and also by secondary organic aerosol (SOA) formation due to oxidation of reactive organic gases (ROG) in indoor air [23, 24]. Furthermore, it has been hypothesized that SOA can form owing to the oxidation of environmental tobacco smoke (ETS) and may lead to certain health effects stemming from ETS exposure [25].

Given the fact that people spent the majority of their time indoors [26] and the reaction rates of SOA precursors are comparable with common indoor air residence times [27], exposure to indoor-formed SOA is likely often meaningful and may cause health concerns. According to Waring [23], most indoor SOA formation is due to ozone (O_3) reactions with reactive organic gases (ROG) such as alkanes. The dominant indoor source of ozone is outdoor-to-indoor transport [28], though it may also be emitted from some appliances and machines [29-31]. The use of consumer products strongly contributes to SOA formation [32-36], because use of those products will emit ROGs such as monoterpenes [37-39]. Monoterpenes ($C_{10}H_{16}$, 136.24 g/mol) are highly reactive due to their one or two unsaturated bonds that may be oxidized by ozone [40], which is a reaction that initiates a complex mechanism that generates products of low volatility that nucleate or partition to form SOA. The monoterpenes of limonene and α -pinene are among the common terpenes used in consumer products [38, 41].

The aerosol mass fraction (AMF) is a metric used to quantify the SOA formation potential of different ROGs, and it is the ratio of the mass of SOA produced to the mass of ROG consumed. The AMF is determined experimentally, and the AMF allows quantification and forecasting of SOA formation without explicitly modeling the myriad reactions that occur due to terpenoid oxidation [42]. SOA formation is mostly an absorptive phenomenon, so AMFs for ROGs span a range of values and increase with organic aerosol (OA) concentrations over 2 to 3 orders of magnitude, depending on the ROG oxidized. Many studies have reported the AMFs of various ROGs [43-54]. For example, unit density AMFs range from

~0.05 to 1.1 for limonene and ~0.01 to 0.35 for α -pinene, over the OA range of ~0.1 to 1000 $\mu\text{g}/\text{m}^3$. The effects of various parameters on SOA formation and the AMF have been explored, such as the air exchange rate [47], temperature [55-57], and humidity [58-61].

The AMF offers no information on the size distribution of produced SOA, and few studies have described the size and number of aerosols formed indoors [62, 63]. However, the diameter of an aerosol plays an important role in defining its fate and transport characteristics, and it additionally determines its rate of lung deposition [64]. The particle diameter is affected by several mechanisms and several studies have addressed dynamics of indoor size distributions including particle emission [65-69], nucleation [70-72], deposition [73-77], coagulation [63, 78-83], air exchange [48, 84-86], and partitioning [87-92]. The partitioning phenomenon is complicated to model since it describes the net mass transfer from the gas to the aerosol phase and requires mechanistic knowledge of the gas phase chemistry, available gaseous products, their mass transfer coefficients, diffusivities, molecular weights, and also transport mechanisms. Relatedly, Ito and Harashima [70] modeled indoor SOA distributions resulting from ozone and limonene reactions in the absence of air exchange, but their model used fitted values for the AMF term, rather than standard partitioning AMF theory as is done in this paper.

In this paper, we focus specifically on simplifying modeling for the dynamic changes in aerosol size distributions due to the partitioning of lower volatility compounds resulting from SOA formation for ozone and monoterpene reactions, using experimental data for model validation. To do so, this work combines our experimentally determined AMF values for limonene ozonolysis in Youssefi and Waring [47] with Fick's first law to predict the distribution of generated SOA mass onto available aerosols as a function of their size. The model also accounts for other indoor relevant processes such as deposition, air exchange, and coagulation mechanisms. With the developed framework, future researchers can predict the evolving size distribution due to indoor SOA formation as well as the fate of that aerosol as it interacts with building control mechanisms and is inhaled by building occupants.

2. METHODOLOGY

2.1. *Experimental inputs to size distribution evolution model as initial conditions*

For model development and validation, the transient SOA formation experiments for limonene ozonolysis from our article focusing on mass formation and AMF quantification, Youssefi and Waring [47] were used. The experimental procedure and apparatus are explained in detail therein (Figure 1 in that article), but both are described briefly here. In a 1-m³ stainless steel chamber operated as a well-mixed, continuous flow mixed reactor (CMFR), with air at a mean temperature of 23 °C and low relative humidity (RH) and initially free from ROGs and aerosols, ozone was generated (Absolute Ozone NANO) until it reached a desired concentration. Then, the limonene was injected with a syringe pump for 30 s at a mass amount that would yield its desired concentration. Once the reactions commenced, neither reactant was replenished. The size distributions from 5.6 to 560 nm of the SOA were measured in one-minute intervals by using a Fast Mobility Particle Sizer (FMPS, TSI 3091), while ozone (1 min averages, 2B Technologies 205) and limonene concentrations (~20 min, GC-FID, SRI Instruments) were also measured.

SOA formation in these transient experiments may be coarsely divided into three main stages, as in Figure 1. The first is the *aerosol nucleation* stage, where many SOA clusters are formed initially due to the ozone and terpene reactions. At the end of this relatively short stage, the SOA number reaches a peak value. Nucleation increases the peak value, while coagulation acts to restrict the peak number that may be reached, since its strength increases with aerosol number. Other mechanisms such as deposition and air exchange are active in this stage, as well as the following two stages. The second stage is the *aerosol partitioning* stage, during which ozone and terpene reaction products nucleate less strongly but instead partition to existing aerosols in a dominant way. Coagulation may still act until number decreases sufficiently. In the third and final stage, which is the *aerosol loss* stage, the reaction rate has decreased so that formation is less impactful than the mass loss mechanisms of deposition and air exchange.

Figure 1. SOA formation for Experiment E5, owing to a pulse injection of ozone (195 ppb) and limonene (30 ppb) and resulting transient reactions inside the stainless steel chamber at an air exchange rate, AER = 0.53 h^{-1} . The marked stages of aerosol behavior are (1) aerosol nucleation, (2) aerosol growth, and (3) aerosol loss.

Though nucleation generates the initial aerosol size distribution, this model does not attempt to predict it theoretically due to its complexity and unpredictability. Also, nucleation is broadly studied elsewhere [70-72]. As such, the initial condition of the size evolution model is right at the point that the nucleation mechanism generated a peak number with a size distribution of certain geometric mean (GM) diameter and geometric standard deviation (GSD). Table 1 reproduces the conditions for the 18 chamber experiments from Youssefi and Waring [47], including the air exchange rates (AER), initial concentrations of ozone and limonene, and the size distribution characteristics of the peak number, GM, and GSD. Also, deposition is not theoretically modeled, but instead experimental deposition rates for each size bin, which were determined once SOA formation ceased [47], were used. For reference, the integrated SOA mass deposition loss rates are listed in Table 1.

Table 1. Experiment conditions for secondary organic aerosol (SOA) formation due to limonene ozonolysis from Youssefi and Waring [47].

Exp. ID ^a	AER ^b (h ⁻¹)	Initial reactants		β_{SOA}^c (h ⁻¹)	Peak size distribution ^d		
		Ozone (ppb)	Limonene (ppb)		Number (#/cm ³)	GM ^e (nm)	GSD ^e (-)
E1	0.53	45	422	0.15	317,901	37	1.5
E2	0.52	45	119	0.32	287,805	31	1.6
E3	0.52	45	67	0.28	107,318	35	1.5
E4	0.53	295	20	0.19	148,867	40	1.7
E5	0.53	195	30	0.25	165,180	46	1.4
E6	0.53	94	35	0.32	107,527	50	1.4
E7	0.26	41	428	0.43	137,165	65	1.5
E8	0.27	46	275	0.36	92,918	65	1.5
E9	0.31	45	118	0.34	154,923	35	1.5
E10	0.28	300	243	0.23	517,218	70	1.5
E11	0.3	192	72	0.18	144,500	67	1.5
E12	0.26	95	122	0.21	162,059	55	1.5
E13	0.95	45	663	0.47	85,686	41	1.5
E14	0.95	45	131	0.10	23,122	72	1.4
E15	0.96	45	67	0.45	27,904	55	1.5
E16	0.96	295	58	0.16	264,731	35	1.5
E17	0.95	195	34	0.18	99,172	37	1.5
E18	0.97	95	42	0.24	70,967	37	1.5

^aExperimental identification label

^bAER = air exchange rate

^c β_{SOA} = SOA surface deposition rate

^dPeak size distribution used as initial condition for size distribution model

^eGM = geometric mean; GSD = geometric standard deviation

2.2. Size distribution evolution model

When considering the mechanisms of coagulation, partitioning, air exchange, and surface deposition, the governing equation is a form of the continuous general dynamic equation is:

$$\begin{aligned} \frac{\partial n(v, t)}{\partial t} = & \frac{1}{2} \int_{v_0}^{v-v_0} K(v-v_0, v_0) n(v-v_0, t) n(v_0, t) dv_0 \\ & - \int_{v_0}^{\infty} K(v, v_0) n(v, t) n(v_0, t) dv_0 - \frac{\partial}{\partial v} I(v, t) n(v, t) \\ & - \lambda n(v, t) - \beta_{SOA}(v) n(v, t) \end{aligned} \quad (1)$$

where $n(v, t)$ (m⁻³) is the number of particles with volume (m³) within v and $(v + dv)$ at time t (s); v_0 and $(v - v_0)$ are volumes of two colliding particles and v is the volume of the resulting particle; K (m³/s) is the coagulation kernel, and it depends on the volumes of the two colliding particles; I (m³/s) is the condensation rate, and it depends on the particle volume and also gaseous compound characteristics such as diffu-

sivity and molecular weight; λ (s^{-1}) is the air exchange rate; and β_{SOA} (s^{-1}) is the particle deposition rate to surfaces, and it depends on the volume of the particle, as well as surface and airflow characteristics.

The left hand side of Equation 1 is the time rate of the change in the number of particles of volume v . On the right hand side, the first and the second terms correspond to coagulation gains and losses, respectively; the third term is partitioning due to net condensation; the fourth term is air exchange loss; and the last term is to particle loss due to surface deposition. As explained above, Equation 1 was solved with the following initial condition: $n(v, 0) = n_{\text{max}}(v)$, where $n_{\text{max}}(v)$ (m^{-3}) is the peak SOA number concentration that is represented by a measured lognormal size distribution with a particular peak number, GM, and GSD, as listed in Table 1 for the 18 experiments.

2.3. Numerical solution to size distribution evolution model

To solve the integro-differential equation, it was discretized using a semiimplicit method [93], for which the state of the system at a current time is obtained by using the state of the system at the previous time. The integro-differential equation is valid for ‘monomer bin distribution,’ where the volume in bin k is the volume in the smallest bin multiplied by k . Such a distribution requires a large number of bins and is computationally intensive; the model range of 5.6 to 560 nm would require 10^6 bins. So instead, this work used a ‘volume-ratio bin distribution,’ in which the volume in the next bin is defined by multiplying the volume in the previous bin by a constant ‘volume ratio,’ VR [93]. Smaller ratios lead to more bins, which increases accuracy. Further, for monomer bin distributions, when a particle grows due to coagulation or partitioning, the resulting particle will have a certain volume corresponding to only one bin size. However, for volume-ratio bin distributions, the resulting particle may have a volume that can be distributed between two bins (discussed more below).

When Equation 1 was discretized, it was also modified to represent change in the volume of particles, rather than the particle number, as follows:

$$\begin{aligned}
\frac{V_{k,t} - V_{k,(t-\Delta t)}}{\Delta t} = & \sum_{j=1}^k \sum_{i=1}^{k-1} f_{i,j,k} K_{i,j} V_{i,t} n_{j,(t-\Delta t)} \\
& - \sum_{j=1}^{N_B} (1 - f_{k,j,k}) K_{k,j} V_{k,(t-\Delta t)} n_{j,(t-\Delta t)} \\
& + \sum_{i=1}^{k-1} \frac{n_{i,t} v_k}{\Delta t} g_{i,k,t} - \frac{n_{k,t} v_k}{\Delta t} (1 - g_{k,k,t}) - (\lambda + \beta_{\text{SOA},k}) V_{k,(t-\Delta t)}
\end{aligned} \tag{2}$$

where t and $(t - \Delta t)$ are notations for current and previous time steps, respectively, which apply to some variables; Δt (s) is the time step duration; $V_{k,t}$ (m^3/m^3) and $V_{i,t}$ (m^3/m^3) are volume concentrations of particles in bins k or i , respectively; v_k (m^3) is the volume of a single particle in bin k ; $n_{j,(t-\Delta t)}$ (m^{-3}) and $n_{k,t}$ (m^{-3}) are number concentrations of particles in bins j and k , respectively; $K_{i,j}$ (m^3/s) is the coagulation coefficient between two colliding particles from bins i and j ; $\beta_{\text{SOA},k}$ (s^{-1}) is the deposition rate of SOA for bin k ; and N_B is the total number of bins. The f and g terms are described next.

On the right side of the equality in Equation 2, the first two terms represent gain and loss owing to coagulation, respectively. Since Equation 2 uses a volume-ratio distribution, the particle resulting from partitioning or coagulation of other particles may not have a volume equal to any defined bin and it may be distributed between upper and lower bins. Therefore, $f_{i,j,k}$ is a coefficient that can be between 0 and 1 and describes the volume fraction of particles in bins i and j that coagulate into bin k , while $f_{k,j,k}$ similarly describes the fraction in bins k and j that coagulate and remain in bin k . The next two terms represent gain and loss owing to partitioning, respectively. The gain due to partitioning in bin k is the sum of the fraction of particles from smaller bins (from bin $1 \rightarrow (k - 1)$) that grow due to partitioning and enter that bin. The loss rate from bin k due to partitioning is the sum of particles in bin k that grow and leave bin k and will move to bins bigger than bin k . Similar to $f_{i,j,k}$ and $f_{k,j,k}$ coefficients, $g_{j,k,t}$ is a coefficient that describes the volume fraction of particles in bin j that will grow to bin k due to partitioning at time t , while $g_{j,k,t}$ similarly describes the fraction that will grow out of bin k into bin j due to partitioning at time t .

As an example, two of the volume fractions are illustrated [93]. The first is the coagulation volume fraction: when two particles from bins i and j collide, the resulting particle will have a volume of $v_{i,j}$ and $f_{i,j,k}$, the coagulation volume fraction for bins i and j that coagulate into bin k is:

$$f_{i,j,k} = \begin{cases} \left(\frac{v_{k+1} - v_{i,j}}{v_{k+1} - v_k} \right) \frac{v_k}{v_{i,j}}, & v_k \leq v_{i,j} \leq v_{k+1} & k < N_B \\ 1 - f_{i,j,k-1}, & v_{k-1} \leq v_{i,j} \leq v_k & k > 1 \\ 1, & v_k \leq v_{i,j} & k = N_B \\ 0, & \text{otherwise} \end{cases} \quad (3)$$

The second is $g_{j,k,t}$, the partitioning volume fraction for bin k that results from the growth of particles in bin j at time t . Assuming that $dv_{j,t}$ is the volume that partitions on a single particle in bin j during the time span dt , the resulting particle will have volume of $vdv_{j,t} = v_j + dv_{j,t}$, then $g_{j,k,t}$ is:

$$g_{j,k,t} = \begin{cases} \left(\frac{v_{k+1} - vdv_{j,t}}{v_{k+1} - v_k} \right) \frac{v_k}{vdv_{j,t}}, & v_k \leq vdv_{j,t} \leq v_{k+1} & k < N_B \\ 1 - g_{j,k-1,t}, & v_{k-1} \leq vdv_{j,t} \leq v_k & k > 1 \\ 1, & v_k \leq vdv_{j,t} & k = N_B \\ 0, & \text{otherwise} \end{cases} \quad (4)$$

It should be noted that unlike the coagulation volume fractions ($f_{i,j,k}$), which are constant during the experiment, the partitioning volume fractions ($g_{j,k,t}$) change as the rate of SOA formation (i.e., dv/dt) changes.

2.4. Simulating coagulation process

The coagulation kernel, K , varies by the diameters of colliding particles, and it is potentially affected by several mechanisms, including Brownian motion, van der Waals forces, and viscous forces [63, 93]. For particles larger than 10 nm, considering only the Brownian motion component results in coagulation rates supported by experimental data [94]. However, for particles smaller than 10 nm, other mechanisms besides Brownian motion are accounted for in calculating the coagulation kernel [63, 95]. Therefore, since the SOA range in this study is 5.6 to 560 nm, the inclusion of Brownian motion along with the Fuchs correction as well as van der Waals forces in estimating the coagulation kernel was evaluated.

The Hamaker constant, A (J), indicates the strength of binding attraction between two colliding particles due to van der Waals forces [93], and it ranges from $(6-150) \times 10^{-20}$ J [64]. Rim et al. [63] sug-

gested $A = 100k_B T$ and $200k_B T$ for organic particles resulting from gas and electric stove operation, where k_B is the Boltzmann constant (1.38×10^{-23} J/K) and T (K) is absolute temperature. Three coagulation models were evaluated: (1) Brownian motion with Fuchs correction (2) Brownian motion with Fuchs correction and van der Waals and viscous forces with $A = 100k_B T$ and (3) Brownian motion with Fuchs correction and van der Waals and viscous forces with $A = 200k_B T$. The coagulation kernel contour plot for the first case with Brownian motion only is shown in Figure 2.

Figure 2. Coagulation kernel for relevant particle diameter (D_p) range considering Brownian motion.

2.5. Simulating partitioning process

For the partitioning element, it was necessary to determine $dv_{j,t}/dt$, which is the rate of the transfer of condensable material from the gas to particle phase for bin j at time t . One practice is to use Fick's first law of diffusion for individual SOA precursors, and doing so requires detailed knowledge of the complex chemistry that produces those precursors, as well as their individual vapor pressures, diffusivities, and molecular weights. However, one can assume the mixture of SOA precursors is a hypothetical single compound and write Fick's first law for a single particle with diameter $d_{p,j}$ in bin j , as in Equation 5:

$$\frac{dv_j}{dt} = \frac{2\pi d_{p,j} DM}{\rho RT} f(Kn, \alpha) (P - P_{eq}) \quad (5)$$

where dv_j/dt (m^3/s) is the volumetric transfer rate of SOA precursors from the gas to particle phase for a single particle in bin j ; D (m^2/s) is the diffusion coefficient of the SOA precursor in air; M (kg/kmol) and ρ (kg/ m^3) are its molecular weight and density; and $f(Kn, \alpha)$ is the correction due to non-continuum ef-

fects and imperfect surface accommodation. The difference between P (Pa), the vapor pressure of SOA precursors far from the particle, and P_{eq} (Pa), the equilibrium vapor pressure, is the driving force ($P - P_{eq}$) for the transport of the SOA precursor from the gas to particle phase.

While dv_j/dt (m^3/m^3s) is the volumetric transfer rate of SOA precursors from the gas to a particle in bin j , the total volumetric transfer rate of SOA precursor of all particles in bin j , dV_j/dt (m^3/m^3s), is the product of the number of particles in bin j , $n_{j,t}$, and dv_j/dt . Also, the total volumetric transfer rate of SOA precursors to particle phase, dV/dt (m^3/m^3s), is the sum of volumetric transfer rate of SOA precursors to each bin ($j: 1 \rightarrow N_B$) as follows:

$$\frac{dV}{dt} = \sum_{j=1}^{N_B} \frac{dV_j}{dt} = \sum_{j=1}^{N_B} n_{j,t} \frac{dv_j}{dt} \quad (6)$$

By replacing dv_j/dt in Equation 6 by its equivalent from Equation 5, one has the total partitioning rate, determined from Fick's first law:

$$\frac{dV}{dt} = \sum_{j=1}^{N_B} n_{j,t} \frac{2\pi d_{p,j} DM}{\rho RT} f(Kn, \alpha) (P - P_{eq}) = \frac{2\pi DM}{\rho RT} (P - P_{eq}) \sum_{j=1}^{N_B} n_{j,t} d_{p,j} f(Kn, \alpha) \quad (7)$$

Furthermore, to eliminate the need for the knowledge of gas phase chemistry and SOA precursor physico-chemical parameters to find D , M , P , P_{eq} and ρ , one can use the well-constrained aerosol mass fraction (AMF) model framework to determine dV/dt , as follows:

$$\frac{dV}{dt} = \frac{AMF \cdot k_{O_3-terp} C_{O_3,t} C_{terp,t} \Gamma}{\rho_{SOA}} \quad (8)$$

where $AMF = (\text{mass formed SOA} / \text{mass reacted terpene})$; k_{O_3-terp} ($ppb^{-1} s^{-1}$) is the reaction rate constant of ozone and the terpene; $C_{O_3,t}$ and $C_{terp,t}$ (ppb) are the ozone and terpene concentrations at time t ; ρ_{SOA} is the SOA density (kg/m^3); and Γ is a temperature-dependent conversion factor to change from ppb to kg/m^3 for the terpene. For the limonene AMF, this work used the empirical formulations that were developed in our previous paper for limonene ozonolysis [47].

The left hand sides of Equations 7 and 8 are similar. Therefore, equating the total amount of partitioning rate determined from the AMF framework (Equation 8) to the one determined from Fick's first law (Equation 7) results in:

$$\frac{2\pi DM}{\rho RT} (P - P_{eq}) = \frac{AMF \cdot k_{O3-terp} C_{O3,t} C_{terp,t} \Gamma}{\rho_{SOA} \sum_{j=1}^{N_B} n_{j,t} d_{p,j} f(Kn, \alpha)} \quad (9)$$

By substituting the right side of Equation 9 into Equation 5 and assuming a unit density ($\rho_{SOA} = 1 \text{ g/cm}^3$), one can solve for the volumetric transfer rate of condensable material that partitions to a single particle with diameter $d_{p,j}$ at time t as follows:

$$\frac{dv_j}{dt} = \frac{d_{p,j} \cdot f(Kn, \alpha)}{\sum_{j=1}^{N_B} n_{j,t} \cdot d_{p,j} \cdot f(Kn, \alpha)} \cdot AMF \cdot k_{O3-terp} C_{O3,t} C_{terp,t} \Gamma \quad (10)$$

Unlike Equation 5, which is for individual precursors that may be unknown and require knowledge of variables such as D , M , and P_{eq} , all parameters in Equation 10, including relevant particle diameters, ozone and terpene concentrations, AMF values, and $f(Kn, \alpha)$, are known, either by theoretical (i.e., $f(Kn, \alpha)$) or experimental or predictable (i.e., AMF, $k_{O3-terp}$, $C_{O3,t}$ and $C_{terp,t}$) means.

To find $f(Kn, \alpha)$, one can use the equation suggested by Fuchs [96], as follows:

$$f(Kn, \alpha) = \frac{0.75 \alpha (1 + Kn)}{0.75 \alpha + Kn + Kn^2} \quad (11)$$

As mentioned, $f(Kn, \alpha)$ is the correction due to non-continuum effects and imperfect surface accommodation, and it depends on interplay of the particle's diameter and its mean free path. When the diameter of a diffusing particle is bigger than the mean free path of the air, the air may be treated as a continuum. In other words, the diffusing particle is surrounded completely by air molecules, and for every movement the particle makes, it encounters air molecules. The accommodation factor, α , can have a value between 0 and 1, and it is assumed as $\alpha = 1$, which is realistic for organic aerosols [97]. A value of 1 indicates that when a condensable molecule from gas phase encounters the aerosol surface, it sticks to the surface with a probability of sticking equal to unity.

Furthermore, according to Equation 10, the distribution of gas phase compounds to the aerosol phase proceeds as a function to the aerosol diameters. In order to improve the model performance, it was tested at different values considering a wholly empirical power term, x , applied to the particle diameter in Equation 10, i.e., as in d_p^x . Though this alteration is admittedly a model tuning parameter, it is loosely

grounded in a physical concept of the changing strength of the condensation mechanism as scaling with particle size (i.e., $x = 1$), surface area ($x = 2$), or volume ($x = 3$).

3. RESULTS AND DISCUSSION

3.1. Setting the volume ratio

MATLAB was used to run all modeling, and the data from the previous limonene ozonolysis chamber experiments [47] were used to evaluate and tune the model. However, first the volume ratio (VR) for the numerical solution to solve the integro-differential equation had to be determined. As discussed previously, smaller VRs lead to more bins, increasing accuracy; however, smaller VRs and more bins require more computational power. Jacobson [93] reported that using a VR = 1.2 to numerically solve the general dynamic equation for particle coagulation only resulted in a size distribution comparable to analytical solutions for coagulation. However, the model herein includes other mechanisms, so to find a reasonable VR for our solution, cases were run with various values of VR, and the smallest one for which the solution did not change appreciably with the next VR reduction was chosen.

The impact of changing VRs on the predicted SOA volume distribution for each of the experiments was explored, starting with VR = 2 and decreasing down to VR = 1 (results not shown). The VR = 1.2 as suggested by Jacobson [93] resulted in 76 bins. As the VR decreased, the SOA volume distribution results continued to change until VR = 1.05, and after that, using a VR = 1.04 yielded no appreciable change in the model results. However, the VR = 1.04 and VR = 1.05 correspond to large differences in the number of discrete bins of 353 and 284 bins, respectively, and much more necessary computing power for the lower VR. As such, the VR = 1.05 was chosen for all subsequent runs and analysis.

3.2. Coagulation models

The three coagulation models, a) Brownian, b) Brownian and van der Waals with Hamaker constant, $A = 100k_B T$ and c) Brownian and van der Waals with $A = 200k_B T$, gave similar results. Therefore, including van der Waals and viscous forces in the coagulation coefficient calculation did not change the results significantly. Van der Waals and viscous forces are very influential on coagulation mechanism in particles smaller than 10 nm [63, 95]. While the SOA size range in the experiments was 5.6 to 560 nm,

except for the very first few minutes of each experiment, available SOA diameter sizes were larger than 24 nm. Therefore, it is reasonable that van der Waals and viscous forces would not affect the model prediction for the range of SOA in this set of model runs.

3.3. Model tuning

With the $VR = 1.05$, the model was tuned and its performance evaluated. First, the model was run for each of the 18 experiments without tuning, so that $x = 1$ for d_p^x . (Note that the model calculated volume distributions, but they were converted to size distributions for model evaluation.) Generally, the untuned model well predicted the time-evolution of the SOA number, but as time progressed the GM of the predicted SOA size distributions were larger than the measured ones and GSD were smaller. The SOA mass formed was also in good agreement, as expected, since the AMFs were good mass predictors in Youssefi and Waring [47]. As discussed earlier, the partitioning mechanism does not affect the particle number, and it only affects particle size (thus the GM and GSD). Therefore, since the time evolution of total number of particles was in good agreement with the experimental values and it was the size of the particles that differed, it was hypothesized that the experiments were not as well represented by the method of distributing the condensable products in the model, i.e., that of Equation 10.

To improve the model performance, different values of the power term, x , applied to the particle diameter in Equation 8, i.e., as in d_p^x , were tested. This tuning power was optimized to best recreate the experimental data, and Figure 3 shows examples, along with the overall best estimate of $x = 2.8$ for Experiment E2. That is, Figure 3(a) illustrates experimental data, while Figures 3(b) to 3(e) illustrate the model results for cases where the power was, $x = 1, 2, 2.8$, and 3, respectively. Additionally, Figure 3(f) demonstrates experimental and predicted GMs for these cases. As the power x increases, the GM of the size distribution decreases. Moreover, increasing x results in larger GSDs, so as x increases, the distributions spread out. The power $x = 2.8$ was ultimately chosen as the best estimate for the tuning parameter, because it yielded the best agreement between real and modeled data, over all 18 experiments.

Figure 3. Effect of the tuning power on model results: (a) Contour plot of E2 experimental data; contour plot of model results with (b) power = 1, (c) power = 2, (d) power = 2.8, (e) power = 3; and (f) experimental geometric mean (GM) and modeled results of GM for the different tuning powers.

As an example of the agreement for other experiments using this optimized power, Figure 4 demonstrates the results with $x = 2.8$ for modeled and measured results, for three different experiments other than Experiment E2. Specifically, it shows results for one experiment performed at each of the three air exchange rate (AER) conditions, including the moderate AER = 0.53 h^{-1} (Experiment E5), low AER = 0.28 h^{-1} (Experiment E11), and high AER = 0.96 h^{-1} (Experiment E17). For these three cases, the initial concentrations of reactants were similar (see Table 1). As shown in Figure 4, at all three AER conditions, the model very well reproduced the experimental trends. In the next section, the evaluation of model performance from a statistical standpoint is discussed.

Figure 4. Model results vs experimental results for (a-c) E5, (d-f) E11, and (g-i) E17, for the SOA number, mass, and size-distribution GM, respectively. Solid lines represent experimental results and dashed lines are model results.

3.4. Statistical evaluation of tuned model

Over all experiments, statistical tests were used to quantify the model performance with $VR = 1.05$ and the tuning power $\alpha = 2.8$. Particularly, performance was quantified following the guidance of ASTM D5157-97 [98], which recommends evaluating predictive performance using the metrics of the normalized mean square error (NMSE) and fractional bias (FB).

The NMSE is a measure of the variation in the predicted values by the model and the observed values in an experiment. For classification of acceptable model performance, the NMSE must be smaller than 0.25 according to ASTM D5157-97 [98]. For the 18 experiments, the NMSE was determined for the SOA number and mass concentrations, as well as the GM and GSD of size distributions, using the real

and predicted values at each minute of the experiment, and these resulting NMSE are listed in Table 2. Number and mass concentrations mean values of NMSE were 0.047 and 0.010, respectively, and ranges were small, at NMSE of 0 to 0.169 for number concentration predictions and NMSE of 0.001 to 0.073 for mass concentrations. For the GM, the mean NMSE was 0.004, and it ranged from 0 to 0.013. For the GSD, the mean NMSE was 0.007, and it ranged from 0 to 0.021. Therefore, as the means and ranges of the NMSE values show, the tuned model is capable of predicting size evolution within an acceptable proximity to the observed experimental values.

Table 2. Model performance: Normalized mean squared error (NMSE) and fractional bias (FB) values for modeled versus measured results for the 18 limonene experiments.

Exp. ID	Number Concentration		Mass Concentration		Geometric Mean		Geometric Standard Deviation	
	NMSE	FB	NMSE	FB	NMSE	FB	NMSE	FB
E1	0.011	-0.015	0.015	-0.081	0.004	-0.007	0.001	0.013
E2	0.036	-0.123	0.002	0.038	0.005	-0.006	0.017	0.126
E3	0.045	-0.194	0.005	0.006	0.002	0.002	0.014	0.114
E4	0.140	-0.150	0.004	-0.007	0.004	-0.012	0.021	0.072
E5	0.077	-0.114	0.011	0.035	0.005	0.009	0.010	0.045
E6	0.097	-0.009	0.002	0.110	0.001	-0.005	0.013	0.051
E7	0.045	-0.097	0.002	0.009	0.008	0.040	0.001	-0.008
E8	0.022	-0.060	0.018	0.012	0.007	0.041	0.002	0.015
E9	0.086	-0.031	0.007	-0.021	0.004	-0.009	0.004	0.058
E10	0.060	0.052	0.006	-0.044	0.006	0.005	0.000	0.000
E11	0.017	-0.073	0.003	0.027	0.002	0.027	0.001	0.014
E12	0.169	0.262	0.014	0.085	0.013	-0.094	0.005	0.049
E13	0.008	-0.087	0.073	0.190	0.004	-0.014	0.012	0.108
E14	0.001	-0.017	0.001	-0.010	0.001	-0.020	0.005	0.058
E15	0.029	-0.163	0.006	-0.061	0.001	-0.031	0.003	0.047
E16	0.014	-0.105	0.014	0.068	0.003	0.011	0.006	0.074
E17	0.000	-0.008	0.002	0.004	0.001	-0.020	0.001	0.008
E18	0.006	-0.068	0.002	-0.031	0.000	-0.007	0.001	0.009

To quantify systematic bias in the model predictions, ASTM D5157-97 [98] recommends using the non-dimensional fractional bias (FB). Its full allowable range is between -2 and 2 , and when the predicted and observed values agree perfectly (i.e., an ideal mode), the $FB = 0$. Larger negative values of the FB indicate that the model under predicts results, while positive values are an indication of over prediction. Table 2 also lists the FB results for number and mass concentrations, as well as for the GM and GSD

of the size distributions. For number and mass concentrations, the means of FB were 0.055 and 0.018 respectively, and they ranged from -0.194 to 0.262 for the number concentration and from -0.081 to 0.190 for the mass concentration. The mean FB for the GM was -0.005 , and it ranged from -0.095 to 0.041 . For the GSD, the mean FB was 0.047 , and it ranged from -0.008 to 0.126 . As these values show, the model has little systematic bias, except perhaps slightly for the number and GSD. Moreover, except for Experiment E7, with $FB = -0.008$, all other FB for the GSD are positive. Although, these values are small, it indicates that the model slightly over predicts the GSD for SOA formation.

3.5. Influence of inputs on size distribution

The importance of various drivers on the resulting SOA size distribution was explored. First off, it was noted that the AER had a substantial impact on the size distribution evolution over time. Over all experiments on average, at low AERs (0.28 h^{-1}), the mechanisms of surface deposition, air exchange, and coagulation had similar impacts on SOA number, at 33% each. For medium AERs (0.53 h^{-1}), the AER impact increased and 50% of SOA number was lost due to air exchange, while surface deposition and coagulation each contributed to 20% and 30% of SOA number loss. At the high AER (0.96 h^{-1}), 69% of SOA number was lost due to air exchange, and surface deposition and coagulation were responsible for 17% and 14% of SOA loss. (In all experiments, coagulation was more impactful at the beginning of an experiment, since there was the most SOA number in the chamber at that time.) Thus, for all experiments, the AER was equal to or greater than other loss mechanisms, showing that ventilation is an effective mechanism to decrease and control the levels of formed SOA indoors.

Furthermore, a sensitivity analysis was performed to study the impacts of underlying parameters on the SOA distribution, including the AER, initial ozone and limonene concentrations, and the friction velocity, u^* (cm/s). Specifically, u^* is an input to calculate the size-resolved surface deposition according to the theory of Lai and Nazaroff [73], where a higher u^* results in more turbulence, a thinner boundary layer, and more aerosol deposition at smaller diameter size ranges. Thus, u^* was varied as an input in that deposition model and the sensitivity analysis to explore the deposition impact. First, the model was run with the expected values of inputs to define a base case and predict SOA formation for those values.

Then, each parameter was varied individually to high and low values, and the differences in SOA formation were quantified owing to each parameter change (9 total runs). The lower bound, expected value, and upper bound of the parameters used in this sensitivity analysis are listed in Table 3. These AER, reactant concentrations, and u^* expected values and upper and lower bounds are typical for residences.

For the limonene AMF, this work used a multiple linear regression ($R^2 = 0.83$) to AMF parameters with predictors of the AER and the initial ozone-limonene product and ratio from Youssefi and Waring [47]. Since the model does not address nucleation and takes the peak size distribution as an initial condition, this work also used the experimental data (in Table 1) from Youssefi and Waring [47] to set the initial (peak) SOA size distribution for the 9 runs, as follows. For peak number, the model used a multiple linear regression generated with peak values in Table 1 as the outcome, with predictor variables being similar ones chosen for AMF parameter regressions in Youssefi and Waring [47], i.e., the AER and the product and ratio of the initial ozone, $C_{O_3,t=0}$ (ppb), and limonene, $C_{lim,t=0}$ (ppb), concentrations:

$$N_{t=0} = 170342 - 134437 (\text{AER}) + 5.04(C_{O_3,t=0}C_{lim,t=0}) + 4873 \left(\frac{C_{O_3,t=0}}{C_{lim,t=0}} \right) \quad (12)$$

where $N_{t=0}$ (cm^{-3}) is the predicted initial, peak SOA number (regression $R^2 = 0.62$). The initial GM was the average of the GMs for each AER condition in Table 1, which were 59, 40, and 46 nm at low, medium, and high AERs, respectively. The initial GSD was set at 1.5 for all runs.

Table 3. Expected value and lower and upper bounds of parameters varied in the sensitivity analysis.

Parameter	Lower bound	Expected value	Upper bound
Air exchange rate, AER ^a (h^{-1})	0.3	0.5	1
Ozone concentration ^b (ppb)	10	25	50
Limonene concentration ^c (ppb)	100	200	300
Friction velocity ^d (cm/s)	0.3	1	3

a. Chosen to roughly mimic 10th, 50th, and 90th percentiles of AER distribution from Murray and Burmaster [99], but preserve the AER input range used to generate the regressions.

b. Typical-to-high range due to outdoor-to-indoor transport [23]

c. Typical range of resulting concentrations from consumer product use [38]

d. Typical range indoors [73]

The results of the sensitivity analysis are in Table 4 and graphically in Figure 5, where the bars show the absolute change from the base case for peak SOA mass and number (associated with the peak

mass) for each variable at its low and upper bounds. The AER had the highest effect on the SOA mass and number concentrations. While base case concentrations were $26 \mu\text{g}/\text{m}^3$ and $78,000 \text{ \#}/\text{cm}^3$, doubling the AER from 0.5 to 1.0 h^{-1} reduced mass and number to $10 \mu\text{g}/\text{m}^3$ and $56,000 \text{ \#}/\text{cm}^3$, corresponding to percent differences (calculated as $[\text{difference}/\text{mean}]$) of -89% and -33% , respectively. Conversely, decreasing the AER to 0.3 h^{-1} increases SOA formation to $61 \mu\text{g}/\text{m}^3$ and $102,000 \text{ \#}/\text{cm}^3$, corresponding to percent differences of 81% and 27% , respectively. Varying the other airflow parameter, the friction velocity, resulted in little change from the base case for the explored range. The largest change occurred when u^* increased from 1 to $3 \text{ cm}/\text{s}$, reducing SOA number by a small percent difference of -2.6% . Further, it should be noted that in this work, the particle removal rate as a result of air exchange was not influenced by particle size. However, in buildings and realistic environments where also air exchange and penetration occur through air handling units and envelope leaks, air exchange is a size-influenced process and introduces particles [23, 100-107] that will have an impact on the results. Also, filtration modifies indoor PM concentrations, but its impact was also neglected in this work [108].

Table 4. Inputs and outcomes of sensitivity analysis. Base case is listed in bold on the first row, then high and low changes are listed for: AER, ozone, limonene, and friction velocity (changed are shaded).

Initial conditions for sensitivity analysis ^a				Outcomes of sensitivity analysis ^b			
AER ^c (h^{-1})	Ozone (ppb)	Limonene (ppb)	Friction velocity (cm/s)	Peak mass ($\mu\text{g}/\text{m}^3$)	Number ($\text{\#}/\text{cm}^3$)	GM ^d (nm)	GSD ^d (-)
0.5	25	200	1	26.2	77,797	65.2	1.6
1.0	25	200	1	10.3	55,964	54.3	1.5
0.3	25	200	1	60.5	102,262	81.7	1.5
0.5	50	200	1	48.3	88,177	76.6	1.6
0.5	10	200	1	13.4	68,481	55.1	1.5
0.5	25	300	1	30	91,936	64.4	1.6
0.5	25	100	1	19.6	63,963	63.5	1.6
0.5	25	200	3	25.7	75,623	65.6	1.6
0.5	25	200	0.3	26.3	79,006	64.9	1.6

- Starting from the base case, parameters were uniquely varied to their high and low values
- All outcomes are for predicted peak SOA mass concentration
- AER = air exchange rate
- GM = geometric mean; GSD = geometric standard deviation

Varying ozone and limonene each influenced the results substantially, with an interesting trend. Changing both reactants affects the SOA number concentration at similar magnitudes, with mean absolute percent differences of the high and low parameter variations being 13% and 18% for ozone and limonene, respectively. However, for mass, changing the ozone had a much larger impact, with mean absolute percent differences of 63% and 20% for ozone and limonene, respectively. Also, increasing ozone yielded the second highest GM of varied cases. This trend occurs because of the manner in which limonene reacts with ozone. Briefly, limonene is a cyclic monoterpene with two unsaturated bonds, of which one is endo-cyclic and one is exo-cyclic. The endo bond is 10–50× more likely to react with ozone [50]. Yet as the ozone-to-limonene ratio increases, the exo- bond may also be oxidized, likely within the aerosol phase, resulting in higher AMFs. This relationship was quantified by Youssefi and Waring [47] in their empirical AMF fits, so it is also embodied within this model.

Figure 5. Results of sensitivity analysis, for both SOA number and mass at the peak mass concentration predicted by the size evolution model. Plot shows impact on SOA formation of changing each parameter from expected values to upper and lower bound values.

4. CONCLUSIONS

In this paper, a model was developed to predict the SOA size distribution evolution owing to ozonolysis of limonene in indoor settings with air exchange. The focus was developing a method to predict

SOA formation due to partitioning, and as such, the peak SOA nucleated size distribution was included as an initial condition to the model. For the partitioning mechanism, this work used the standard framework to predict SOA mass formation of the aerosol mass fraction ($AMF = \text{mass SOA formed} / \text{mass terpenoid reacted}$). The AMF framework was successfully integrated into a numerical solution to predict changes in aerosol volume by aerosol size. This integration is advantageous because the AMF framework is well grounded in the literature, so this method provides a practical and tractable solution to predict the evolution of the SOA size distribution. Though nucleation is treated as an initial condition, the model explicitly includes other relevant mechanisms that influence the size distribution, including coagulation, partitioning, air exchange, and surface deposition.

Experimental data from Youssefi and Waring [47] for 18 chamber experiments that quantified SOA formation due to limonene ozonolysis was used to refine and then evaluate the performance of the optimized model. Good agreement between modeled and measured results was obtained by tuning one parameter, which was an introduced power for the particle diameter in Equation 8, which predicted the change in volume of a single particle due to partitioning (using the AMF). To evaluate the model performance, this work followed the ASTM D5157-97 [98] method and determined the normalized mean square error (NMSE) and fractional bias (FB). The numerical solution successfully predicted the SOA size evolution resulting from limonene ozonolysis indoors. A sensitivity analysis showed that air exchange is a highly influential mechanism in SOA formation due to limonene ozonolysis, and that higher air exchange rates can strongly lessen the formed SOA (and thus the subsequent exposure).

5. ACKNOWLEDGMENTS

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6. NOMENCLATURE

t	Time
Δt	Time span

$d_{p,i}$	Diameter of a particle in bin i
v	Volume of a particle
v_i	Volume of a single particle in bin i
$V_{i,t}$	Total volume of particles in bin i at time t
$n(v, t)$	Number of particles with volume v at time t
$n_{i,t}$	Number of particles in bin i at time t
$n_{max}(v)$	Number of particles with volume v at the peak
$\frac{dv_j}{dt}$	Growth rate of a single particles in bin j due to partitioning
$\frac{dV_j}{dt}$	Volumetric growth rate of all particles in bin j due to partitioning
$\frac{dV}{dt}$	Total volumetric growth rate of particles due to partitioning
VR	Volume ratio; ratio of single particle volumes in two successive bins
AMF	Aerosol mass fraction
$C_{O_3,t}$	Ozone concentration at time t
$C_{terp,t}$	Terpene concentration at time t
$k_{O_3\cdot terp}$	Ozone and terpene reaction rate constant
Γ	Temperature-dependent conversion factor to change from ppb to kg/m ³ for the terpene
Kn	Knudsen number
λ	Ventilation rate
β_{SOA_k}	Surface deposition loss rate of SOA particles in bin k
β_{SOA}	Surface deposition loss rate of SOA particles
λ_{air}	Air molecules' mean free path
K	Coagulation kernel
$K_{i,j}$	Coagulation coefficient of a particle in bin i colliding with a particle in bin j
I	Condensation rate of SOA precursors on particles
k_B	Boltzmann constant
N_0	Initial total number of SOA particles
T	Temperature
D	SOA precursors' diffusivity
M	SOA precursors' molecular weight
ρ	SOA precursors' density
P	Vapor pressure of SOA precursors far from a particle

P_{eq}	Equilibrium vapor pressure of SOA precursors
$v_{i,j}$	Volume of a particle results from coagulation of two particles from bins i and j
$v dv_{j,t}$	Volume of a particle which is initially in bin j and grows due to partitioning at time t
$f_{i,j,k}$	Volume fraction of particles that resulted from collision of particles from bins i and j and enter bin k
$g_{i,j,t}$	Volume fraction of particles from bin i that grow and enter bin j at time t
N_B	Total number of bins
ρ_{SOA}	SOA density
$\beta_{SOA}(v)$	Surface deposition loss rate of a particle with volume v
$f(Kn, \alpha)$	A correction factor to take into account non-continuum regime
α	Accommodation factor
GM	Geometric mean diameter of particles with a lognormal size distribution
GSD	Geometric standard deviation of particles with a lognormal size distribution

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486 7. REFERENCES

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