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The Influence of HVAC Systems on Indoor Secondary Organic Aerosol Formation

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ABSTRACT

Chemical reactions between ozone and terpenoids can yield secondary organic aerosol (SOA), which are potentially a large source of indoor particles that are harmful to human health. The mass of SOA formed in a building is influenced by the operation of the heating, ventilation, and air-conditioning (HVAC) system. This investigation models the influence of HVAC systems on SOA concentrations in residential and commercial buildings. A parametric analysis explores the role of ventilation and recirculation rates, filtration efficiency and loading, and the operation of heat exchangers. In a rural setting, the median residential and commercial SOA concentrations for all simulations were 17.4 μg/m³ (1.09×10^{-9} lb/ft³), with a range of 2.47 to 27.0 μg/m³ ($1.54 \times 10^{-10} - 1.68 \times 10^{-9}$ lb/ft³), and 10.6 μg/m³ (6.61×10^{-10} lb/ft³), with a range of 1.81 to 26.3 μg/m³ ($1.13 \times 10^{-10} - 1.64 \times 10^{-9}$ lb/ft³), respectively. In an urban setting, the median predicted residential and commercial SOA concentrations were 68.0 μg/m³ (4.24×10^{-9} lb/ft³), with a range of 14.7 to 108 μg/m³ ($9.17 \times 10^{-10} - 6.74 \times 10^{-9}$ lb/ft³), and 44.8 μg/m³ (2.80×10^{-9} lb/ft³), with a range of 11.6 to 105 μg/m³ ($7.24 \times 10^{-10} - 6.55 \times 10^{-9}$ lb/ft³), respectively. The most influential HVAC parameters are the flow rates through the system, particle filtration efficiency, and indoor temperature for the residential and commercial models, as well as ozone removal on used filters for the commercial model. The results presented herein can be used to estimate the effects of altering HVAC system components and operation strategies on indoor SOA concentrations and subsequent exposure.

INTRODUCTION

Particulate matter (PM) diameter spans many orders of magnitude, from a few nanometers to tens of micrometers, and exposure to particles has been associated with harmful effects on human health. Fine particles (<2.5 μm in diameter) can penetrate deep into the alveolar regions of the lung (Hinds 1999). In a review of studies in the last 20 years on associations between exposure to particles and increased human mortality, Pope and Dockery (2006) conclude that fine particles have adverse effects on cardiopulmonary health. Also, exposure to ultrafine particles (<0.1 μm in diameter) have an adverse effect on cardiopulmonary function that is independent of fine particle exposure (Pekkanen et al. 2002). Most epidemiological research on the influence of particle exposure on human health has focused on the outdoor environment. However, the average American spends 18 hours indoors for every hour outdoors (Klepeis et al. 2001), and particle concentrations indoors are often much higher than outdoors, due to the strong influence of indoor sources (Wallace 2006). Common indoor sources of fine particles include cooking (Wallace et al. 2004), cigarettes (e.g. Waring and Siegel 2007), and vented clothes dryers (Wallace 2005).

Another source of indoor fine particles are chemical reactions. Weschler and Shields (1999) and other researchers (e.g., Wainman et al. 2000; Long et al. 2000; Rohr et al. 2002; Sarwar et al. 2003; Sarwar and Corsi 2007) showed that gas-phase reactions between ozone (O_3) and various terpenoids yield particles in the form of secondary organic aerosol (SOA). The reactions proceed at fast enough reaction rates to compete with loss due to air exchange (Weschler and Shields, 1996), and the reactants both commonly occur indoors. Ozone infiltrates the indoors from the outdoors (Weschler 2000), and is directly emitted indoors from office electronics (Lee et al. 2001), portable ionizers (e.g. Waring et al. 2008) or in-duct electrostatic precipitators (ESPs) (Viner et al. 1992). Also,

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terpenoids are commonly emitted indoors from consumer products, cleaners, and fragrances (Wallace et al. 1991; Singer et al. 2006b; Corsi et al. 2007). The SOA is composed of condensed products that either nucleated or partitioned onto preexisting seed particles, and SOA is in the ultrafine and lower portion of the fine particle size ranges. Further, the health effects due to SOA may be different and more deleterious than those from primary aerosols (Rohr et al. 2002 and 2003).

The heating, ventilation, and air-conditioning (HVAC) system of a building can affect the concentration of SOA formed by influencing the reactant and seed particle concentrations, as well as the indoor air temperature and relative humidity (RH). For example, ventilation rates influence indoor ozone (Weschler 2000) and seed particle concentrations (Riley et al. 2002) by affecting the rate of transport between the indoors and outdoors. Recirculation rates affect ozone, seed particle, and SOA concentrations by altering both the removal to filters in the recirculation air stream and the deposition to indoor surfaces (Sabersky et al. 1973; Lai and Nazaroff, 2000; Zuraimi et al. 2007). Seed particle and SOA concentrations are reduced by HVAC filters (Hanley et al. 1994) and in-duct ESPs (Wallace et al. 2004). Ozone is either removed by particle-laden filters (Zhao et al. 2007) or can be generated by in-duct ESPs (Viner et al. 1992). The HVAC heat exchanger (or coil) alters both the temperature and RH in the space, and the temperature affects the chemical reaction rates and resulting product vapor pressures and the RH the mass of SOA yielded (Leungsakul et al. 2005). Since the HVAC system design and operation can affect the mass of SOA formed, we explore its effects by developing a simulation that predicts the size-resolved mass of SOA formed in typical residential and commercial spaces with HVAC systems. In both the residential and commercial models, we vary the (i) ventilation and recirculation rates, (ii) the HVAC filter efficiency, (iii) the ozone removal on HVAC filters due to particle loading, and the indoor (iv) temperature and (v) relative humidity. A parametric analysis is used to explore each of these factors on SOA formation.

METHODOLOGY

The residential and commercial models are similar to models in Riley et al. (2002) and Waring and Siegel (2008), with the addition of gaseous transport and emission and SOA formation. For size-resolved parameters, including the mass of SOA formed, a particle diameter (d_p) range of 0.01 to 10 μm was considered. Residential and commercial spaces were modeled separately because of differences in values of input parameters and HVAC system operation.

The Residential and Commercial Models

Figure 1 shows the schematic that was used to account for pollutant fate and transport. The residential model does not have ventilation air intake, as most U.S. residential spaces get fresh air only from infiltration. V (m^3 or ft^3) is the volume of the space. T (°C or °F) and RH (%) are the indoor temperature and relative humidity, respectively. Q_i, Q_v, and Q_r (m^3/h or CFM) are the infiltration, ventilation, and recirculation volumetric flow rates, respectively. The ratio of a particular flow rate to the building volume is the air exchange rate, λ (h^{-1}). β_p and β_{O_3} (h^{-1}) are the loss rates of particles, including SOA, and ozone to indoor surfaces, respectively. Both loss rates are equal to the terms $v_d S$, where v_d (m/h or ft/h) is a mass transfer coefficient that is often called the deposition velocity and S (m^{-1} or ft^{-1}) is the surface area to volume ratio in the space. η_p and η_{O_3} (–) are the removal of particles, including SOA, and ozone by the HVAC filter, respectively. Particle removal by HVAC filters is intentional, but ozone removal is unintentional and occurs due to reactions with particle-laden filters. E_{O_3} and E_{terp} (μg/h or lb/h) are mass emission rates of ozone by an ESP or of terpenoids by building contents (e.g., consumer products), respectively. C_{O_3}, $C_{O_3,out}$, C_{terp} (μg/m^3 or lb/ft^3) are mass concentrations of indoor ozone, outdoor ozone, and indoor terpenoids, respectively, and $C_{O_3,m}$, $C_{O_3,m,out}$, and $C_{terp,m}$ (ppb) are molar concentrations of those compounds, respectively. n_{seed}, $n_{seed,out}$, and n_{SOA} (μg/μm•m^3 or lb/μm•ft^3) are mass distributions of indoor and outdoor seed particles, and indoor SOA, respectively.

The models assume steady-state, well-mixed conditions, constant air density, and no indoor sources of particles other than SOA formation. These assumptions are not realistic over all indoor conditions, but this is an appropriate approach to compare the relative influence of HVAC system parameters on SOA formation. The steady-state indoor mass concentration of SOA, C_{SOA} (μg/m^3 or lb/ft^3), may be calculated as the ratio of the mass formation rate of SOA and the loss rate of SOA, and C_{SOA} is calculated with Equation (1):

$$C_{SOA} = \int_{d_p} n_{SOA}\,\mathrm{d}d_p = \int_{d_p} \frac{(F_T F_{RH})\Gamma C_{terp,m}(kC_{O_3,m})Y_{g,sr}}{\beta_p + \lambda_i + \lambda_v + \lambda_r \eta_p}\,\mathrm{d}d_p \quad (1)$$

where F_T(–) and F_{RH}(–) are formation factors that adjust for changes in T and RH, respectively; k (ppb^{-1} h^{-1}) is the reaction rate constant of terpenoids and ozone; $Y_{g,sr}$ (μm^{-1}) is the size-resolved mass distribution yield of SOA formed by gas-phase reactions between ozone and the terpenoid, which is the ratio of the change in mass of SOA formed to the change in mass of terpenoid consumed; Γ(–) is a conversion factor to change units of ppb/h to μg/m^3·h or lb/ft^3·h; and λ_i, λ_v, and λ_r (h^{-1}) are air exchange rates due to infiltration, ventilation, and recirculation, respectively. Equation 1 is integrated over the modeled range of d_p = 0.01 – 10 μm. All terms in Equation (1) except k are influenced by the HVAC system and varied in the parametric analysis. In reality, k is a function of indoor temperature and is also thus affected by the HVAC system, but our model incorporates this effect in the F_T term. $Y_{g,sr}$ is a function of the seed particle concentration that infiltrates

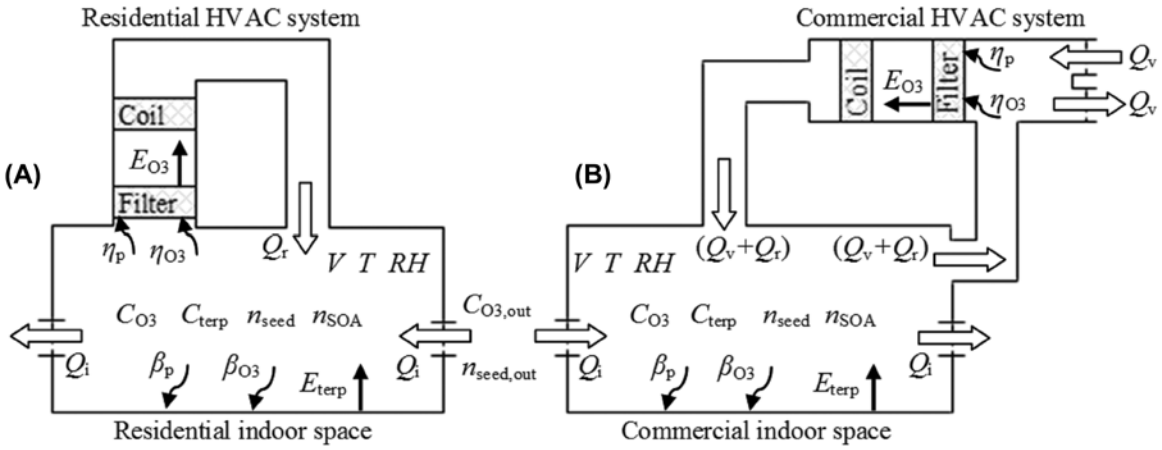

Figure 1 *Schematic for the (A) residential and (B) commercial models. Block arrows represent airflows and line arrows represent species losses or gains. (Symbols are defined in text.)*

indoors from outdoors. The indoor steady-state seed particle concentration of outdoor origin, C_{seed} (μg/m³ or lb/ft³), is calculated with Equation (2), which also contains an integral since n_{seed}, $n_{seed,out}$, p_p, η_p, and β_p are particle size-resolved:

$$C_{seed} = \int_{d_p} n_{seed} \mathrm{d}d_p = \int_{d_p} n_{seed,out} \frac{p_p \lambda_i + \lambda_v (1 - \eta_p)}{\beta_p + \lambda_i + \lambda_v + \lambda_r \eta_p} \mathrm{d}d_p \quad (2)$$

where p_p (–) is the size-resolved penetration of particles through the building envelope. The steady-state indoor terpenoid mass concentration, C_{terp}, is calculated with Equation (3) and was considered to be independent of the indoor ozone concentration. In reality, some amount of terpenoid would react with the ozone and thus decrease, but we assume that this decrease is small compared to the amount of reacted ozone and is thus neglected.

$$C_{terp} = \frac{E_{terp}/V}{\lambda_i + \lambda_v} \quad (3)$$

The steady-state indoor ozone mass concentration, C_{O_3}, is calculated with Equation (4):

$$C_{O_3} = C_{O_3,out} \frac{\lambda_i + \lambda_v (1 - \eta_{O_3}) + \dfrac{E_{O_3}}{C_{O_3,out} V}}{\beta_{O_3} + \lambda_i + \lambda_v + \lambda_r \eta_{O_3} + k C_{terp}} \quad (4)$$

All integration was performed numerically. Particle size-resolved parameters were divided into 80 different bins, with 20 to 30 bins for each order of magnitude range. The HVAC parameters in Equations (1) to (4) were varied to parametrically explore their influence on C_{SOA}.

Building Model Parameters

The volume, V, for the residential model was 392 m³ (13,843 ft³), which is based on the typical floor area from the U.S. Bureau of the Census (2005) of 163.3 m² (1758 ft²) multiplied by an assumed ceiling height of 2.4 m (8 ft). The volume, V, for the commercial model was assumed as 1000 m³ (35,315 ft³), which was arbitrarily chosen because commercial buildings span a wide range of volumes, depending on their use. The commercial floor area is 416.7 m² (4485 ft²), which assumes the same ceiling height of 2.4 m (8 ft). Using the methods and parameter values of Riley et al. (2002), the size-resolved penetration of particles through the building envelope (p_p) was calculated according to the theory of Liu and Nazaroff (2001). These calculated penetration factors closely resemble the measured penetration factors reported in Long et al. (2001) for all but the very largest of particle diameters. Because of the lack of commercial penetration factors in the literature, identical penetration factors were used for both models.

Ambient Parameters

For both models, a Rural and an Urban ambient case were modeled to compare how the HVAC system affects SOA formation in different climates. Two parameters were varied for each ambient case, the outdoor ozone concentration and the outdoor particle distribution. An outdoor ozone molar concentration, $C_{O_3,m,out}$, was assumed as 25 ppb for the Rural case and 100 ppb for the Urban case. The Urban case had higher ozone because automobile pollution often leads to ozone in the form of photochemical smog (Seinfeld and Pandis 1998). The outdoor and indoor seed particle mass distribution functions, $n_{seed,out}$ and n_{seed}, respectively, both depend on the size-resolved outdoor particle number distribution, $n_{seed,N,out}$ (#/μm•m³ or #/μm•ft³). For the outdoor distribution in both the residential and commercial models, we

Table 1. For the Rural and Urban Ambient Cases

The outdoor ozone concentrations and particle number distributions, including the total number concentrations (#/cm³ or multiply by 2.83×10^4 for #/ft³) and the geometric mean diameters (GM) and log of geometric standard deviations [log(GSD)] for each mode.

		Particle Distributions								
		Mode 1			Mode 2			Mode 3		
Ambient Case	Ozone Conc. (ppb)	Number (#/cm³)	GM (μm)	log(GSD) (–)	Number (#/cm³)	GM (μm)	log(GSD) (–)	Number (#/cm³)	GM (μm)	log(GSD) (–)
Rural*	25	6650	0.015	0.225	147	0.054	0.557	1990	0.084	0.266
Urban*	100	99,300	0.013	0.245	1100	0.014	0.666	36,400	0.05	0.337

*Ozone concentrations were assumed, and particle number distributions are from Jaenicke (1993).

used Rural and Urban number distributions from Jaenicke (1993), which are synthesized distribution from various sources. These are described as tri-modal lognormal distributions, and the number, geometric mean diameter (GM), and log of the geometric standard deviation (GSD) that describe the three modes are listed in Table 1. These same distributions were used in Riley et al. (2002) and Waring and Siegel (2008). Each $n_{seed,N,out}$ was converted to $n_{seed,out}$ by assuming the particles are spherical and multiplying the volume of the geometric mean of each size bin by an assumed particle density of 1 g/cm³ (62.4 lb/ft³). The resulting Rural $PM_{2.5}$ and PM_{10} concentrations are 7.3 and 15 μg/m³ (4.6×10^{-10} and 9.4×10^{-10} lb/ft³), respectively, and the Urban $PM_{2.5}$ and PM_{10} concentrations are 43 and 60 μg/m³ (2.7×10^{-9} and 3.7×10^{-9} lb/ft³), respectively.

Terpenoid Reactant Parameters

The parameters E_{terp}, $Y_{g,sr}$, and k were derived from a series of papers that reported SOA formation from ozone reactions with a set of common consumer products (Destaillats et al. 2006; Singer et al. 2006a and 2006b; Coleman et al. 2008). The results for a pine-oil cleaner (POC) are used to determine E_{terp}, $Y_{g,sr}$, and k in both the residential and commercial models, since its use in both settings is realistic. The pine-oil cleaner labeled GPC-1 in Singer et al. (2006b) and POC in Coleman et al. (2008) are the same consumer product, and E_{terp} is derived from Singer et al. (2006b) and $Y_{g,sr}$ and k from Coleman et al. (2008).

The modeled emission, E_{terp}, is a floor-mopping event based off a technique described in Singer et al. (2006b). Floor-mopping with the POC emits 7.26 mg of terpenoids/g POC (7.26×10^{-3} lb of terpenoids/lb POC) and the mopping technique uses 3.3 g POC/m²•h (6.8×10^{-4} lb POC/ft²•h). In the residential model, the emission is a floor-mopping of one-fourth the total floor area, so E_{terp} = 978 mg/h (2.15×10^{-3} lb/h) of terpenoids. In the commercial model, the emission is a floor-mopping of one-half the total floor area, so E_{terp} = 3743 mg/h (8.23×10^{-3} lb/h) of terpenoids. Of these emissions, 78.5% are due to two terpenes, d-limonene and terpinolene, and one terpene-alcohol, α-terpineol. Of these, d-limonene has the largest SOA formation potential (Weschler and Shields 1999; Ng et al. 2006). Three experiments in Coleman et al. (2008) were conducted under nearly identical conditions, except two had deliberately low seed particle concentrations and one used laboratory air with seed particles, with $PM_{1.1}$ = 4 μg/m³ (2.5×10^{-10} lb/ft³). $Y_{g,sr}$ was modeled as depending on the seed particle concentrations and was calculated with results for the average of the low seed particle experiments (0.111) and for the seed particle experiment (0.197). The following linear relationship was determined: $Y_{g,sr} = 0.0213\ (C_{seed,\ PM1.1}) + 0.1107$, where $C_{seed,\ PM1.1}$ (μg/m³ or lb/ft³) is the concentration of indoor seed particles less than 1.1 μm in our models. This relationship is only used in our residential and commercial models if $C_{seed,\ PM1.1} \leq 4$ μg/m³ (2.5×10^{-10} lb/ft³). If $C_{seed,\ PM1.1} >$ 4 μg/m³ (2.5×10^{-10} lb/ft³) then $Y_{g,sr}$ is constant at 0.197. The size-resolved mass yield, $Y_{g,sr}$, is modeled as a lognormal distribution, and its parameters were fitted to the POC-Seed experiment by converting the tri-modal lognormal distribution of the steady-state SOA number concentration into a uni-modal lognormal mass distribution, with GM = 0.37 μm and GSD = 1.52. The ozone and terpenoid reaction rate constant, k, was calculated as follows. Neglecting ozone decay due to irreversible wall deposition (experiments were in Teflon-lined chamber), a steady-state mass balance with ozone and terpenoid concentrations yields $k = 0.05$ ppb^{-1} h^{-1}. $Y_{g,sr}$ and k are identical in both models.

HVAC Parameter (i): HVAC Flow

The air exchange rates used in the models are listed in Table 2. The HVAC system directly controls the ventilation and recirculation rates, and these were varied to explore their influence on SOA formation. The Flow cases used infiltration and recirculation air exchange rates from Riley et al. (2002) and Waring and Siegel (2008). The residential Duty case assumed cycling of conditioning equipment, and therefore recirculation for one-sixth of the total time, and the Continuous case considered the air handler fan to be running the entire time. For the commercial HVAC Flow cases, all operation was continuous and three air makeup cases were considered, with assumed air exchange rates based on engineering judgment that were also used in Waring and Siegel (2008). The 100% outside air (OA) case represents a building for which air recirculation is undesirable. The 50% OA/50% recirculated air

Table 2. Summary of Air Exchange Rates Used in the Residential and Commercial Models

Air Exchange Rate (h^{-1})	Residential		Commercial		
	Duty	Continuous	100% OA	50% OA/ 50% RA	10% OA/ 90% RA
λ_i	0.75	0.75	0.25	0.25	0.25
λ_v	0	0	4	2	0.4
λ_r	0.67	4	0	2	3.6

(RA) case represents a heavily occupied building, and the 10% OA/90% RA case represents a lightly occupied building.

In the residential model, the Duty and Continuous flow cases also cause different airflow regimes in the modeled indoor space. Indoor spaces without continuous recirculation are assumed to have average air flows of lower velocity than those indoor spaces with continuous recirculation. Since higher velocity flows lead to a decreased thickness of the boundary layers adjacent to surfaces, the deposition parameters of β_p and β_{O_3} are expected to increase with higher velocity flows. Thus, the residential model uses different values of β_p and β_{O_3} for the Duty and Continuous cases. The commercial model has continuous flow for all cases, so it uses one constant value for both β_p and β_{O_3}.

Similar to in Riley et al. (2002) and Waring and Siegel (2008), we used the model of Lai and Nazaroff (2000) to determine specific values of β_p. One input in their model for β_p is the friction velocity, u^* (cm/s or ft/h), which is an empirical parameter that describes the level of turbulence intensity near a surface. This parameter thus represents the air flow conditions in a space, with higher values for u^* associated with higher velocity flows. Typical values of u^* for indoor environments are 0.3 to 3 cm/s (35.4 to 354 ft/h) (Lai and Nazaroff, 2000). For the residential model, the Duty case was assigned the β_p for u^* = 1 cm/s (118 ft/h) and the Continuous case for u^* = 3 cm/s (354 ft/h). For the commercial model, all three flow cases were assigned β_p for u^* = 3 cm/s (354 ft/h).

Sabersky et al. (1973) described β_{O_3} for two residential cases in the same home, without and with the forced air system operating, at 2.9 and 5.4 h^{-1}, respectively. In the residential model, our Continuous case assumes the forced air system is always on, so it was assigned as $\beta_{O_3} = 5.4\ h^{-1}$. For the Duty case, we assumed a value of 5.4 h^{-1} when the system was on and a value of 2.9 h^{-1} when the system was off, for an overall value of $\beta_{O_3} = 3.3\ h^{-1}$. For the commercial model, β_{O_3} was assigned for all flow cases as 4.2 h^{-1}, which is an average of the office ozone deposition loss rates summarized in Weschler (2000).

HVAC Parameter (ii): PM Filtration

Five removal devices were used in the residential and commercial models: four porous-media filters and one electrostatic precipitator (ESP). The efficiency curves for all five filters are displayed in Figure 2. We assumed that each filter

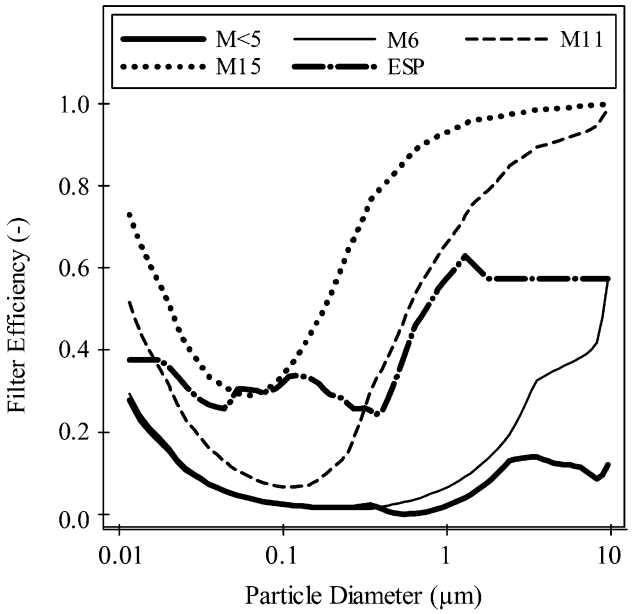

Figure 2 *Filter efficiency curves for the MERV <5, 6, 11, and 15 filters (M<5, M6, M11, and M15) and the electrostatic precipitator (ESP) used in this modeling study.*

retains the efficiency shown in Figure 2 and is constant over time, though filter removal efficiency typically changes with loading (Hanley et al. 1994; Wallace et al. 2004). Filter efficiency data for the four porous-media HVAC filters were obtained from ASHRAE Standard 52.2 tests (ASHRAE 2007) provided by filter manufacturers. The ASHRAE Standard 52.2 procedure challenges filters with particles from 0.3 to 10 μm, so the fibrous filtration theory described by Hinds (1999) was used to extend the data into the full range used in this study, following the procedure in Riley et al. (2002). These are the same filter curves used in Waring and Siegel (2008).

The fifth filter used in our models was an in-duct ESP, and its efficiency curve was derived from Wallace et al. (2004), who reported size-resolved mean deposition rates in a townhome with the central house fan operating continuously, both without and with an in-duct ESP operating. The size-resolved efficiency of the in-duct ESP was calculated for each reported particle diameter in Wallace et al. (2004) with the relationship, $\Delta\beta_{ESP} = (\lambda_{r,townhome})(\eta_{ESP})$, where $\Delta\beta_{ESP}$ is the difference in deposition loss rates with and without the ESP operating, $\lambda_{r,townhome}$ is the rate of recirculated air in the townhome (reported by the authors as 5.4 h^{-1}), and η_{ESP} is the calculated size-resolved efficiency of the in-duct ESP. Wallace et al. (2004) reported size-resolved deposition rates for the particle diameter range of 0.0181 to 1.843 μm, so the efficiencies of modeled particle diameters that were lower than this range were assigned the η_{ESP} for the particle diameter of 0.0181 μm and those higher were assigned the η_{ESP} for the particle diam-

eter of 1.843 μm. The same efficiency curve for η_{ESP} was used in the residential and commercial models due to the lack of efficiencies reported for ESPs in commercial systems. The use of an in-duct ESP produces ozone, so our models coupled an indoor ozone emission rate, E_{O_3}, to the residential and commercial scenarios that employed the ESP. Wallace et al. (2004) did not report ozone emission rates for the studied ESP, so our residential and commercial models used the rate of a commercially available unit in Viner et al. (1992) of E_{O_3} = 21.6 mg/h (4.76×10^{-5} lb/h) for the continuous HVAC Flow cases in the residential and commercial models. For the Duty case in the residential model, the ozone emission rate is one-sixth the Continuous case and is E_{O_3} = 3.6 mg/h (7.93×10^{-6} lb/h).

HVAC Parameter (iii): O_3 Filtration

As ozone-laden air passes through a porous-media HVAC filter, ozone can be removed by the filter, predominately due to reactions with loaded particles (Hyttinen et al. 2003; Bekö et al. 2006, 2007; Zhao et al. 2007). We modeled New and Used filter cases for ozone removal. Both models assumed that η_{O_3} = 0% for their New cases. The models assumed Used values of η_{O_3} = 10% and 41% for residential and commercial buildings, respectively (Zhao et al. 2007). The residential value of 10% is the mean of eight particle-laden filters taken from actual residences, and the commercial value of 41% is the mean of five particle-laden filters from commercial environments. Though the ozone removal efficiency of a porous-media filter is likely associated with its particle removal efficiency, our model does not link the two since there are insufficient data to make such an association. All scenarios with an ESP are assigned the ozone removal value of η_{O_3} = 0%.

HVAC Parameter (iv): Temperature

The heating or cooling coil operation influences the air temperature in a modeled space. Three different indoor temperatures, 18.3, 23.9, and 29.4°C (65, 75, and 85°F) were considered. The total mass of SOA formed increases as temperature decreases (Leungsakul et al. 2005; Sarwar and Corsi 2007). Though the reaction rate of ozone and terpenoids decreases, this decrease is surpassed by the increase in gas-to-particle partitioning that occurs as the vapor pressures of condensing products decrease. For reactions between ozone and d-limonene (the primary SOA forming reactant in the POC), Leungsakul et al. (2005) report that the mass of SOA formed changes at a rate of –0.016°C^{-1} (–0.0089°F^{-1}) and Sarwar and Corsi (2007) report a rate of –0.04°C^{-1} (–0.022°F^{-1}). The experiments reported in Coleman et al. (2008) were conducted at a temperature of 23°C (73.4°F), and temperature formation factors, F_T, were calculated with the averages of the two rates from Leungsakul et al. (2005) and Sarwar and Corsi (2007). These F_T adjust the mass of SOA formed at the experimental temperature to represent that which would occur at the modeled temperatures. For both models, F_T equals 1.13, 0.98, and 0.82 for the cases of 18.3, 23.9, and 29.4°C (65, 75, and 85°F), respectively.

HVAC Parameter (v): Relative Humidity

The heating or cooling coil also influences the relative humidity (RH) in a space. Both models utilize three different values for indoor RH of 25, 50, and 75% to model the range of RH that occurs in buildings in different climates. As RH decreases, the water available for reactions becomes limited. Some products of the ozone and d-limonene reaction (stabilized Criegee intermediates) that can react with water instead react with other products of the ozone and d-limonene reactions (aldehydes) to form less volatile compounds, increasing total SOA mass formed (Leungsakul et al. 2005). Leungsakul et al. (2005) report that the mass of SOA formed changes at a rate of –0.0009%$^{-1}$. The experiments reported in Coleman et al. (2008) were conducted at an RH of 50%, and RH formation factors, F_{RH}, were calculated with the rate from Leungsakul et al. (2005). These F_{RH} adjust the mass of SOA formed at the experimental RH to represent that which would occur at the modeled RH. For the residential and commercial models, F_{RH} equals 1.02, 1.0, and 0.98 for the cases of 25, 50, and 75%, respectively.

Base Case Definitions and Number of Reported Scenarios

For the residential and commercial models, the varied HVAC parameters, as well as the literature sources used for input values, are summarized in Table 3. A Rural and Urban base case for both the residential and commercial models was selected based on typical values for each parameter. Within the Rural and Urban distributions, the residential base case consisted of a Duty flow cycle, a MERV 6 filter [the requirement for new homes in ASHRAE Standard 62.2 (ASHRAE 2004)], a Used filter with an ozone removal efficiency of 10%, and indoor temperature of 23.9°C (75°F) and an RH of 50%. Within the Rural and Urban distributions, the commercial base case consisted of a 10% OA/90% RA flow cycle, a MERV 6 filter, a Used filter with an ozone removal efficiency of 41%, and indoor temperature of 23.9°C (75°F) and an RH of 50%. Each combination of the parameters was modeled. The residential model had 324 unique scenarios (162 each of Rural and Urban) and the commercial model had 486 unique scenarios (243 each of Rural and Urban).

RESULTS AND DISCUSSION

Using Equations (1) through (4), the resulting SOA concentrations, C_{SOA}, varied over an order of magnitude, depending on the HVAC input parameters. SOA yield and other size-resolved parameters are lognormally distributed, so the median was used as a descriptive statistic. For the residential model, the median C_{SOA} over all 162 Rural scenarios was 17.4 μg/m^3 (1.09×10^{-9} lb/ft^3), with a range of 2.47 to 27.0 μg/m^3 ($1.54 \times 10^{-10} - 1.68 \times 10^{-9}$ lb/ft^3) and over all 162 Urban scenarios was 68.0 μg/m^3 (4.24×10^{-9} lb/ft^3), with a range of 14.7 to 108 μg/m^3 ($9.17 \times 10^{-10} - 6.74 \times 10^{-9}$ lb/ft^3). The resi-

Table 3. Summary of Varied HVAC Parameters for the Residential and Commercial Models

The base cases are listed in bold.
Literature sources are listed below the table.

Parameter	Model	Cases (Base Case in bold)
HVAC Flow[a]	Residential	**Duty**, Continuous
	Commercial	100% OA, 50% OA/50% RA, **10% OA/90% RA**
PM Filtration[b]	Residential, Commercial	MERV <5, **6,** 11, 15; ESP
O_3 Filtration[c]	Residential, Commercial	New, **Used**
Temperature[d]	Residential, Commercial	18.3, **23.9**, 29.4°C (65, **75**, 85°F)
Relative Humidity[e]	Residential, Commercial	25, **50**, 75%

[a]Riley et al. (2002), Waring and Siegel (2008)
[b]Waring and Siegel (2008), Hinds (1999), and Wallace et al. (2004)
[c]Zhao et al. (2007)
[d]Leungsakul et al. (2005) and Sarwar and Corsi (2007)
[e]Leungsakul et al. (2005)

dential base case C_{SOA} for the Rural case was 22.8 μg/m³ (1.42 × 10⁻⁹ lb/ft³) and for the Urban case was 90.9 μg/m³ (5.67 × 10⁻⁹ lb/ft³). For the commercial model, the median C_{SOA} over all 243 Rural scenarios was 10.6 μg/m³ (6.61 × 10⁻¹⁰ lb/ft³), with a range of 1.81 to 26.3 μg/m³ (1.13 × 10⁻¹⁰ – 1.64 × 10⁻⁹ lb/ft³) and over all 243 Urban scenarios was 44.8 μg/m³ (2.80 × 10⁻⁹ lb/ft³), with a range of 11.6 to 105 μg/m³ (7.24 × 10⁻¹⁰ – 6.55 × 10⁻⁹ lb/ft³). The commercial base case C_{SOA} for the Rural case was 15.4 μg/m³ (9.61 × 10⁻¹⁰ lb/ft³) and for the Urban case was 62.9 μg/m³ (3.92 × 10⁻⁹ lb/ft³). These ranges are of the same order as reported in real buildings under a variety of experimental conditions (Weschler and Shields 1999; Hubbard et al. 2005). The size-resolved distributions of SOA for these base cases, as well as the outdoor and indoor seed, SOA, and total indoor particle distributions are in Figure 3.

The median C_{SOA}, the ranges of formation, and the base case results for C_{SOA} illustrate that much more SOA is formed in the Urban than the Rural area, in both models. The indoor terpenoid concentrations, C_{terp}, were the same in both areas, and the higher formation is due to the greater urban outdoor ozone concentration, $C_{O_3,m,out}$, of 100 ppb over the rural concentration of 25 ppb. The integrated concentrations for

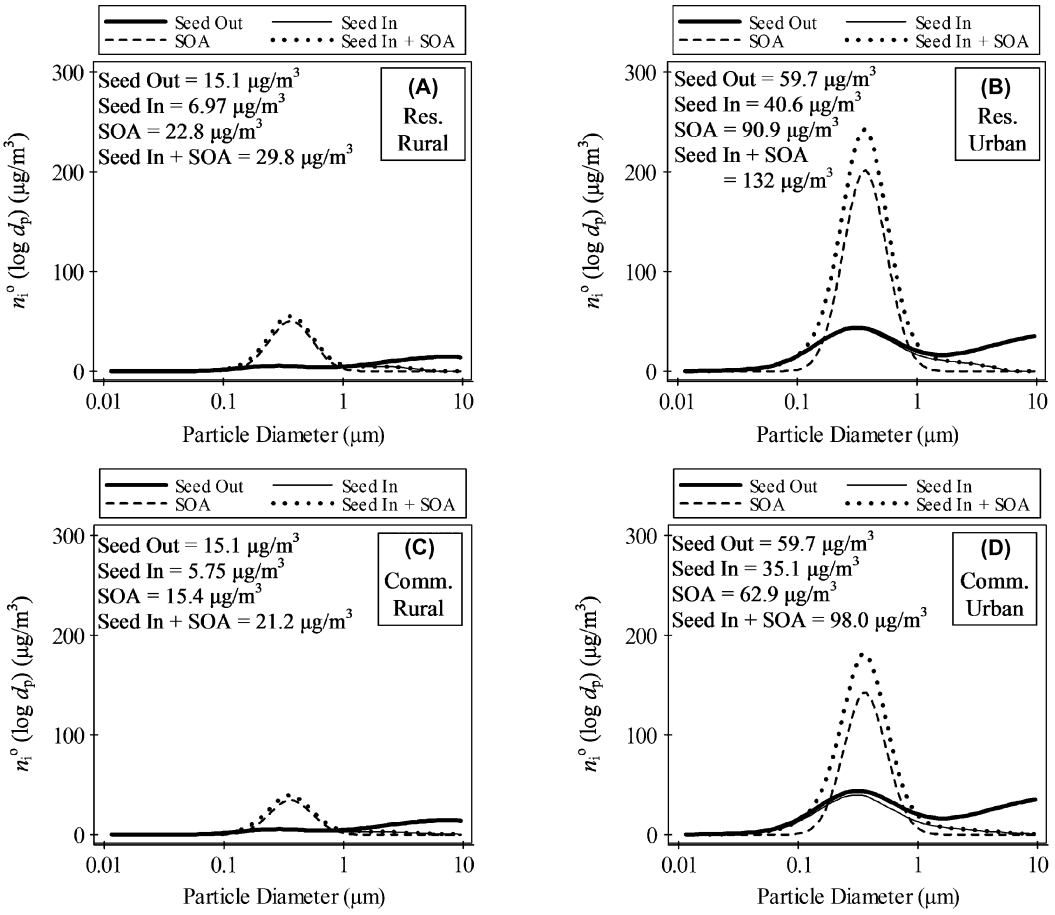

Figure 3 *Outdoor seed, indoor seed, SOA, and Seed + SOA particle distributions (μg/m³ or multiply by 62.4×10⁻¹¹) to convert to lb/ft³) for residential (A) rural and (B) urban base cases and commercial (C) rural and (D) urban base cases.*

Table 4. SOA Change Ratios (SCR = Adjusted C_{SOA}/Base Case C_{SOA}) for Rural and Urban Base Cases in the Residential and Commercial Models

Parameter	Model	Base Case	Going to	SOA Change Ratio (SCR)	
				Residential	Commercial
Rural Base Case					
Flow	Residential	Duty	Continuous	0.84	
	Commercial	10% OA/90% RA	50% OA/50% RA		0.73
			100% OA		0.52
PM Filtration	Residential, Commercial	MERV 6	MERV <5	1.01	1.06
			MERV 11	0.76	0.34
			MERV 15	0.53	0.14
			ESP	0.94	0.70
O_3 Filtration	Residential, Commercial	Used	New	1.00	1.37
Temperature	Residential, Commercial	23.9°C (75°F)	18.3°C (65°F)	1.15	1.15
			29.4°C (85°F)	0.84	0.84
RH	Residential, Commercial	50%	25%	1.02	1.02
			75%	0.98	0.98
Urban Base Case					
Flow	Residential	Duty	Continuous	0.86	
	Commercial	10% OA/90% RA	50% OA/50% RA		0.71
			100% OA		0.51
PM Filtration	Residential, Commercial	MERV 6	MERV <5	1.01	1.04
			MERV 11	0.80	0.44
			MERV 15	0.62	0.22
			ESP	0.85	0.68
O_3 Filtration	Residential, Commercial	Used	New	1.00	1.37
Temperature	Residential, Commercial	23.9°C (75°F)	18.3°C (65°F)	1.15	1.15
			29.4°C (85°F)	0.84	0.84
RH	Residential, Commercial	50%	25%	1.02	1.02
			75%	0.98	0.98

Rural base case: residential model = 22.8 μg/m³ (1.42×10^{-9} lb/ft³); commercial model = 15.4 μg/m³ (9.61×10^{-10} lb/ft³).
Urban base case: residential model = 90.9 μg/m³ (5.67×10^{-9} lb/ft³); commercial model = 62.9 μg/m³ (3.92×10^{-9} lb/ft³).

each of the four plotted particle mass distributions are also displayed in Figure 3. For the base cases in both models, the C_{SOA} in the Rural environment was a higher fraction of the total indoor particles than in the Urban environment, at 76.5% versus 68.9% for the residential base cases and 72.6% versus 64.2% for the commercial base cases. Figure 3 shows for all bases cases, most of the resulting SOA distribution is in the 0.1 to 1 μm diameter size range, consistent with the findings of other researchers (Weschler and Shields 1999; Coleman et al. 2008).

The parametric influence of each of the HVAC parameters on the base cases was determined with the SOA Change Ratio (SCR), which is listed in Table 4. The SCR equals the *adjusted* C_{SOA} divided by the *base case* C_{SOA}, and the adjusted C_{SOA} is the result of holding all parameters in the base case constant except for the varied parameter. Thus, the SCR is a measure of how sensitive C_{SOA} is to a change of a given HVAC parameter, relative to the base case. Non-influential parameters have SCRs at or near unity. Parameter changes that lead to lower C_{SOA} have an SCR less than unity, and parameter changes that lead to higher C_{SOA} have an SCR greater than unity. The product of the SCR and the base case C_{SOA} yields the adjusted C_{SOA}.

Table 4 shows that there was little change in the SCR for the same case in the Rural versus the Urban environments. The different $C_{O_3,m,out}$ of 25 and 100 ppb affect the absolute C_{SOA} formed for the different ambient cases. However, these different $C_{O_3,m,out}$ do not affect the SCR since it is a measure of change relative to the base case. The differences that are observed in SCRs for the Rural versus the Urban cases are due to the different seed particle concentrations, C_{seed}, for the two ambient conditions, since lower seed particle concentrations lead to a lower $Y_{g,sr}$. However, this effect is small because of limitations in available input parameters. The experimental data in Coleman et al. (2008) that were used to generate the linear relationship between $Y_{g,sr}$ and $C_{seed,PM1.1}$ were only for the particle mass range of $C_{seed,PM1.1} \leq 4$ μg/m³ (2.5×10^{-10} lb/ft³). For most of the modeled scenarios, $C_{seed,PM1.1}$ was greater than 4 μg/m³ (2.5×10^{-10} lb/ft³), so $Y_{g,sr}$ was at a constant value that did not change with the indoor seed particle concentration. In reality, the SOA yield could increase further with higher seed particle concentrations, but there is limited

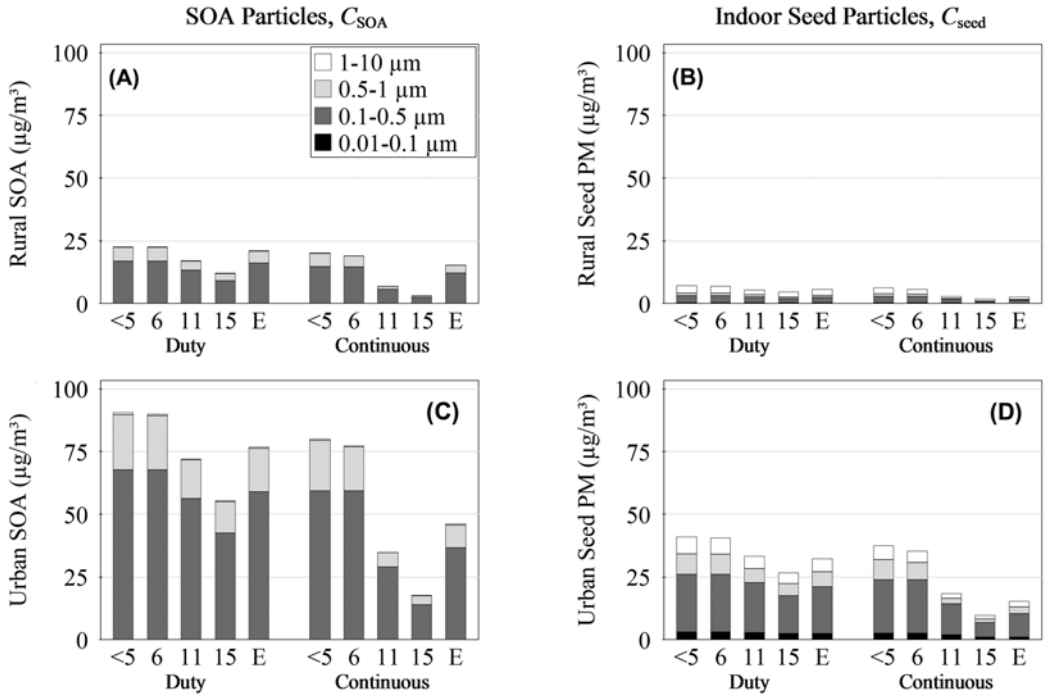

Figure 4 *Size-bin-resolved residential rural (A) C_{SOA} and (B) C_{seed} and residential urban (C) C_{SOA} and (D) C_{seed} (µg/m³ or multiply by 6.24×10^{-11} to convert to lb/ft³) for PM filtration parameters within each flow case.*

data in the literature on the subject to more fully model this effect.

The SCRs in Table 4 show that the most influential HVAC parameters are those in the cases of Flow, PM Filtration, and Temperature for the residential and commercial models, as well as O_3 Filtration for the commercial model. In the residential model, changing flow cases from Duty to Continuous decreases the SCR since continuous air flow allows the HVAC filter to remove more of the SOA formed. The different Flow cases have a larger relative effect in the commercial building, since volumetric air flow is larger and always continuous with an air exchange rate through the HVAC system of $(\lambda_r + \lambda_v) = 4\ h^{-1}$. For the changes to the two different flow strategies, the 100% OA case causes the largest relative decrease in the SCR since no air is recirculated and both the terpenoids emitted indoors and the SOA formed indoors are completely ventilated (though the C_{O_3} of outdoor origin is higher in this case). The 50% OA/50% RA case also yields a smaller C_{SOA} than the base case, but it has a larger C_{SOA} than the 100% OA case since some of the air is recirculated. Filtration affects the results with the SCR generally increasing with lower efficiency filters and decreasing with higher efficiency filters. The SCR for going to a MERV <5 filter is near unity since the MERV <5 and 6 filters have similar removal for the size range of SOA formation of 0.1 to 1 µm (see Figure 2). An increase in filter efficiency has a larger relative effect on the SCR within the commercial than the residential model due to its continuous flow and larger volumetric flow rates through the HVAC filter. In the residential model, the O_3 Filtration had little effect on the SCR since there was ozone removal by the Used filter, η_{O_3}, of 10% and duty air flow. However, the commercial model exhibited a large SCR since it had a Used η_{O_3} of 41% within a continuous flow case with as much as 90% recirculated air. The Temperature and RH cases affect C_{SOA} with F_T and F_{RH} in Equation (1), and their SCRs vary by approximately 15% over the temperature range considered and about 2% over the relative humidity range.

Table 4 displays results derived from the integrated C_{SOA} results. More nuanced trends in the SOA formation can be determined by examining summary results from a size-resolved perspective, also within the context of parametric influence on C_{SOA}. Figure 4 displays the median particle size-resolved residential Rural (A) C_{SOA} and (B) C_{seed} and residential Urban (C) C_{SOA} and (D) C_{seed}, for PM Filtration parameters within each Flow case. Each bar summarizes the results that use the two parameters listed below it as inputs, with the fraction of C_{SOA} and C_{seed} attributed to each size bin demarcated by a different shade within the bar. In Figure 4A, for example, the size-resolved bar for the MERV <5 filter within the Duty case was created by summing the median size-resolved geometric means of C_{SOA} and C_{seed} for any modeled result that had MERV <5 and Duty parameters as inputs. The total number of Rural and Urban scenarios were each 162, and each bar is the result of 18 scenarios except the ESP, which is the result of 9 scenarios (since the ESP does not have a Used case for O_3 Filtration).

As with the base cases, the Rural cases yielded lower C_{SOA} than the Urban cases, due to the lower rural $C_{O_3,out}$. Indoor seed particles, C_{seed}, were higher for the Urban than the Rural cases, since the outdoor mass concentrations were higher, particularly for particles within the range of 0.1 to 1 μm. This size range readily penetrates through the building envelope with the infiltration air exchange, which is the only source of outdoor particles in the residential model. Most of the resulting C_{SOA} mass were particles in the size range of 0.1 to 0.5 μm, with the rest in the range of 0.5 to 1 μm. This particle size range also tends to have the longest residence time in the air, since it has the lowest values for removal by both filtration and deposition to indoor surfaces. The fact that the C_{SOA} are larger than C_{seed} and most of the C_{SOA} is in the range of 0.1 to 1 μm implies the following about particle source apportionment in indoor environments with substantial SOA formation. Particles in the range of 0.1 to 1 μm are likely to be SOA products of chemical reactions, and particles in the size ranges smaller than or larger than 0.1 to 1 μm are likely to be of outdoor origin, assuming no other indoor sources of particles.

This result has implications for applying density assumptions to concentrations of indoor particles with a substantial fraction of SOA, implying that one density assumption for all sizes may not be ideal. Our models and Coleman et al. (2008) assumed a density of 1 g/cm^3 (62.4 lb/ft^3) for all SOA and particles of outdoor origin, but other researchers have assumed different values. SOA density assumptions in the literature commonly fall into the range of 1 to 1.4 g/cm^3 (62.4 to 87.4 lb/ft^3). Our SOA density assumption did not affect the GM or the GSD of the lognormal yield of SOA that was calculated from Coleman et al. (2008), but only the magnitude of formation. Outdoor particles are often assigned a density of 1 g/cm^3 (62.4 lb/ft^3), but sometimes they are assigned a density of 2.5 g/cm^3 (156 lb/ft^3) for particles over 2.5 μm, since this size range may be more likely to contain crustal material (Seinfeld and Pandis 1998). However, our density assumptions do not affect the utility of the results of our model. To adjust any reported C_{SOA} or C_{seed} to the value it would have with a different density assumption, multiply that C_{SOA} or C_{seed} by the factor change of density (e.g., multiply C_{SOA} by 1.2 to change the C_{SOA} results to those for a density of 1.2 g/cm^3).

Figure 4 also shows that higher efficiency filters lead both to reduced C_{SOA} and C_{seed}. Furthermore, the higher efficiency filters have a larger effect for the residential scenarios with continuous HVAC operation. Continuously recirculated air moves through the HVAC system with an air exchange rate of 4 h^{-1} versus 0.67 h^{-1} for the cyclical duty flow, which allows the filter to remove six times more mass in the Continuous versus the Duty cases. The ESP removes particles much better than the MERV <5 or 6 filters and better than or similarly to the MERV 11 filter for the size range of 0.1 to 1 μm (see Figure 2). However, the ESP leads to approximately equal C_{SOA} when compared to the MERV <5 or 6 filters for Duty or Continuous flow within the Rural cases and for Duty flow within the Urban cases. These comparable resulting C_{SOA} values are because the indoor ozone concentration, C_{O_3}, increases due to the ozone emission of the ESP, E_{O_3}. The relative effect of E_{O_3} on C_{SOA} is larger in the rural environment since $C_{O_3,out}$ is lower. Though not included in this model, filter bypass would have a relatively large influence on reducing the effect of changing filtration efficiency, since the range of SOA formation is the size range of particles very likely to follow fluid streamlines and flow around the filter with the bypass air (Ward and Siegel 2005; Waring and Siegel 2008). Our models also assumed a constant filter removal efficiency, though filter loading over time will likely change the removal efficiency of the porous-media filters (Hanley et al. 1994) and decrease the removal efficiency of the ESP (Wallace et al. 2004).

Figure 5 is similar to Figure 4, but for the commercial Rural (A) C_{SOA} and (B) C_{seed} and commercial Urban (C) C_{SOA} and (D) C_{seed}, for PM Filtration parameters within each Flow case. Figure 5 does not display the MERV < 5 and 11 filters for brevity and since their trends may be inferred from the results of the MERV 6 and 15 filters. The total number of Rural and Urban scenarios were each 243, and each bar is the result of 36 scenarios except the ESP, which is the result of 18 scenarios (since the ESP does not have a Used case for O_3 Filtration). As with the base cases, there are smaller C_{SOA} in the commercial versus the residential model. Similar to the residential model, in the commercial model the effect of PM Filtration depends on the Flow case. However, in contrast to the residential model, all flow is continuous in the commercial model, and higher filtration efficiency leads to lower C_{SOA} in cases with higher fractions of recirculated air. For the 100% OA case, more efficient filtration had no effect on SOA concentrations since all indoor air was continuously ventilated. The ESP led to a higher C_{SOA} for the 100% OA cases due to its emission of ozone, E_{O_3}. The C_{SOA} for scenarios with the ESP changed little for different Flow cases, since the filtration of SOA by the ESP is challenged by the extra SOA that is formed due to the increased influence of the E_{O_3} within recirculated air.

The parametric analysis with the SCRs demonstrates that the O_3 Filtration parameter influences C_{SOA} in the commercial model. Figure 6 is similar to Figures 4 and 5, and it shows the size-bin-resolved commercial (A) Rural and (B) Urban C_{SOA}, for O_3 Filtration parameters within each Flow case. Figure 6 does not display C_{seed} since it is not dependent on η_{O_3}. The total number of Rural and Urban scenarios were each 243, and each of the New bars are the result of 45 scenarios and the Used bars the result of 36 scenarios (since the ESP does not have a Used case for O_3 Filtration). The Rural and Urban cases exhibit similar trends, but with different magnitudes of C_{SOA} since $C_{O_3,out}$ is different. The change in C_{SOA} going from a New to a Used case is the smallest within the 10% OA/90% RA case since these cases have the lowest C_{O_3} due to their having the lowest fraction of outdoor air. The differences in New versus Used cases are very similar within the other two flow cases of 100% OA and 50% OA/50% RA. This similarity is because going from the 100% OA case to the 50% OA/50% RA increases C_{terp} (due to indoor emission) but decreases the

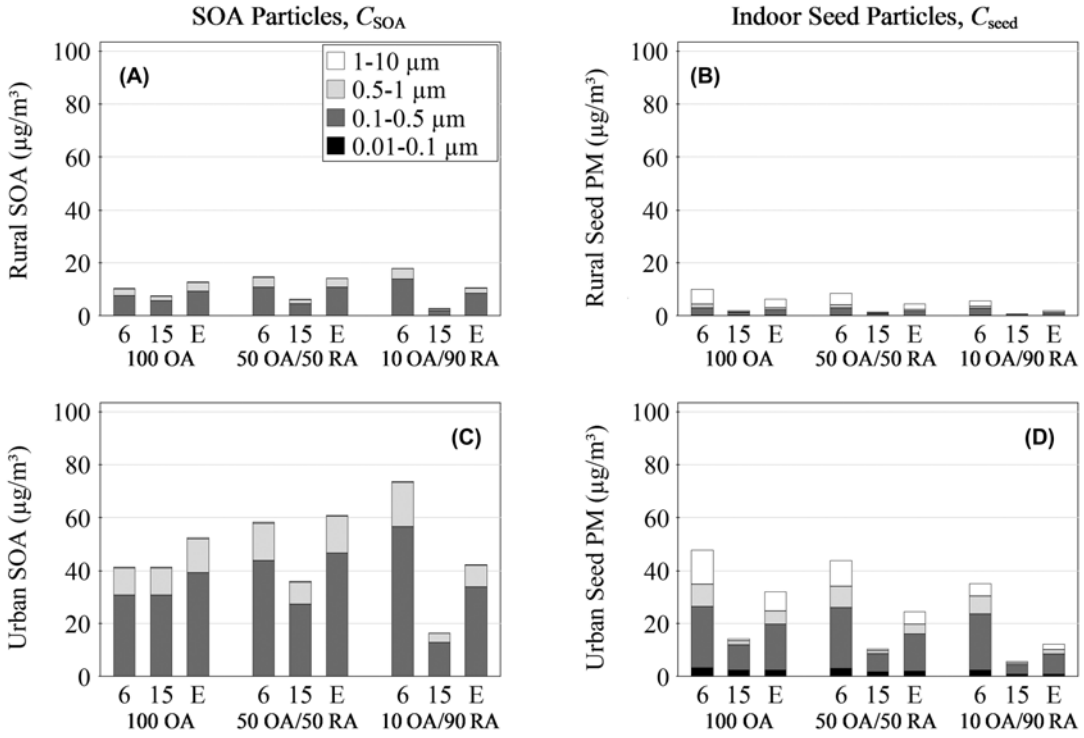

Figure 5 *Size-bin-resolved commercial rural (A) C_{SOA} and (B) C_{seed} and commercial urban (C) C_{SOA} and (D) C_{seed} (µg/m³ or multiply by 6.24×10^{-11} to convert to lb/ft³) for PM filtration parameters within each flow case.*

C_{O_3} (due to outdoor-to-indoor transport) at amounts that result both in comparable values of absolute C_{SOA} and comparable differences in New versus Used cases.

In general, the η_{O_3} parameter has less of an effect on C_{SOA} in the residential than the commercial model, for two reasons. The first is that the Used value for η_{O_3} is much less in the residential than the commercial model. The second reason for the reduced relative influence of η_{O_3} in the residential model is because ozone-laden air is only introduced indoors from outdoors in the residential model through infiltration. In the commercial model, ozone is introduced indoors from outdoors via both infiltration and ventilation [see Equation (4)], so η_{O_3} has an effect on source as well as removal mechanisms. One potential limitation of our approach is that the values for η_{O_3} from Zhao et al. (2007) were determined with ozone removal tests at face velocities of 0.4 cm/s (47.2 ft/h), which is much lower than would likely occur in most HVAC systems. However, their mean residential value of 10% is comparable to values reported in two other studies conducted at larger face velocities (Hyttinen et al. 2003; Bekö et al. 2007). Their mean commercial value of 41% from Zhao et al. (2007) is somewhat higher than other reported commercial values, but these larger values could reflect a choice of venue from which used filters were procured (e.g., a filter from an office would likely have a lower value of η_{O_3} than a restaurant because of unsaturated cooking oils that would settle on the particle-cake on a used filter from a restaurant).

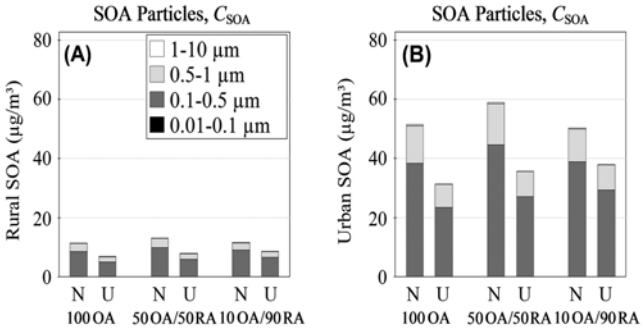

Figure 6 *Size-bin-resolved commercial (A) rural and (B) urban C_{SOA} (µg/m³ or multiply by 6.24×10^{-11} to convert to lb/ft³) for O_3 filtration parameters within each flow case.*

To compare the influence of HVAC system components and operation on SOA concentrations, we developed a steady-state model. In reality, one or both reactant sources and resulting concentrations are likely to be transient in nature. Outdoor ozone concentrations rise and fall according to a diurnal cycle, and indoor ozone concentrations due to outdoor-to-indoor transport thus also fluctuate diurnally, albeit at lower concentrations than and lagging slightly in time behind those outdoors (Weschler 2000). However, indoor emissions of

ozone, such as from in-duct ESPs (Viner et al. 1992), portable ion generators (e.g., Waring et al. 2008), or office equipment (Lee et al. 2001), can produce either a steady-state or time-averaged indoor emission, which leads to a portion of the indoor ozone concentration having a steady or nearly steady baseline with the influence of the diurnal outdoor ozone concentration added over it. Also, most terpenoid emissions indoors are likely to be nearly pulse emissions, such as the floor mopping event used here, or the application of a surface cleaner (Singer et al. 2006b) or consumer product (Corsi et al. 2007). However, plug-in air fresheners emit terpenoids at a nearly steady rate over a time-scale of days (Singer et al. 2006b). Thus, the resulting C_{SOA} from our models should not necessarily be interpreted as those to which occupants would be exposed over long periods of time. Nevertheless, this modeling effort effectively illustrates the relative effects that HVAC components and operation strategies can have on indoor SOA concentrations.

It is important to revisit other assumptions of our approach. The assumptions that have the largest impacts on SOA formation are those related to the input parameters of the models. Where possible we used values from the literature, but there are numerous gaps in the research as well as assumptions and limitations described in the text. Therefore, we suggest caution in applying the results to a real building without sufficient knowledge of parameter values and without validation of the results presented here. Another important assumption is that the entire building was well-mixed, despite the fact that the emission of the terpenoids and the removal of ozone on HVAC components are likely to occur in a single zone in the buildings. However, given the relatively small losses of SOA due to deposition onto surfaces as compared to air exchange and filtration losses, this assumption will likely have a small impact on the final results. A further assumption is that this entire analysis considered particles as spheres. This assumption is better for SOA than particles of outdoor origin, since SOA are formed from gas-phase products that condense into spheres, the form that results in the lowest free energy of the aerosol. Finally, we assumed one volume, V, each for the residential and commercial model. For the model Equations (1) through (4), V occurs in Equations (3) and (4), which include indoor emissions of terpenoids and ozone, respectively. Equation 3, however, is unaffected by a change in V, since the terpenoid emission is function of the floor area, which changes linearly with V, since the ceiling height is fixed. Only Equation (4) when it includes an indoor emission of ozone by the ESP, E_{O_3}, is affected by a change in V. As V decreases, the influence of E_{O_3} on C_{O_3} increases, and vice versa. Thus, for all scenarios without an ESP, C_{SOA} is independent of V.

The terpenoid emission and reaction assumptions deserve their own discussion. The floor mopping emission in the residential model led to a $C_{terp,m}$ of 592.9 ppb for all scenarios. The floor mopping emission in the commercial model led to different $C_{terp,m}$ values, depending on the ventilation rate, and the range of $C_{terp,m}$ was 157.0 to 1026 ppb, the median 296.5 ppb, and the mean 493.3 ppb. These concentrations are largely on the same order as those in Singer et al. (2006b) for the floor mopping emission in a 50 m^3 (1765 ft^3) test chamber with an air exchange rate of 0.5 h^{-1}. For reference, the odor threshold of d-limonene is 440 ppb (Devos et al. 1990). We also assumed that the only loss of terpenoids was by ventilation and that there was no change in terpenoid concentrations due to the reaction with ozone [see Equation (3)], which is a reasonable assumption because we focus on the relative effect caused by the HVAC system operation. Moreover, in Coleman et al. (2008), the reacted ozone changed much more than the reacted terpenoids, at 90% decrease versus a 25% decrease, respectively. Terpenoids do adsorb to building surfaces (Singer et al. 2004) and HVAC system components (Fick et al. 2005), but adsorption was neglected since it does not affect steady-state concentrations. The terpenoid emission in our model and its SOA yield was based on a pine-oil cleaner with d-limonene as one of its major constituents. The terpene d-limonene has the highest SOA mass formation potential of common indoor terpenoids (Weschler and Shields 1999; Ng et al. 2006), so different consumer products with other reactive terpenes or terpene alcohols would likely result in lower SOA concentrations.

Using the HVAC system to reduce the amount of SOA that forms in buildings is a worthwhile goal. To cause the biggest reduction in SOA exposure, using a high-efficiency filter will have the most impact, followed by ozone control strategies, such as eliminating ozone sources such as an ESP or reducing outdoor-to-indoor transport of ozone with an activated carbon filter. HVAC flow strategies can also have a large effect at reducing SOA concentrations indoors. For instance, at times when ozone concentrations are low outdoors, a commercial HVAC system could deliver 100% ventilation air to dilute concentrations resulting from indoor terpenoid emissions. Conversely, when there are terpenoid emissions that coincide with high outdoor ozone concentrations, the highest allowable level of recirculated air could be used. In a residential home, setting the HVAC system on continuous recirculation will likely reduce SOA concentrations.

As shown in Figures 3 through 5, the amount of SOA formed has the potential to constitute a large fraction of the total mass concentration of indoor particles, which has implications for particle loading onto HVAC equipment. The deposition of particles onto HVAC filters, coils, and ducts can lead to increased energy use and secondary indoor air quality problems. The SOA size range of 0.1 to 1 μm predominately deposits only on HVAC filters and penetrates through coils and ducts (Waring and Siegel 2008). Filter loading can increase the pressure drop across a filter, which can lead to increased energy use over time in HVAC systems with a variable speed fan. Filter loading also affects indoor air quality, in both positive and negative ways. Positively, removal of SOA by a filter can improve the indoor air by removing the SOA particles themselves as well as potentially increasing the removal efficiency of the filter, which can increase with loading for porous-media

filters (Hanley et al. 1994). Negatively, however, filter loading can also reduce indoor air quality by decreasing the efficiency of ESPs (as well as some filters) and increasing the reactivity of porous-media filters with ozone and free radicals, which can react with the particle-cake on used filters to yield gaseous byproducts (Hyttinen et al. 2003; Bekö et al. 2006, 2007; Zhao et al. 2007).

SOA formation also has further implications for human exposure. SOA particles in the size range of 0.1 to 1 μm have the ability to penetrate the upper airway regions and deposit in the alveolar sacs of the lungs (Hinds 1999). Additionally, Rohr et al. (2002) exposed mice to the oxidation products (both particle and gas-phase) of limonene and ozone reactions and noted acute upper airway irritation in the mice. However, Wolkoff et al. (2008) later showed that the gaseous elements, rather than the particle phase elements, of the d-limonene/ozone reaction may be responsible for acute upper airway irritation. There has been no research into the chronic effects of exposure to SOA. Since the reactions between terpenoids and ozone lead to both particle-phase SOA and gas-phase products (such as formaldehyde, other more complex aldehydes, and carboxylic acids), for HVAC strategies that lead to the same final concentration of SOA those that reduce the actual formation are preferable to those that remove SOA after formation. Similarly, an SOA reducing strategy of removing ozone in the HVAC stream would preferably be accomplished with an activated carbon filter rather than a used porous-media particle filter, since the activated carbon filter would lead to lower concentrations of gaseous reaction products.

CONCLUSION

This paper presented the results of a modeling investigation that predicted the relative influence of HVAC system components and operation strategies on SOA formation due to ozone reactions with terpenoids. We reported the results for 324 unique residential scenarios (162 each of in a rural and urban climate) and 486 unique commercial scenarios (243 each of in a rural and urban climate). The resulting loading rates varied over a range of an order of magnitude, depending on the inputs of the varied HVAC parameters. For each set of unique parameter combinations, the median and range of resulting SOA concentrations were as follows. In a rural setting, the median residential and commercial SOA concentrations for all simulations were 17.4 μg/m^3 (1.09×10^{-9} lb/ft^3), with a range of 2.47 to 27.0 μg/m^3 ($1.54 \times 10^{-10} - 1.68 \times 10^{-9}$ lb/ft^3), and 10.6 μg/m^3 (6.61×10^{-10} lb/ft^3), with a range of 1.81 to 26.3 μg/m^3 ($1.13 \times 10^{-10} - 1.64 \times 10^{-9}$ lb/ft^3), respectively. In an urban setting, the median predicted residential and commercial SOA concentrations were 68.0 μg/m^3 (4.24×10^{-9} lb/ft^3), with a range of 14.7 to 108 μg/m^3 ($9.17 \times 10^{-10} - 6.74 \times 10^{-9}$ lb/ft^3), and 44.8 μg/m^3 (2.80×10^{-9} lb/ft^3), with a range of 11.6 to 105 μg/m^3 ($7.24 \times 10^{-10} - 6.55 \times 10^{-9}$ lb/ft^3), respectively. Based on our model and its input parameters, the following further conclusions can be drawn from this work:

- More indoor SOA is formed in urban areas due to higher ambient ozone concentrations. However, SOA concentrations in rural areas may be a higher fraction of the total indoor particles than urban areas.
- Much of the resulting SOA distribution is in the 0.1 to 1 μm diameter size range, and excluding those from indoor sources, indoor particles above or below this range are likely of outdoor origin.
- Residential SOA concentrations are most influenced by the particle filtration efficiency, whether the HVAC system cycles on and off or runs continuously, and the indoor set-point temperature.
- Commercial SOA concentrations are most influenced by the particle filtration efficiency, whether ozone is removed by HVAC filters or other ozone sinks, the fraction of ventilation versus recirculated air, and the indoor set-point temperature.
- The enhanced particle removal capability of an electrostatic precipitator (ESP) can be substantially dampened by its ozone generation and consequent SOA formation.
- The relative effect of the ozone emissions of an ESP on SOA formation are larger in a rural than an urban area since ambient ozone concentrations are lower.
- The filtration of ozone by an HVAC component is an effective way to reduce indoor SOA concentrations. To reduce indoor SOA, methods that lead to the lowest formation of gaseous byproducts, such as ozone filtration by activated carbon, are preferable.

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NOMENCLATURE

C_{O_3}	= steady-state indoor mass concentration of ozone (μg/m^3 or lb/ft^3)
$C_{O_3,m}$	= steady-state indoor molar concentration of ozone (ppb)
$C_{O_3,out}$	= steady-state outdoor mass concentration of ozone (μg/m^3 or lb/ft^3)
$C_{O_3,m,out}$	= steady-state outdoor molar concentration of ozone (ppb)
C_{seed}	= steady-state indoor seed particle concentration of outdoor origin (μg/m^3 or lb/ft^3)
$C_{seed,PM1.1}$	= steady-state indoor seed particle concentration of outdoor origin < 1.1 μm (μg/m^3 or lb/ft^3)
C_{SOA}	= steady-state indoor SOA mass concentration (μg/m^3 or lb/ft^3)
C_{terp}	= steady-state indoor mass concentration of terpenoids (μg/m^3 or lb/ft^3)
$C_{terp,m}$	= steady-state indoor molar concentration of terpenoids (μg/m^3 or lb/ft^3)

d_p = particle diameter (μm)
E_{O_3} = steady-state ozone emission rate of the electrostatic precipitator (μg/h or lb/h)
E_{terp} = steady-state terpenoid emission rate of the floor mopping event (μg/h or lb/h)
F_{RH} = formation factor that adjusts for differences in relative humidity (–)
F_T = formation factor that adjusts for differences in temperature (–)
k = reaction rate constant of terpenes and ozone ($ppb^{-1}\ h^{-1}$)
n_{seed} = steady-state indoor seed particle mass distribution function ($\mu g/\mu m \cdot m^3$ or $lb/\mu m \cdot ft^3$)
$n_{seed,N,out}$ = outdoor particle number distribution function ($\#/\mu m \cdot m^3$ or $\#/\mu m \cdot ft^3$)
$n_{seed,out}$ = outdoor particle mass distribution function ($\mu g/\mu m \cdot m^3$ or $lb/\mu m \cdot ft^3$)
n_{SOA} = steady-state indoor SOA mass distribution function ($\mu g/\mu m \cdot m^3$ or $lb/\mu m \cdot ft^3$)
p_p = penetration of particles through cracks in the building envelope (–)
Q_i = infiltration air volumetric flow rate (m^3/h or CFM)
Q_r = recirculation air volumetric flow rate (m^3/h of CFM)
Q_v = ventilation air volumetric flow rate (m^3/h or CFM)
RH = relative humidity of the space (%)
S = surface area to volume ratio in the space (m^{-1} or ft^{-1})
T = temperature of the space (°C or °F)
u^* = friction velocity (cm/s or ft/h)
V = volume of the space (m^3 or ft^3)
v_d = deposition velocity (m/h or ft/h)
$Y_{g,sr}$ = size-resolved mass distribution yield of SOA (μm^{-1})
β_p = particle deposition loss rate (h^{-1})
β_{O_3} = ozone deposition loss rate (h^{-1})
$\Delta\beta_{ESP}$ = difference in β_p with and without the ESP operating in Wallace et al. (2004) (h^{-1})
η_{ESP} = size-resolved efficiency of the in-duct ESP in Wallace et al. (2004) (–)
η_p = particle removal efficiency for the filter (–)
η_{O_3} = ozone removal efficiency for the filter (–)
λ_i = infiltration air exchange rate (h^{-1})
λ_r = recirculation air exchange rate (h^{-1})
$\lambda_{r,townhome}$ = recirculated air exchange rate in Wallace et al. (2004) (h^{-1})
λ_v = ventilation air exchange rate (h^{-1})

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