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Numerical modeling of particle generation from ozone reactions with human-worn clothing in indoor environments

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HIGHLIGHTS

- We modeled particle generation from ozone reactions with human-worn clothing.
- The models can predict size-resolved particle number and mass concentrations.
- The models provided insights into the overall particle-generation mechanism.
- Ozone/clothing reactions could be important sources of indoor ultrafine particles.

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ABSTRACT

Ozone-terpene reactions are important sources of indoor ultrafine particles (UFPs), a potential health hazard for human beings. Humans themselves act as possible sites for ozone-initiated particle generation through reactions with squalene (a terpene) that is present in their skin, hair, and clothing. This investigation developed a numerical model to probe particle generation from ozone reactions with clothing worn by humans. The model was based on particle generation measured in an environmental chamber as well as physical formulations of particle nucleation, condensational growth, and deposition. In five out of the six test cases, the model was able to predict particle size distributions reasonably well. The failure in the remaining case demonstrated the fundamental limitations of nucleation models. The model that was developed was used to predict particle generation under various building and airliner cabin conditions. These predictions indicate that ozone reactions with human-worn clothing could be an important source of UFPs in densely occupied classrooms and airliner cabins. Those reactions could account for about 40% of the total UFPs measured on a Boeing 737-700 flight. The model predictions at this stage are indicative and should be improved further.

1. Introduction

Particulate air pollution is a matter of serious health concern for humans. Several studies have associated exposure to outdoor particles with increased morbidity and mortality risks (Dominici et al., 2006; Pope and Dockery, 2006). However, outdoor exposure to particles is usually less significant than indoor exposure because people spend the majority of their time indoors. Therefore, it is presumable that many of the adverse health effects that are apparently due to outdoor particles are actually caused by exposure indoors (Jones, 1999). Hence, it is imperative to characterize people's exposure to particles in indoor environments.

Several investigations have identified ozone reactions with terpene-containing consumer products as an important source of indoor particles (Sarwar and Corsi, 2007; Weschler and Shields, 1999). For example, Long et al. (2000) found that ozone/terpene reactions drastically increased (with a peak increase of 7–100 times) the particle number concentrations in Boston-area homes. They also found that more than 50% of those particles (by volume) were ultrafine in nature. Such ultrafine particles (UFPs) can deposit deep into the lungs and presumably are responsible for many of the adverse health effects associated with particles (Donaldson et al., 1998; Sioutas et al., 2005). Hence, several researchers focused on

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measuring UFP generations from ozone/terpene reactions (Coleman et al., 2008; Destaillats et al., 2006). They found that such reactions were indeed a major source of indoor UFPs. A recent investigation has identified ozone reactions with squalene (a triterpene) present in human skin-oils as another potential source of indoor UFPs (Wang and Waring, 2014).

In addition to experimental investigations, some researchers have developed models for predicting particle generation from ozone/terpene reactions on the basis of detailed chemical mechanisms (Leungsakul et al., 2005; Sarwar et al., 2003). These models can provide reasonable predictions of particle mass generation. However, they cannot compute particle number concentrations and size distributions, which are essential indices for characterizing exposure to UFPs (Donaldson et al., 1998; Peters et al., 1997). A recent study by Ito and Harashima (2011) developed a sectional model for simulating particle generation from ozone reactions with D-limonene (a terpene) together with particle size distributions. However, their model did not include condensational growth, which is the predominant mechanism for increase in particle size in such reactions.

Therefore, to better understand ozone-initiated particle generation and human exposure to the particles, this investigation developed a numerical model for computing size-resolved particle concentrations based on physical formulations of particle dynamics. We used the model to analyze particle generation from ozone reactions with human-worn clothing, which were recently identified as a potential source of indoor UFPs (Rai et al., 2013). The model was then used to compute concentrations of such ozone-initiated particles in various indoor environments such as buildings and airliner cabins.

2. Method

This investigation simulated the generation of ozone-initiated particles from reactions with human-worn clothing as measured in an environmental chamber. The details of the chamber experiments were provided in Rai et al. (2013) and are briefly summarized here. Fig. 1 shows the schematic of the chamber and the anticipated particle-generation mechanism. Experiments were conducted in a medium-scale environmental chamber (dimensions: 1.8 m × 1.7 m × 1.7 m) containing a steel box (dimensions: 0.2 m × 0.4 m × 1.2 m) at its center. The box was used as a human simulator, with its temperature maintained at 31 ± 1 °C and a cotton T-shirt stretched over it. The T-shirt was soiled with skin-oils by a human subject's sleeping in it and was the primary site for ozone reactions. The chamber was ventilated with outdoor air enriched with ozone, which reacted with the T-shirt to generate particles. The experiments measured the time-varying concentrations of ozone and particles at the chamber inlet and exhaust. The measurements therefore presented a well-controlled and challenging case study for developing models to study ozone-initiated particle generation from the reaction of ozone with clothing.

To model particle generation, this investigation assumed that ozone reacted with skin-oils on the T-shirt to produce a hypothetical semi-volatile organic compound (SVOC) in the vapor phase, as shown in Fig. 1. Our assumption was based on the fact that skin-oil constituents such as squalene react with ozone and produce SVOCs (Fadeyi et al., 2013). The concentration of the SVOC vapor then increased in the chamber because its production rate from ozone/skin-oil reactions was higher than its removal rate by ventilation and deposition. The SVOC vapor concentration subsequently crossed its nucleation threshold, and new particles (liquid droplets of SVOC) were generated. Note that if ventilation and deposition rates were sufficiently high, particle generation would not take place because the SVOC vapor concentration would never cross its nucleation threshold.

These freshly nucleated particles then served as condensation sites for the SVOC vapor and subsequently reduced its concentration below the nucleation threshold, preventing further nucleation. From this time onward, condensation was predominant, which led to growth in particle size and consumption of the SVOC vapor. Furthermore, the number of particles in the chamber was also decreasing because nucleation had stopped and particles were continuously removed by ventilation and deposition, as illustrated in Fig. 1. This in turn reduced the number of available condensation sites for the SVOC vapor, and its concentration started to increase again, producing another nucleation burst of particles. This cycle of particle generation, growth, and removal continued until the ozone/skin-oil reactions could not generate sufficient SVOC vapor in the chamber. Note that coagulation of particles was ignored in the above description because the particle concentrations in most cases were too low for coagulation to have a significant influence (Hussein et al., 2009).

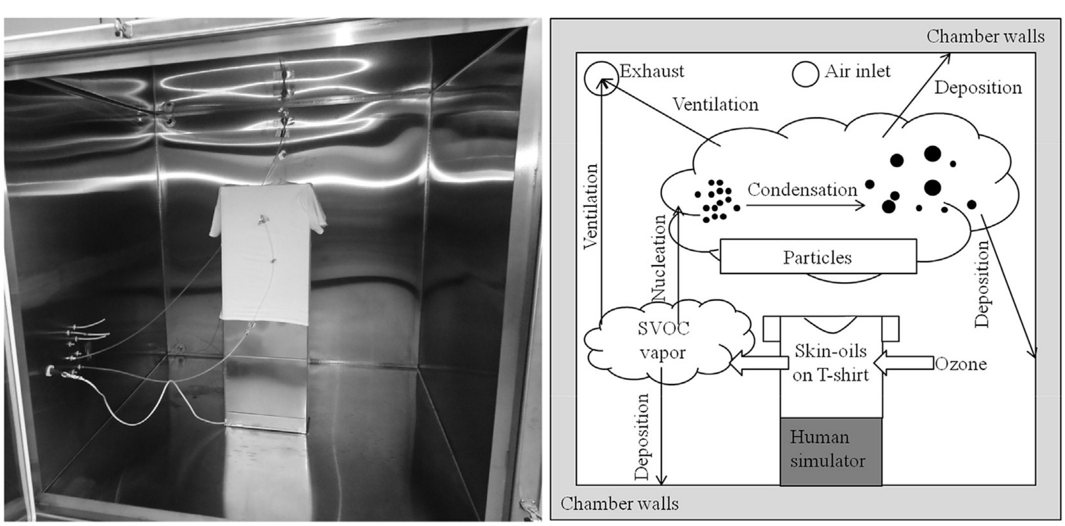

Fig. 1. Schematic of the environmental chamber used to study ozone reactions with a T-shirt on a human simulator and an illustration of the particle-generation mechanism.