

Experimental study of environmental tobacco smoke particles under actual indoor environment

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Abstract

Environmental tobacco smoke (ETS) is a major source of human exposure to airborne particles. In order to provide more information necessary for human exposure investigations, the aim of the work presented here is to investigate experimentally the variation of the ETS particle concentration and size distribution under an actual indoor environment, in a room of 30 m³, using human smokers. The effect of number of cigarettes and brands of cigarettes, the effect of sampling location and the effect of ventilation rates were investigated. The results indicated little difference in the geometric mean diameter (GMD) of the ETS particles from those in background air. Under a ventilation rate of 0.03 m³/s, the concentration of the ETS particles reached a peak value at the sampling point shortly after completing the smoking process. The GMD first increased due to coagulation and diffusion deposition, and finally decreased due to the effect of ventilation. Smoking two cigarettes at the same time would increase the initial concentration and led to an increase in GMD of the ETS particles. Two different brands of cigarette with different tar contents released ETS particles of different GMDs but similar particle concentrations. Spatial variation in particle concentration was obvious only in the first 600 s of the tests and tended to fade out subsequently. Stronger ventilation would reduce the concentration and GMD of the particles.

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1. Introduction

Indoor air pollution is a major health concern in the work place (Subramanian et al., 2000; Niven et al., 2000). Since people typically spend the most of their time in various indoor environments, the level of exposure to potentially toxic and carcinogenic compounds is of particular concern. In the world, cigarette smoking is allowed in many indoor environments,

including smoking in restricted locations at the work place. Because of the known harmful effects of many of the compounds found in environmental tobacco smoke (ETS), there is much concern over exposure to ETS. This is especially true in the indoor environment where ETS concentrations can be many times the levels found in the outdoor ambient environment (Guerin et al., 1994).

ETS is a combustion product composed of sidestream smoke (SS) as well as exhaled mainstream smoke (MS) (First, 1985; Benner et al., 1989; Guerin et al., 1994). Upon smoking, ETS particles are generated and diluted with room air and undergo chemical and physical changes over time (Eatough et al., 1990).

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ETS is a complex aerosol consisting of vapor and particulate phases. At least 4700 constituents of ETS have been identified, and the number of unidentified components has been estimated to be as high as 100,000 (Green and Rodgman, 1996). ETS contains many chemical elements and compounds, and the health-impairing mechanisms caused by exposure to this mixture are not well understood. In ETS, toxic species are found in both the gas phase (for example, 1,3-butadiene) and particle phase (for example, benzo[*a*]pyrene). Sizing studies have shown that ETS particles have a size distribution that can be deposit into the human lungs efficiently. Scientific evidences have indicated that exposure to ETS can increase the risks of coronary heart disease, cancer and chronic obstructive pulmonary disease (Hackshaw et al., 1997; He et al., 1999; NCI, 1999).

Smoking indoors can significantly affect the airborne particle concentrations. It has been found that homes with smokers have increased average indoor particle levels by the order of 20–30 $\mu\text{g}/\text{m}^3$ in comparison to homes without smokers (Leaderer and Hammond, 1991; Ozkaynak et al., 1996). For non-smokers in smoking households, the home may be the dominant site of ETS exposure (Emmons et al., 1992; Jenkins et al., 1996).

The concentration of ETS particles in an actual indoor environment varies temporally and spatially. The temporal variation is strongly influenced by the frequency of smoking and the ventilation rate of the indoor environment, and the spatial variability depends on the details of airflow rates and patterns within the indoor environment (Miller and Nazaroff, 2001). Experimental investigations on the behavior of the ETS particles in indoor environment could be performed under strictly controlled laboratory conditions or using smoking machine for generating the ETS (Morawska et al., 1997; Miller and Nazaroff, 2001). Morawska et al. (1997) conducted experiments both in controlled chamber and actual indoor environments and found that the chamber experiments could not fully simulate indoor measurements. At present, there are little investigations on the changes of the concentration and size distribution of ETS particles upon generation under actual smoking conditions in a small room. However, it is of value to investigate these changes. The results of such investigation, coupled with results on the volatile organic compound contents (Xie et al., 2003), will be of value to human exposure analysis and to risk assessment. The aim of the present study is to investigate experimentally, on the actual range of particle concentration and size distribution, and the changes of their

concentration and size distribution with time, of the ETS particles generated by a human smoker in a small office.

2. Experiments and discussion

2.1. Experimental condition and facility

Experiments were carried out in a room located at the Hong Kong Polytechnic University. The room is a typical office for the academic staff of the university. It has plastered surfaces, and has a volume of 4m×3m×2.5m. The room has no windows and is served by a central air conditioning system. There is a diffuser for supply air and a return grille in the ceiling of the room. The rate of the supply air can be adjusted. During the experiments, the door of the room was closed. The closed door was not sealed, so air could escape through the gap between the door and the floor.

During the experiments, there was no special control over the temperature and humidity inside the room, but care was taken to perform the measurements during stable weather condition. The temperature and relative humidity during the measurements were in the range from 20°C to 22°C and from 50% to 55%.

In the present study, the scanning mobility particle sizer (SMPS; TSI Model 3934) was used to measure the particle distribution and total particle number concentration of the ETS particles generated upon smoking. The SMPS was operated to count particles of 14.3 nm to 749 nm using 32 size channels during the process of measurement. Each measurement of the particle size distribution took 150 s. The measurements were continued one after the other with some switching over time between consecutive measurements. The measurement started immediately upon smoking the cigarette and lasted for a period of approximately 2 h or until the ETS particle peak was no longer clearly distinguishable from the background indoor particle peak. In the results shown below, the first measurement was labeled to start at time 0 and a measurement labeled as 330 s refers to the measurement that was started at 330 s from the time smoking was started.

Particles measurements were carried out for three smoking conditions: one low-tar cigarette; two low-tar cigarettes and two high-tar cigarettes. The spatial variation of the particle concentration and the effect of ventilation rate on the particle concentration were also investigated.

Prior to each experiment, the size distribution and concentration of the background air in the room was measured to ensure that the effect of the former experiments had faded away. During the experiments, the cigarettes were smoked by people with over 10 years

of smoking experience. The smokers were able to control the rate of smoking such that each cigarette would last for approximately 5 min. During this period of time, roughly 600 mg of tobacco plus cigarette paper was combusted if one cigarette was smoked. Two commercially available brands of cigarettes with tar contents of 8 mg and 15 mg were used for the tests. The cigarettes were not special treated for the experiments.

In the case of one-person smoking, the person was smoking at a level of 1 m from the floor, and smoking facing the center of the room. The sampling tube inlet was located at 1.7 m above the floor and 1.5 m from the smoking point. In the case of two-person smoking, unless otherwise specified, the two persons were smoking at the same horizontal level of 1 m from the floor, and smoking facing the center of the room. The sampling tube inlet was located at 1.7 m above the floor and 1.5 m from each person, while the distance between the two persons was 1.5 m. The arrangement was to simulate the effect of smoking to a person standing or sitting face to face with the smokers, at a distance of 1.5 m apart.

2.2. Results and discussion

In the experiments, there was a continuous supply of fresh air from the supply diffuser. Unless otherwise specified, the rate of ventilation was adjusted to the minimum value of $0.03 \text{ m}^3/\text{s}$. The rate of ventilation was calculated by multiplying the average wind speed, which was measured at the air diffuser, with the area of the diffuser. The room has a positive pressure, compared with the outdoor environment. Moreover, the room was filled with background air at the commencement of the experiments. In order to analyze the influence of the background aerosol on the ETS particle distribution and its temporal variation, the concentration and size distribution of the background aerosol in the room was measured before the commencement of each experiment. Fig. 1 presents the measured size distribution of the

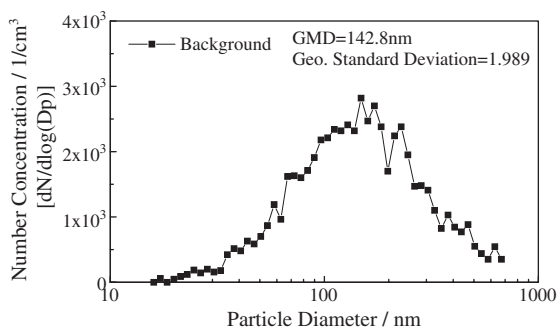


Fig. 1. Size distribution of background aerosol.

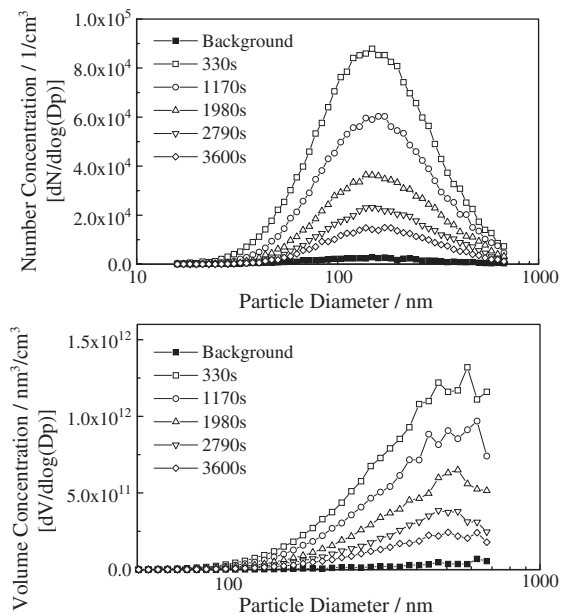


Fig. 2. Temporal variation of number and volume distributions of ETS particles due to one low-tar cigarette.

background aerosol, which had a geometrical mean diameter (GMD) of 142.8 nm, a geometric standard deviation of 1.989; and the total concentration of particles was 1.889×10^3 per cm^3 .

Fig. 2 shows the variations of the particle number distributions and volume distributions at different times after a person started to smoke a low-tar cigarette at a normal rate. The smoking lasted for approximately 5 min or 300 s. During the experiments, the door was closed and the fresh air supply was $0.03 \text{ m}^3/\text{s}$. As the particle mass is proportional to the particle volume, the volume distributions of the particles actually reflect the mass distribution of the particles. The distribution of the background aerosol is also shown in Fig. 2 for comparison. Fig. 2 shows that, essentially, the concentration of particles measured at different time after starting smoking was much larger than the concentration of background particles. Hence, we can conclude that the measured particles were mainly ETS particles, and it is reasonable to refer the measured particle concentration and size distribution as the ETS particles concentration and size distribution. The proportional of particles lying outside the measuring range of the particle sizer was small. Therefore, we can conclude that the measured particle concentration could reasonably reflect the total concentration of the particles at the sampling point. However, as far as volume is concerned, Fig. 2 shows that the measured range could not reflect the total volume of particles at the sampling point.

It could be seen from the results presented in Fig. 2 that the peak location of the number distributions and volume distributions at different times remained almost unchanged. For the distribution of the particle number, the GMD of the ETS particles shortly after the smoking was 147.0 nm, with a geometric standard deviation of 1.886. The corresponding volumetric mean diameter was 416.3 nm, with a geometric standard deviation of 1.548. The measurement results presented here showed that the initial GMD of the ETS particle was close to that of the background air. The ETS particle number distributions and volume distributions were monomodal distribution in the submicron range. Even after 3600 s, the ETS peak concentration was still clearly distinguishable from the background particle spectrum, probably because of the low fresh air supply.

The changes of the ETS particle number concentration with time for several particle sizes, chosen from the experimental data shown in Fig. 2, are shown in Fig. 3. It can be seen from the results that the concentration peaked at about 400 s after smoking started, and the particle number concentration subsequently decreased with time for all particles selected. The higher the initial concentration of the ETS particles, the faster the particle concentration decreased, especially during the period of 2500 s after the smoking started. The smoking process, which lasted for about 300 s, generates the ETS particles. Hence, all the ETS particles were generated within the 300 s. Based on the measured results, the concentration of the ETS particles came to a peak value shortly after completing the smoking action. Thereafter, the concentration dropped due to a number of particle-loss processes.

The dependence of the particle GMD and geometric standard deviation as a function of time are presented in Fig. 4. It could be seen from the results that the GMD of the ETS particles increased at the initial stage. After 3000 s, there was not much change in the GMD, for about 1000 s. Subsequently, the GMD decreased. The temporal

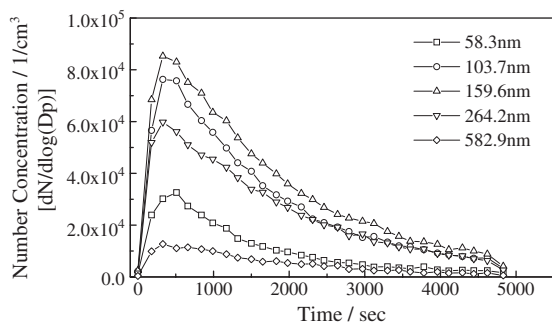


Fig. 3. Variation of number concentration of several ETS particle sizes with time.

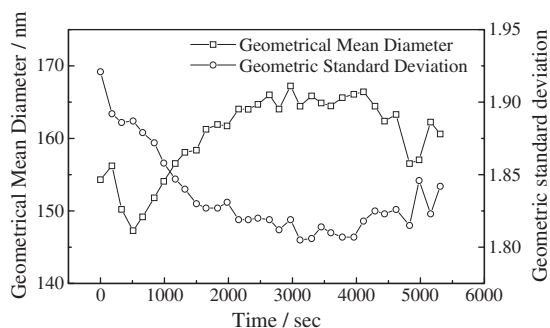


Fig. 4. Temporal variation of the GMD and geometric standard deviation.

variation of the geometric standard deviation of the ETS particles was opposite in trend to that of the GMD.

Morawska et al. (1997) reported a similar rise and fall of the count median diameter of ETS particles in a house with normal ventilation, measured over a period of 60 min; and a monotonic increase in the count median diameter in the same house with minimum ventilation, over a period of 150 min. The change of the concentration and particle GMD in a room is governed by the processes of coagulation, diffusion deposition, gravity deposition and ventilation. While all the processes will lead to a loss of particle concentration, they have different effect on the particle GMD. Coagulation will lead to an increase in particle GMD due to the coalescence of particles to form larger ones. Diffusion deposition will lead to an increase in particle GMD because the smaller particles are more liable to diffusion deposition. Gravity deposition will lead to a decrease in particle GMD because mainly the larger particles are deposited by gravity. Ventilation will lead to a dilution of the ETS particles by the supply air, and its effect depends on the GMD of the particles in the air supply. Since the initial measurements indicated that the ETS particles and background particles have similar GMD, the initial increase should be a result of the coagulation and diffusion deposition effect, which were then balanced and finally offset by the dilution effect of ventilation. Gravity deposition should be minor for submicron particles. Furthermore, it is quite possible that some of the particle growth has to do with the condensation of semi-volatile species on the surface of particles in addition to coagulation.

Fig. 5 shows the comparison of the particle GMD and concentration of one low-tar cigarette with those of two low-tar cigarettes. It could be seen from the results that the initial concentration of the ETS particles produced by two cigarettes was significantly higher than that produced by one cigarette. However, the difference in these two concentrations decreased with time and became almost

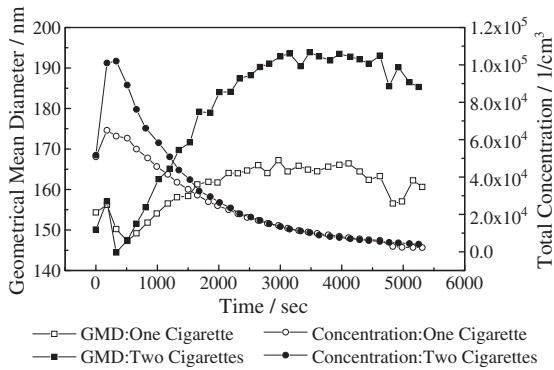


Fig. 5. Comparison of GMD and concentration of ETS particles for one low-tar and two low-tar cigarettes.

negligible about 2000s from the commencement of smoking. Fig. 5 also shows that, in both cases, the particle GMD increased during the first 3000s of the test and then remained unchanged for a period of time and then decreased. Also in both cases, the initial GMDs were not significantly different from each other. But the increase in particle GMD was more remarkable for the two low-tar cigarettes, which had a much higher initial concentration of particles, than the one low-tar cigarette, which had a much lower initial concentration of particles. The higher the initial concentration of the ETS particles, the larger the GMD grew and the faster the particle concentration decreased. Similar conclusions as to the decreasing rate of the particle concentration could be seen from Fig. 6 that presents the change of normalized concentration of the ETS particles with time for one and two low-tar cigarettes. The results presented in Fig. 6 show that, when the particle initial concentration was higher, the decrease rate of the particle concentration was faster.

It could be concluded that the variations in the particle GMD and concentration for the same kind of cigarette brand are associated with the difference in the initial concentration of the ETS particles. The decrease

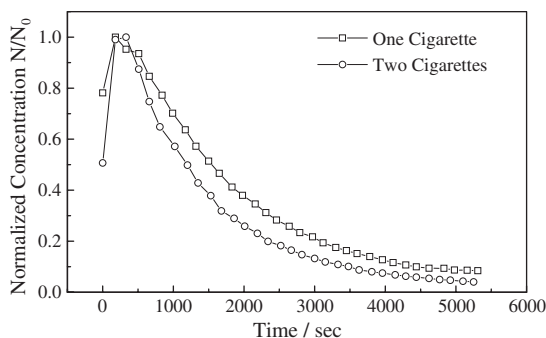


Fig. 6. Normalized concentration of ETS particles for one low-tar and two low-tar cigarettes.

of particle concentration, under such experimental conditions, is due to wall deposition, particle coagulation and ventilation. Higher particle initial concentration is more favorable for the particle coagulation and wall deposition, especially for the smaller particles.

Fig. 7 compares the size distributions of the ETS particles at different time from the commencement of smoking of two low-tar and two high-tar cigarettes by two persons at the same time. The tar contents of the two brands of cigarettes used in the experiments were 8 mg and 15 mg, respectively, and the experimental conditions were the same as those mentioned above.

It could be seen from the results presented in Fig. 7 that the ETS particle distributions for both low-tar cigarettes and high-tar cigarettes depended on the cigarette brand. For the low-tar cigarettes, the GMD of the ETS particles at the initial stage of smoking was 144.44 nm, with a geometric standard deviation of 1.974, and for the high-tar cigarettes, the corresponding GMD and geometric standard deviation of the ETS particles were 165.69 nm and 1.875, respectively. The high-tar cigarettes generated ETS particles of larger GMD and smaller geometric standard deviation.

In addition, a comparison of Figs. 2 and 7 shows that there was little change in the location of the peak values of the size distribution with time for the one low-tar case but a slight shifting of the location of the peak values of the size distribution toward larger size.

The changes of the particle GMD and concentration over time for the two brands of cigarettes are presented in Fig. 8; while Fig. 9 presents the changes of the corresponding normalized concentration with time. Fig. 8 shows that the trends of the changes in particle concentration and GMD were almost the same, but the GMD sizes were different. In these tests, the GMD of particles produced by the high-tar cigarettes were obviously larger than that produced by the low-tar cigarettes. It is believed that the difference in particle GMD is

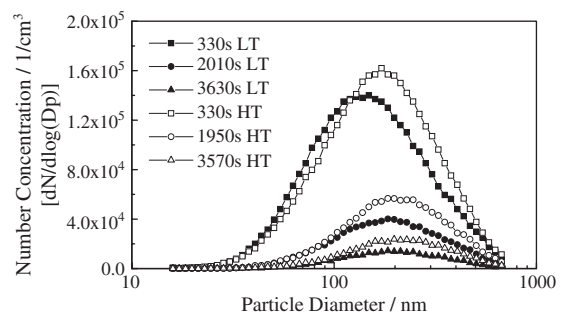


Fig. 7. Number distributions of ETS particles produced by two low-tar and two high-tar cigarettes.

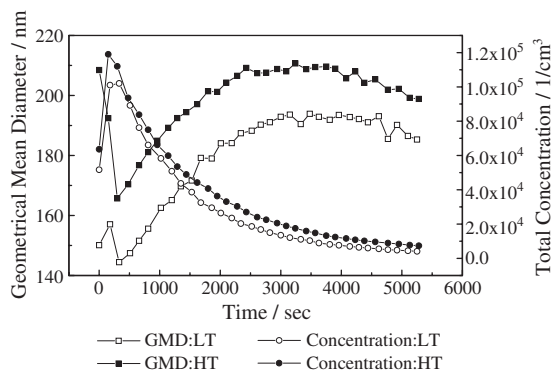


Fig. 8. GMD and concentration of ETS particles for two low-tar and two high-tar cigarettes.

most likely associated with the differences in the material of the two brands of cigarettes and the tar contents.

Figs. 8 and 9 show that there was little difference in the particle concentrations and the decreasing rate of the particles, despite of the large difference in the GMDs of the two groups of particles. This proves that the initial particle concentration has a stronger influence on the decreasing rate of the ETS particle concentration, while the particle GMD has little influence on the decreasing rate. This means that the decreasing rate of the ETS particle concentration with time is controlled mainly by the particle concentration other than the particle size.

Indoor environments are not always well-mixed, and the pollutant concentrations can vary significantly inside a room. Fig. 10 presents the variation of the particle GMD and particle concentration with time at two locations of the room. In these experiments, two high-tar cigarettes were smoked simultaneously by two persons at a normal rate in the room. The sampling point was located 1.5 m from the smoking points and 1 m above the floor for position 1, and 2 m from the smoking point and

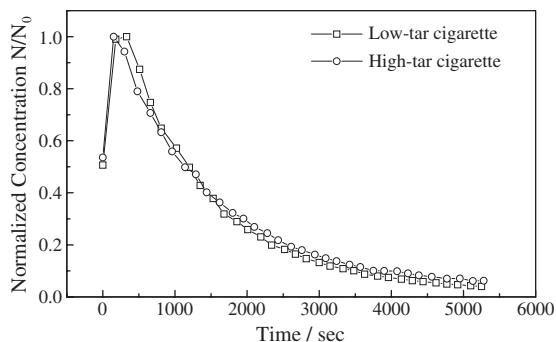


Fig. 9. Normalized concentration of ETS particles for two low-tar and two high-tar cigarettes.

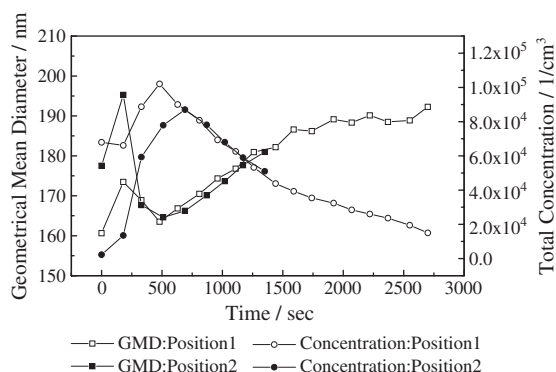


Fig. 10. GMD and concentration of ETS particles at two positions.

1.7 m above the floor for position 2. Hence, position 2 was further away from the smoking points.

The results presented in Fig. 10 show the temporal and spatial variation of the particle GMD and particle concentration. The particle GMD and particle concentration at position 1 differed significantly from that at position 2 during the first 600 s or so after the beginning of the experiments. After that, the particle GMD, the particle concentration and the decreasing rate of the particle concentration with time for the two sampling positions remained almost the same. The reason for this could be the delay of the particle diffusion. Under such experimental condition in a room with the door closed, as the air exchange and flow rate was low, the particle concentrations at different locations were different at the initial stage of the experiment. After completing the smoking process, which lasted for about 300 s, the particles would diffuse to different locations and the spatial variation was even out gradually.

The changes of the particle GMD and the normalized particle concentration with time for one low-tar cigarette at three ventilation rates of 0.03 m³/s, 0.46 m³/s and 0.64 m³/s are presented in Figs. 11 and 12. The results

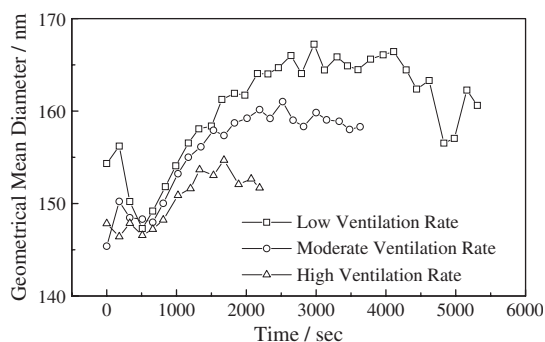


Fig. 11. Change of the particle GMD with time for different ventilation rates.

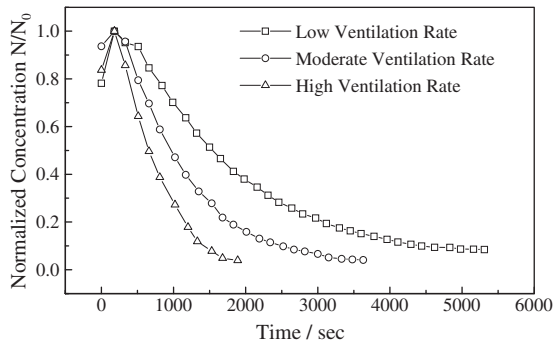


Fig. 12. Normalized concentration of ETS particles for different ventilation rates.

presented in Fig. 11 show that the GMD of the ETS particles were strongly related to the ventilation condition. However, the trends were the same. There was an initial increase in the GMD and a subsequent drop. The increase in GMD was smaller with an increase in ventilation and the decrease occurred much earlier. With an increase in ventilation rate, the rapid dilution of the particle concentration by the supply air reduced particle coagulation and diffusion deposition, and then resulting in smaller GMD.

The results presented in Fig. 12 show that the higher was the rate of ventilation, the higher the decreasing rate of the particle concentration. This suggests that ventilation is the dominant removal process for ETS particles in a room, and ventilation is a more important process for the ETS particle removal than the wall deposition process. An increase in ventilation rate can reduce the human exposure to ETS particles effectively.

The variation of the ETS particle concentration in a room is governed by the processes of coagulation, diffusion deposition, gravity deposition and ventilation. In order to analyze the effect of different processes on the reduction of the particle concentration in the room, the particle coagulation model of Otto et al. (1999) and the particle diffusion deposition model of Park and Lee (2002) were used to estimate the effect of coagulation, gravity, turbulent diffusion and ventilation on the change of the normalized concentration of ETS particles with time inside the room. The simulated results are presented in Fig. 13 for the case of smoking one low-tar cigarette. Each effect was simulated independently. In the simulation of the particle coagulation, the initial total particle concentration, the initial particle GMD and the initial geometric standard deviation obtained from the experiment were used as input. In the simulation of the effect of ventilation, the dimensions of the room and the ventilation rate of $0.03 \text{ m}^3/\text{s}$ were used. In simulating

the effect of gravity deposition, the effects of turbulent diffusion and ventilation were neglected. In simulating the effect of diffusion deposition, the effects of gravity deposition and ventilation were neglected. The values of the different parameters as contained in Park and Lee (2002) were adopted in the simulation. In Park and Lee (2002), the turbulent energy dissipation rate of $ke=36 \text{ s}^{-1}$ corresponds to the case of very strong turbulent diffusion process. In our experiments, the turbulence level was not high and the turbulent energy dissipation rate should be much lower. However, the proposed value was used in our simulation for comparison purpose, understanding that the simulated results will tend to overestimate the actual loss of particle due to diffusion deposition.

It could be seen from Fig. 13 that the decrease of the particle concentration is mainly due to the ventilation even under the condition of the lowest ventilation rate and a very large turbulent energy dissipation rate. The effect of gravity deposition is negligible, and the effect of coagulation is much lower than that of ventilation and turbulent diffusion deposition. The simulated results are similar with the conclusion deduced from the experiments.

3. Summary and conclusions

The submicron ETS particles constitute potentially human health hazard due to their ability to deposit deeper in the respiratory tract and due to their large numbers. In order to provide more information necessary for human exposure investigations, it seems especially important to conduct some studies in an actual indoor environment with experienced smokers, rather than in controlled laboratory conditions or using smoking machine, for the investigation of the evolution

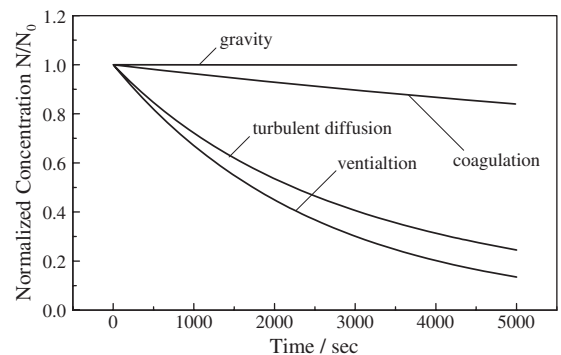


Fig. 13. Comparison of effect of gravity, coagulation, turbulent diffusion and ventilation on the change of the normalized concentration of ETS particles in a room.

behaviors of ETS particles. In this study, the temporal variation of the concentration and size distribution of ETS particles in a small office was investigated experimentally. The general conclusions from this study are as follows:

- (1) For one cigarette smoked in a 30m³ room, the concentration of the ETS particles at the sampling location peaked at about 400s after smoking started, which was shortly after finishing smoking the cigarette. The particle distribution was monomodal and the peak location of the distributions at different times remained almost unchanged. The GMD of the ETS particles increased during the first 3000s after the ETS particle generation and then remained unchanged and finally decreased for the duration of the experiments. The trend of temporal variation of the geometric standard deviation of the ETS particles was opposite to that of the GMD.
- (2) The initial concentration of the ETS particles produced by two cigarettes was significantly higher than that produced by one cigarette but the difference in particle concentration became negligible after about 2000s. The higher the initial particle concentration, the larger the GMD grew and the faster the particle concentration decreased, especially during the initial stage of cigarette smoking. Higher initial particle concentration is more favorable for the particle coagulation and wall deposition, especially for the smaller particles.
- (3) The ETS particle distributions depend on the brand of cigarette. The initial particle concentration and concentration decreasing rate for different brands of cigarettes were almost the same, but the GMD sizes were different remarkably. The GMD of high-tar cigarettes was obviously larger than that of the low-tar cigarettes, but the geometric standard deviation was smaller. It is believed that the particle size distribution has direct correlation with the material of the cigarette and the tar content.
- (4) The decreasing rate of ETS particle concentration with time is controlled mainly by the particle concentration other than the particle size.
- (5) The experimental results show that the concentration and distribution parameters of the ETS particles depended significantly on the ventilation rate in the room. Increasing ventilation rate can effectively reduce the human exposure to ETS particles.

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