



ELSEVIER

The Science of the Total Environment 196 (1997) 43–55

**the Science of the
Total Environment**
An International Journal for Scientific Research
on the Environment and its Relationship with Man

Size characteristics and ageing of the environmental tobacco smoke

Lidia Morawska^{a,*}, Milan Jamriska^a, Neville D. Bofinger^b

^aCentre for Medical and Health Physics, Queensland University of Technology, Gardens Point Campus, 2 George Street, GPO Box 2434, Brisbane Q4001, Australia

^bCentre for Medical and Health Physics and Centre for Instrumental and Developmental Chemistry, Queensland University of Technology, Gardens Point Campus, 2 George Street, GPO Box 2434, Brisbane Q4001, Australia

Received 17 July 1996; accepted 8 October 1996

Abstract

The work presented here is a study of the behaviour of the particulate phase of ETS under controlled laboratory conditions and in real indoor environments with the aim of providing information for assessment of human exposure to ETS. This paper reports investigations of the size distribution of ETS and changes to the distribution with time under a range of environmental conditions. Measurements were performed using two instruments, the Scanning Mobility Particle Sizer and the Aerodynamic Particle Sizer, which enabled the determination of the precise locations of ETS peaks at frequent short time intervals. While total particle concentrations or changes in concentrations are not specific markers of ETS, peaks related to ETS in the spectral distribution of atmospheric particles, for a properly designed experiment, are. The presence and locations of these peaks are characteristic of ETS in indoor environments and are clearly distinguishable from the background particle distribution. It is demonstrated that an initial ETS size distribution in an indoor environment about 10 min after generation by a human smoker has a major peak in the submicron range between 60 and 90 nm. The location of the peak does not depend on the relative humidity, but does depend on the way the cigarette is smoked. An increase in particle size in the range of 20 to 50%, takes place in the first 30 to 60 min after ETS generation and then remains unchanged for the duration of the experiment. A decrease in particle size (shrinkage), was not observed during these experiments. Particle shrinkage has been reported in the literature. Both the SS and the MS smoke reveal bimodal size distribution. In both cases the most significant, in terms of particle numbers, is the submicron peak. Natural ventilation, which is the most common type of ventilation for residences, is often not sufficient for effectively reducing human exposure to ETS. Controlled chamber experiments are useful for investigations of general trends in ETS size distribution and concentration and the results from such experiments, in most cases, correlate well with those from real indoor measurements. There are however, aspects which show certain differences between the two types of experiments. These differences indicate that chamber experiments can not fully simulate indoor measurements, and results from such experiments should be treated with caution when applied to exposure assessment. © 1997 Elsevier Science B.V.

* Corresponding author.

1. Introduction

Environmental tobacco smoke (ETS) is one of the most common man-made airborne pollutants yet is difficult to comprehensively characterise because of the complexities of the cigarette combustion process and of particle interactions.

Several thousand chemicals are produced by a burning cigarette (Baker, 1980). The products, the composition of which varies with burning conditions, are distributed between the gas phase and aerosol particles.

Major differences in product composition and concentration are observed between mainstream smoke (MS) which is smoke drawn through the tobacco and taken in by the smoker, and side-stream smoke (SS) which is emitted by the smouldering cigarette between the puffs (Johnson et al., 1973).

Once generated and introduced to the air, both gas and particle phases interact with each other (gas to particle conversion), with atmospheric aerosols and with the environment. Human exposure to ETS is affected by these interactions.

While numerous studies have been performed over several decades on the chemistry of ETS, its physical characteristics and its epidemiological impact, the mechanisms of ETS interactions with other components of the air under various environmental conditions are not fully understood due to difficulties in the experiments and in analysis of the data in terms of the fundamental processes occurring.

One fundamental problem is that comprehensive characterisation of ETS requires measurements of different airborne chemical compounds in different physical forms, necessitating the use of a number of different instruments.

As a result, the characterisation exercise becomes complicated and costly and in many cases, not feasible. To simplify this, certain parameters or compounds have been identified as ETS markers. Among the most commonly used markers are suspended particles, carbon monoxide and nicotine. Other markers include alkanes (Ramsey et al., 1990), solanesol, ultraviolet particulate matter and fluorescent particulate matter as indicators of particulate phase and 3-ethenylpyridine as an indicator for the gas phase (Nelson et al., 1992).

In principle, the markers should provide a good overall characterisation of the smoke. None of these markers is ideal because it is either not specific to ETS (like particles) or does not necessarily represent the behaviour of smoke in general (Rando et al., 1992). A good example is nicotine which is specific to ETS and important because of its addictive properties, but it is not found in a consistent ratio to other ETS constituents (Nelson et al., 1992) and the equilibrium between its particulate and vapour phases is unstable (McAughy et al., 1993). The use of multiple markers is often suggested as a remedy for these problems, adding to the experimental complexity.

The particulate phase of ETS is of special importance because it contains significant ETS components, for example all of the tar and most of the nicotine (First, 1985), and it can remain suspended in the air for long periods of time.

Experimental problems with characterisation of particulate phase are, firstly, related to the fact that the greatest numbers of ETS particles are in the submicron size range. Particles in this range can be detected only by using sophisticated instrumentation, which is costly and requires a skilled operator. Secondly, interactions of ETS particles in the air involve a number of physical and chemical processes which should be identified and included in the planning stages of the experiment. Finally, data analysis involves complex mathematical procedures and the application of theoretical models which provide only simplified approaches to the problems.

The previously cited experimental investigations on the behaviour of ETS particles in the air and on interactions with other constituents of the air were, in most cases, performed under strictly controlled laboratory conditions with a focus on one particular aspect. Most often, cigarettes were specially selected (or research cigarettes were used) and stored, and the measurements were structured to provide ideal conditions: for example rapid dilution of the generated smoke to prevent coagulation, separation of side stream smoke (SS) from main stream smoke (MS) etc. Smoke behaviour is often analysed for a very short period of time after generation, ranging from seconds to a few minutes. Such measurements provide reproducible

results for the particular brand of cigarettes used and for particular laboratory conditions but can rarely be applied to the characterisation of ETS under natural environmental conditions or to health risk analysis.

While there is value in such measurements, there is a need for measurements to be performed under conditions which are closer to natural environmental conditions. The results of such measurements will be more applicable to human exposure analysis and to risk assessment. The experiments should aim more at establishing general trends of ETS behaviour under a variety of natural environmental conditions, and determining the actual ranges of particle concentration and size distributions, rather than at trying to establish the repeatable conditions necessary for single numbers for concentrations and peak locations to be determined.

Many investigations concentrated on ETS (or MS or SS) behaviour immediately after the smoke was formed. Although this is important for the fate of the smoke in the air, from the point of view of human exposure it is at least of equal importance to establish ETS behaviour in the air during the entire period of time when it constitutes an exposure danger. Being in the cumulative size range, the lifetime of ETS in the air depends strongly on the air exchange rate and other environmental factors and could be up to several hours.

The properties of MS smoke emerging from a cigarette during a puff are of importance in assessment of exposure of the smoker to this component of smoke only. MS smoke is inhaled by the smoker shortly after formation in the cigarette and only a certain amount of it, with changed characteristics, is exhaled, contributing to airborne ETS. Measurements of MS smoke immediately after formation are particularly complicated because of the short time scale (of a few seconds) during which they can be conducted.

Most of the smoke is introduced to the air in the smouldering SS phase which, is inhaled by both smokers and non smokers (Davies, 1988). Inhalation of SS smoke does not take place immediately after it is formed, but sometimes hours later.

Changes in the characteristics of ETS upon aging under realistic smoking conditions have not attracted sufficient attention and there is little information about systematic studies being conducted on this issue.

The aims of the work presented here are two fold. Firstly to perform systematic investigations on the behaviour of ETS particulate phase in the air under controlled laboratory conditions and in real indoor environments for a period of time of up to a few hours in order to provide information necessary for human exposure investigations. Secondly, to evaluate to what extent chamber experiments simulate real indoor environment conditions.

These were achieved by investigations of size distribution of ETS and changes to the distribution with time and under a range of environmental conditions. The measurements were performed using two instruments, the Scanning Mobility Particle Sizer and the Aerodynamic Particle Sizer (details of the instruments are provided below) which enabled the determination of the exact locations of ETS peaks almost instantaneously. While total particle concentrations or variations in the concentrations are not specific markers of ETS, peaks related to ETS in the spectral distribution of atmospheric particles for a properly designed experiment, are. The presence and location of these peaks are characteristic of ETS in indoor environments and the peaks are clearly distinguishable from the background (indoor and ambient) particle distribution.

In the course of these studies, no systematic investigations were performed on differences in particle size characteristics between different brands of cigarettes or different smokers.

Studies on ETS interactions are also of value in the understanding of the interaction processes of many other types of aerosols, in particular those resulting from other combustion processes.

2. Experimental technique

The experimental equipment consisted of an experimental chamber, the Scanning Mobility Particle Sizer (SMPS), the Aerodynamic Particle Sizer (APS) and an ETS generator.

2.1. Experimental chamber

The large experimental chamber of volume 3 m³ was designed such to ensure a uniform flow of the air introduced to the chamber. This was achieved by locating perforated inlet and exit manifolds in opposite corners of the chamber. The air exchange rate for the chamber was controlled at 0.6 h⁻¹. For most of the experiments, chamber air was drawn from the laboratory and air from the chamber exhausted into a fumehood. Some of the measurements were performed without environmental aerosol present in the chamber, the inlet air being filtered by a HEPA filter. Humidity in the chamber was controlled by passing the air through a column chilled with liquid nitrogen before introducing it to the chamber to lower the humidity or by introducing water vapour to increase humidity. When water vapour was introduced to the clean chamber with no ETS, no changes in the background particle size distribution or concentration were observed. A small fan was installed in the chamber to ensure uniform mixing after ETS was introduced to the chamber. Under such conditions dilution of ETS was similar to normal smoking conditions.

2.2. Size distribution measurements

The TSI Model 3934 SMPS was used for size classification of submicron particles in the range from 0.01 to 0.9 μm. The instrument was connected to the chamber at approximately half height of the chamber by a very short connecting tube in order to reduce losses of particles on the walls of the tube. Larger particles in the range from 0.5 to 30 μm were studied with the TSI Model 33 APS. This latter instrument became available at a later stage of the project and was not used for measurements in residential locations.

2.3. ETS generator

An ETS generator was developed for the purpose of these studies. The generator was designed to simulate the following aspects of the smoking patterns of the tobacco user: (i) the frequency,

period and volume of inhalations; (ii) the deposition of mainstream smoke in the lungs of the smoker. It did not, however simulate the chemical and physical changes that occur in the mainstream smoke whilst in the smokers lungs.

The cigarette is contained within the Cigarette Chamber, a perspex cylinder into which three tubes are connected. One is the mainstream line into which the cigarette is inserted, another is the sidestream line and the third is the return. A flowmeter incorporating a flow restriction valve is included in the mainstream line so that the flow rate through the cigarette may be adjusted. Both the mainstream and sidestream lines enter a universal valve which selects, under computer control, which line is open.

A low throughput diaphragm pump draws alternately on mainstream and sidestream smoke. The smoke then flows through a second universal valve which rejects, through a filter, every second mainstream inhalation. This simulates the deposition of smoke in the smokers lungs. Various studies have shown that the proportion of smoke deposited in the smokers lungs varies greatly, however the average is approximately 50% (First, 1985).

The operation of the two universal valves and hence the smoking regime is user controlled by a PC through a Microbits interface card.

The same commercially available brand of cigarettes was used for all the measurements. The cigarettes were stored in an air conditioned laboratory and were not specially treated.

2.4. Experimental procedure

The experimental procedure involved the following steps:

- Purging the chamber with laboratory air or alternatively with air passed through a HEPA filter.
- Establishing a desired humidity level in the chamber. Measurements were performed for relative humidities (RH) of 38–42%, 50–55%, 75% and 95%.
- Measurements of size distribution and concentration of background aerosol in the chamber.

- Introduction of ETS to the chamber. Different regimes of ETS generation were employed:
 - A—4 puffs per min, 2 s puff duration; volume of a puff 27 cm³, 50% of MS smoke + 100% SS smoke
 - B—only SS smoke
 - C—only MS smoke, 2 s puff duration, volume of a puff 27 cm³
 - D—7 puffs per min } puff duration, volume and MS to SS ratio the same as (a) above
 - E—1 puff per min
- Measurements of the size distribution and concentration of ETS in the chamber. The first measurement was performed 10 min after ETS was introduced to the chamber and further measurements at intervals of 10 min for the first half an hour and at intervals of 30 min for the remainder of the experiment. The duration of each experiment was 180 min or until ETS peaks were no longer clearly distinguishable from the background.

In one test, in addition to environmental aerosol and ETS, smoke from a small petrol engine was introduced to the chamber to investigate the behaviour of a mixture of ETS with another combustion product.

2.5. Indoor measurements

Three residential houses and two university clubs were selected for these measurements. The measurements were performed under controlled or semi controlled ventilation conditions. There was no control over temperature and humidity, but care was taken to perform the measurements during stable weather conditions.

Two of the selected houses were modern brick houses (houses 1 and 2) and one was an old timber 'Queenslander' style house, elevated and with high ceilings (house 3).

In each of the houses, measurements were performed for two ventilation rates. Minimum ventilation occurred when all windows and doors of the house were closed and 'normal' ventilation occurred when only certain windows were open.

The relative humidity during the measurements was in the range from 38 to 58%.

Club 1 selected for the measurements was a large room, approximately 600 m² with a ceiling height of 3.5 m. There was no mechanical ventilation installed in the club; however, a number of large windows were almost always opened to increase the air exchange rate. Club 2 was approximately three times smaller than Club 1.

In residential locations measurements commenced after one cigarette was smoked in the centre of a room selected for investigations and were conducted (also in the centre of the room) until the ETS peak was no longer clearly distinguishable from the background indoor particle distribution. Background measurements of indoor and ambient particle size distribution were conducted before ETS was introduced to the indoor environment. Measurements in the clubs were performed with varying numbers of smokers present in the club and during a rock concert. Background ambient aerosol condition was monitored during the measurements.

The ETS for residential measurements was generated by a person smoking one cigarette. Initial tests revealed that the concentration of the smoke produced by the generator was lower than that generated by a smoker. A higher initial concentration was desirable for studies on particle behaviour trends.

3. Results and discussion

Most of the laboratory measurements were performed for generator-produced ETS in the chamber. Attempts were made to establish whether the generator-produced ETS differs markedly in size distribution from smoke produced by a human smoker. A comparison was made between SS smoke produced by the generator, coming from a smouldering cigarette placed directly in the experimental chamber, and from a cigarette smoked by a person, of which only SS smoke was introduced to the chamber. A similar comparison was done for generator and human produced MS smoke.

Fig. 1 presents size distribution of SS smoke of one cigarette smoked by a person. Measurements performed with the SMPS and APS are presented on one graph. Smoking duration was 7 min with

puff frequency of about 4 per min. In this experiment, Count Median Diameters (CMD) of the submicron peak was 136 nm with the geometric standard deviation of 1.77. Volume median diameter of the submicron peak was 334 nm and geometric standard deviation of 1.57. Very similar results were obtained with a smouldering cigarette left in the chamber. In this case, CMD was 135 nm, geometric standard deviation 1.75, volume median diameter 332 nm and geometric standard deviation 1.58. These results were very close to those obtained by using the generator: CMD was 135 nm, geometric standard deviation 1.86 nm, volume median diameter 292 nm, geometric standard deviation 1.55. This comparison proves that the size distribution of generator produced SS smoke is a good representation of real SS smoke.

It is demonstrated in Fig. 1 that as well as the major submicron peak, there is a second peak present in the SS smoke spectrum. The particle number in this peak is significantly lower than in the submicron peak. The CMD of the larger peak is $3.9 \mu\text{m}$ and geometric standard deviation 4.5.

The size distribution of MS smoke produced by a human smoker is presented in Fig. 2. In this particular case, the CMD of the submicron peak was 238 nm, geometric standard deviation 1.65, volume median diameter 391 nm and geometric standard deviation 1.34.

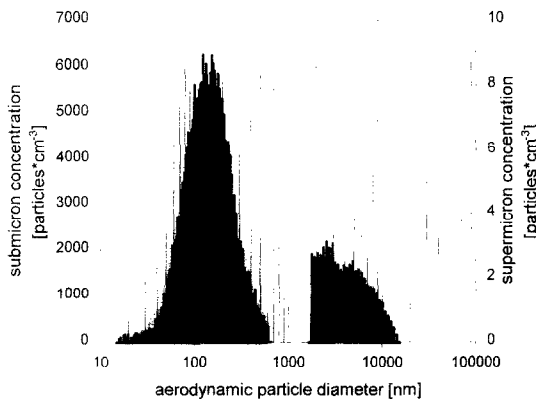


Fig. 1. Size distribution of side stream smoke (SS) produced by a human smoker. The measurements were performed independently by the SMPS and APS and are presented on one diagram.

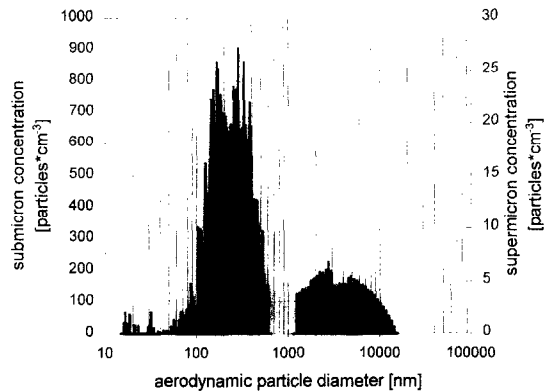


Fig. 2. Size distribution of main stream smoke (MS) produced by a human smoker. The measurements were performed independently by the SMPS and APS are presented on one diagram.

Inspection of Fig. 2 reveals that MS smoke is also bimodal with the second mode of very low concentration, with CMD estimated to be $3.4 \mu\text{m}$ and geometric standard deviation 1.9. This mode is better defined than the second mode in the SS smoke spectrum and the particle concentration in this mode is higher than in the SS smoke mode.

MS smoke produced by the generator differs significantly from that exhaled by a human smoker, the particles being smaller and having CMD of about 110 nm. The geometric standard deviation of a peak from one cigarette is about 2.3. Its higher value is due to usually much lower smoke concentration.

For a human smoker, the number concentration ratio of MS to SS smoke in the air was about 0.2 but there were no systematic studies made of the variability of this ratio. However, from the different generator tests performed, it was concluded that it was typically between 0.14 and 0.25.

The measurements presented here show that both SS and MS smoke are bimodal. Previously Chang et al., 1985 and Ueno and Peters, 1986 reported a bimodal distribution for MS smoke only. Ueno and Peters, 1986 also reported that the size distribution of SS smoke was very broad in comparison with MS smoke and there were no major differences in geometric standard deviation between the two types of smoke.

Variations in peak shape and location between the measurements taken at 10 min and 180 min after smoke introduction to the chamber are presented in Fig. 3. These measurements were performed for RH of 75% and smoke generation regime of type A. During the latter measurement, the ETS peak is still clearly distinguishable from the background environmental aerosol spectrum.

For all the measurements smoke from one cigarette was introduced to the chamber. The smoke generation and dilution processes were always the same, however, significant variations in particle concentration were encountered between individual tests. It is thought that variations are most likely associated with differences in cigarette characteristics.

While in all the tests performed, SS smoke particles produced by the generator were larger than MS smoke particles, the initial particle size for both SS and MS smoke depends on the smoke generation procedure and also varies with cigarette brand. For example, a small increase in flow rate during puff simulation can result in an increase in MS smoke particles of over 30%.

Fig. 4a and b show the dependence of peak location and concentration on time for RH of 42%, 58%, 75% and 95%. Although these measurements were performed in the absence of environmental aerosol in the chamber, results were very similar for measurements performed with environmental aerosols in the chamber.

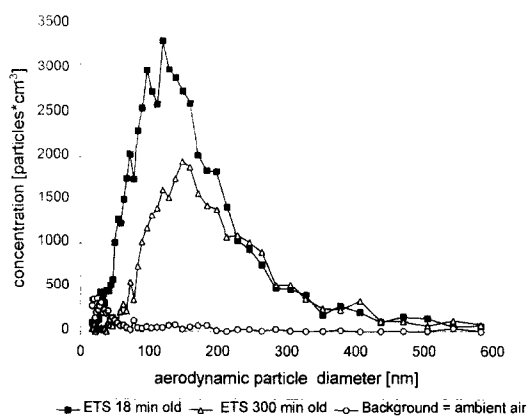


Fig. 3. ETS size distribution at the beginning and at the end of the measurement period. ETS was generated according to smoking regime A (see Section 2.4.).

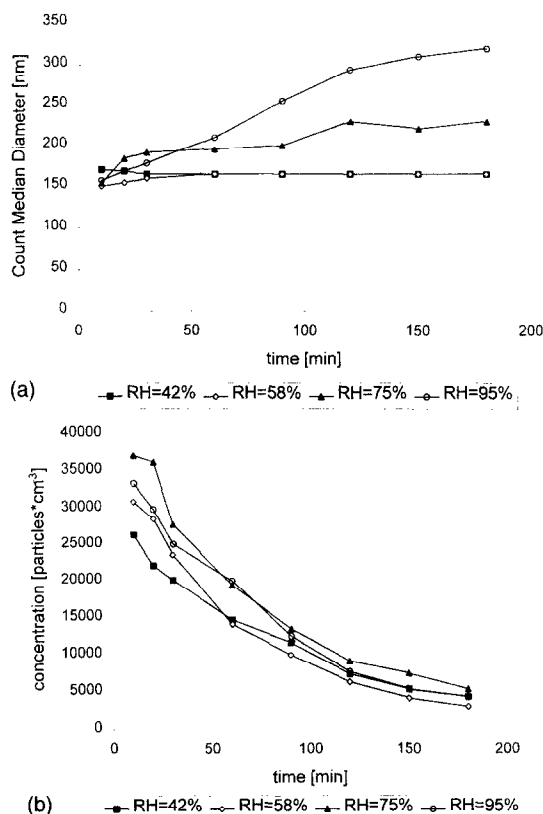


Fig. 4. Dependence of (a) count median diameter of ETS peak and (b) concentration vs. time for varying relative humidity in the chamber. Air exchange range for the chamber was 0.6 h^{-1} .

It can be seen from the results presented in Fig. 4a that peak location at the beginning of the measurements, 10 min after introduction of ETS to the chamber, does not depend on relative humidity.

For lower relative humidities of 40–42 and 52–58% the peak location remains almost unchanged for the duration of the experiment (180 min). Small increases (up to 10%) sometimes takes place after 10 or 30 min.

For higher relative humidities of 75 and 95% the peak location shifts towards larger values during the entire measurement period. The effect is more significant for RH of 95% where the total growth of particle is up to 175%. For RH of 75% the total growth of particles is up to 65%.

The increase in particle size at higher humidities observed in these studies is not in agreement with the results of Kousaka et al. (1982) who reported particle growth only at supersaturation conditions, not at 100% or lower relative humidities. The reason for this could be the differences in time scale of the investigated processes. While in this study ETS ageing was investigated for up to 180 min after formation, Kousaka's studies focussed on the period between 10 and 20 s after smoke formation.

The decrease of ETS particle concentration in the chamber can be represented by an exponential dependence:

$$C(t) = C_0 e^{-\lambda t}$$

where C_0 is ETS particle concentration at the beginning of the experiment, $C(t)$ is the concentration of time t , and λ is constant.

For lower humidities where peak locations remain more or less unchanged, the value of λ was found to be 0.57 h^{-1} . This value corresponds well with the value of 0.6 h^{-1} , the average exchange rate in the chamber. This suggests that ventilation was the dominant removal process for ETS particles in the chamber, being significantly more important than the wall deposition process. More complex analysis would have to be performed on the concentration decreases at higher humidities where particle growth by coagulation contributes to the concentration depletion.

Fig. 5a presents the size distribution of petrol smoke from a small generator introduced to the chamber and Fig. 5b presents a mixture of petrol smoke and ETS.

Fig. 6 shows the dependence of peak location on time for petrol smoke and a mixture of petrol smoke and ETS. No environmental aerosol was present in the chamber during these measurements and RH was in the range 55–60%.

The initial peak location for petrol smoke is significantly below the peak locations for ETS and changes slightly towards large sizes with time for RH of 60%. A mixture of ETS and petrol smoke (of more or less the same concentration) produces a broad peak which slightly changes towards larger sizes for the first 100 min of the measurement and then remains constant.

Fig. 7 presents the dependence of CMD of ETS and aerosols from frying food as a function of time for static conditions (air exchange rate in the chamber close to zero), the relative humidity during the measurements being about 50%. For both of these measurements particle growth continued during the entire measurement period. In the case of the dynamic conditions discussed above, the peak location remained unchanged after initial growth, suggesting that growth did not continue.

Particle growth is governed by Brownian coagulation and heterogenous evaporation (for unsaturated vapour conditions) which in theory can balance each other. For a zero air exchange rate when the decrease in particle concentration was due to deposition and coagulation only and was very slow, particle growth continued throughout the experiment. In the light of this, it can be concluded that for dynamic conditions with de-

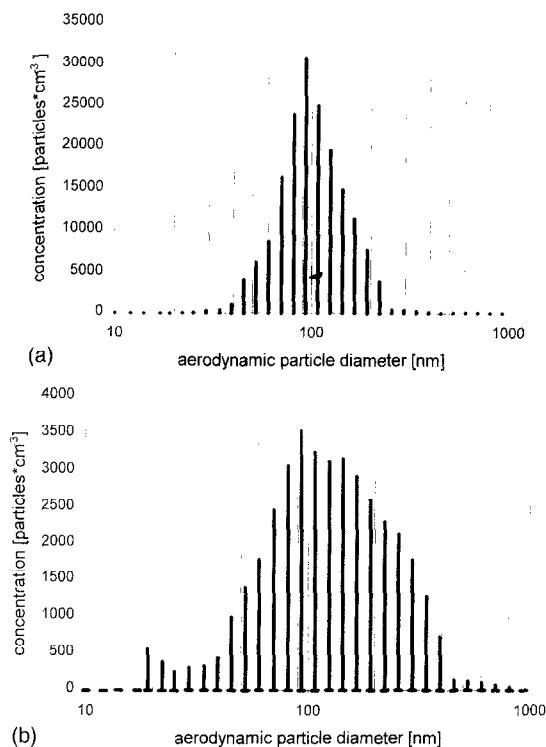


Fig. 5. (a) Size distribution of smoke from a small petrol generator shortly after introduction to the chamber. (b) Size distribution of a mixture of petrol smoke and ETS shortly after introduction to the chamber.

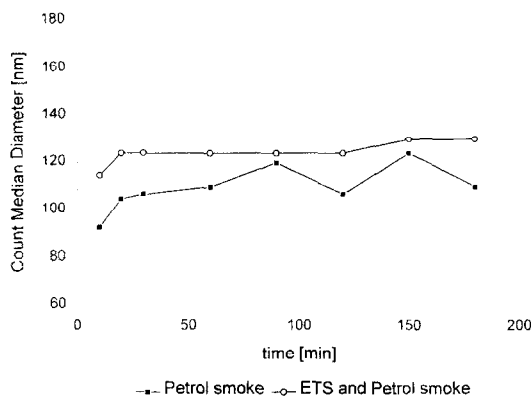


Fig. 6. Dependence of count median diameter of petrol smoke and a mixture of petrol smoke and ETS versus time. RH was in the range of 55–60%.

creasing particle concentrations, the rate of coagulation was reduced by dilution and growth occurred much more slowly than in the static case.

The decrease of particle concentration, under static conditions in the chamber, was due to wall deposition and particle coagulation. In the absence of ventilation, surface deposition is one of the main removal mechanisms of airborne aerosol particles. Under normal environmental conditions, the ventilation process is usually the dominant process in particle concentration reduction in comparison with surface deposition. As most of the studies performed here were for dynamic con-

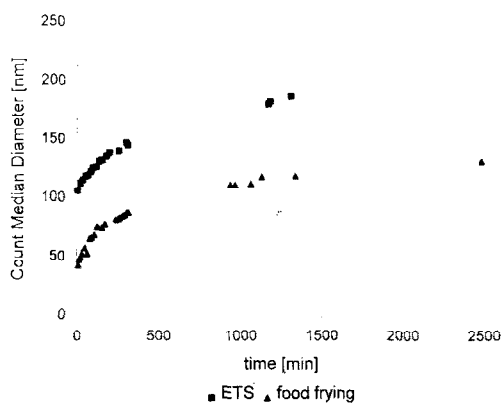


Fig. 7. Count median diameter of ETS and aerosol resulting from frying food vs. time for static conditions in the experimental chamber.

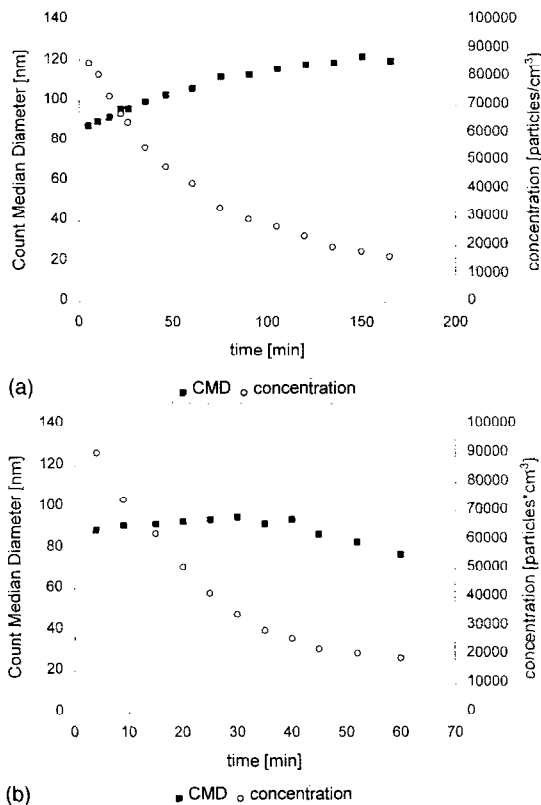


Fig. 8. Concentration and count median diameter of ETS vs. time in House 1 (brick house) with (a) minimum ventilation and, (b) normal ventilation. RH was in the range of 34–45%.

ditions, surface deposition was not investigated. Systematic studies of surface deposition were presented previously (Morawska and Jamriska, 1996). The results of chamber experiments for static conditions are included here to shed more light on particle interactions in the air. Such experiments provide, however, only limited information for exposure analysis. The half life of the ETS peak in the chamber under static conditions in these experiments was found to be of the order of a few hours. For comparison, Duc and Huynh, 1987 found that the half life for particles $< 0.3 \mu\text{m}$ in a chamber was 25.5 h.

The dependence of particle CMD and of concentration on time for minimum and normal ventilation in house 1 (brick house) are presented in Fig. 8a and b and for house 2 (brick house) for minimum ventilation, in Fig. 9.

Fig. 10a and b present results of similar measurements for house 3 (wooden house). The relative humidity during these measurements ranged from 36 to 58%.

Figs. 8–10 show that for minimum ventilation in a brick house a distinctive ETS peak is present in the air, even 3 h after one cigarette was smoked, while in a wooden house the peak is present for up to 2 h. Similar conclusions as to the concentration changes in the submicron ETS peak in a residential location of low air exchange rate, were drawn by Li et al., 1993.

For normal ventilation these times were reduced to about 1 h for a brick house and less than half an hour for the wooden house. An increase in ventilation significantly reduced exposure in both cases, nevertheless exposure time was still considerably long. When concentration remains high for longer time (as in the brick house at minimum ventilation) the CMD of the peak increases from about 90 to about 150 nm. At lower concentrations, the size distribution does not markedly change. These results are consistent with chamber experiments discussed above and can be explained by a slowing down of the coagulation process and particle growth at lower concentrations.

The air exchange rate was estimated from an exponential decay model as described above for the chamber, and was in the range of 0.55 to 0.79 h^{-1} for the measurements performed in brick

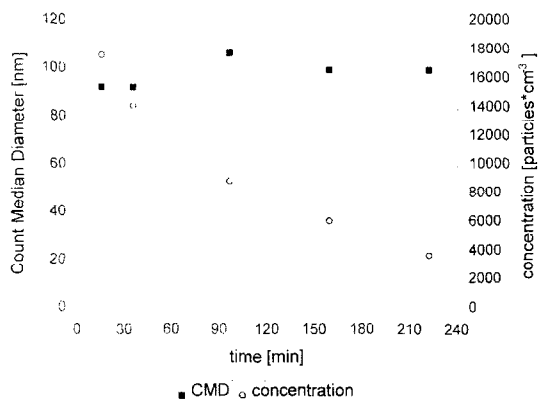


Fig. 9. Concentration and count median diameter of ETS vs. time in House 2 (brick house) with minimum ventilation. RH in the range 53–58%.

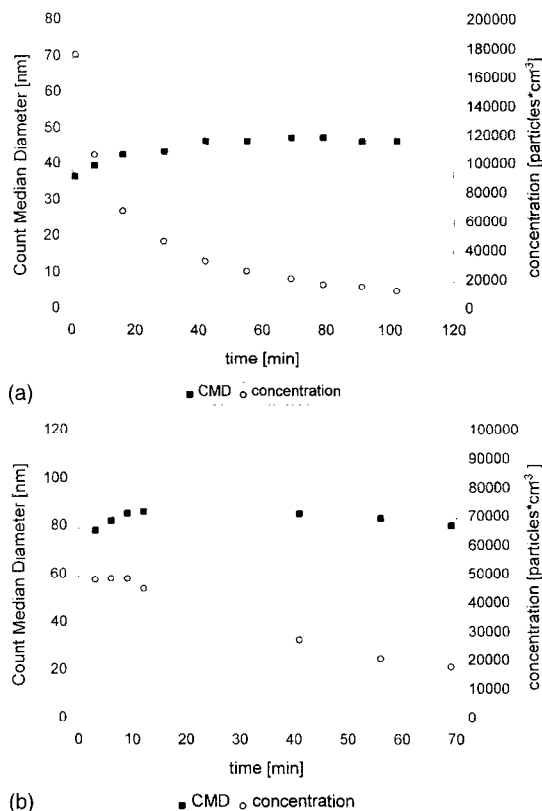


Fig. 10. Concentration and count median diameter of ETS vs. time in House 3 (wooden house) with (a) minimum ventilation and, (b) normal ventilation. RH was in the range 35–39%.

houses for minimum ventilation, and from 1.93 to 4.48 h^{-1} for normal ventilation. For the measurements performed in the wooden house, the values were 1.05 and 4.73 h^{-1} respectively.

Fig. 11 presents results for aging of ETS in house 1 for measurements performed at a high relative humidity of 85%. The increase in particle size for these measurements did not appear to be as significant as it was during chamber measurements performed at similar humidities.

Measurements of ETS characteristics in Club 1 showed that at most times when the number of smokers was small (< 10), the smoke concentration decreased rapidly with time and the ETS peak was not detectable 10–15 min after a cigarette was smoked. Under similar conditions in Club 2, the presence of ETS peak was detectable only immediately after and near to where the

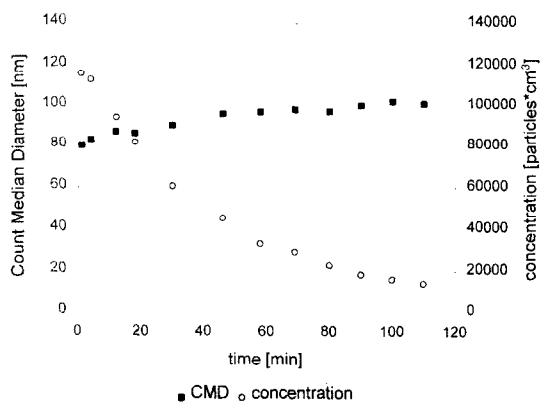


Fig. 11. Concentration and count median diameter of ETS vs. time in House 1 with minimum ventilation. RH was 75%.

cigarette was smoked. However, when the number of smokers was large, for example, during a rock concert, ETS concentration in the room was very high, up to 5×10^4 particles cm^{-3} and characterised by the stable, well defined broad peak shown in Fig. 12. Fig. 13 shows the time evolution of ETS in Club 1 during the concert.

The results of indoor experiments on ageing of ETS indicate that there is always an increase in particle size and particle shrinkage in the smoke almost never takes place. Shrinkage of ETS with time was reported by Guerin et al., 1987 and Ueno and Peters, 1986.

The initial CMD of the submicron ETS mode for all the residential and club measurements was

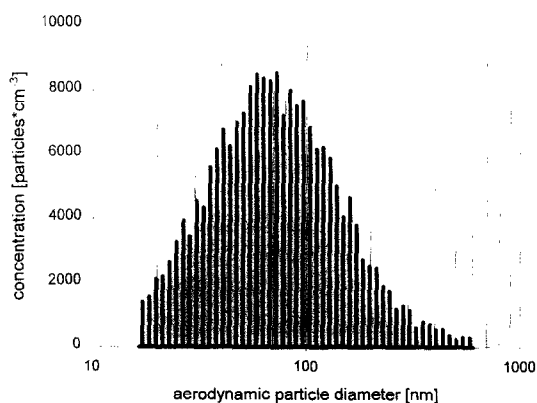


Fig. 12. Size distribution of ETS in Club 1 during a rock concert.

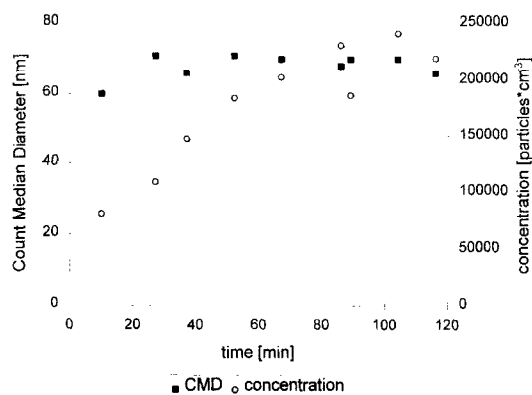


Fig. 13. Concentration and count median diameter of ETS vs. time in Club 1 during a rock concert.

lower than for the chamber measurements and was in the range from 60 to 90 nm. The lower value was for Club 1 where ETS was a mixture of smoke generated by a large number of smokers. As discussed before, the SS smoke in the chamber experiments had CMD of about 135 nm, and MS smoke generated by a smoker was over 200 nm. The reason for the discrepancy between chamber experiments and real indoor measurements is not clear and is currently being investigated further. It is expected that it could be related to some environmental factor not taken into account during chamber experiments. These factors would not necessarily be related just to the size difference between the chamber and interiors of the houses, as care was taken to conduct measurements for levels of particle concentrations of the same order of magnitude.

Particle sizes reported in the literature are scattered over a large range of values. For SS smoke, number median diameter of $0.10 \mu\text{m}$ was reported by Ueno and Peters (1986), $0.16\text{--}0.17 \mu\text{m}$ by Ishizu et al. (1980), $0.24 \mu\text{m}$ by Porstendörfer and Schraub (1972) and $0.52\text{--}0.67 \mu\text{m}$ by McCusker et al. (1980). For MS smoke Chen et al. (1986) reported values from $0.23\text{--}0.44 \mu\text{m}$, Chang et al. (1985) $0.235 \mu\text{m}$ and McCusker et al. (1982), $0.44\text{--}0.43 \mu\text{m}$.

The considerable variations in the reported values could be the result of variations in smoke generation methods and the use of different in-

strumentation. The CMD of SS smoke and MS smoke determined in this study for the chamber and indoor measurements are at the lower end of values reported in the literature.

The ETS characteristics presented here are focused on number concentration and peak location of the dominant ETS peak, not on the total particulate mass or size dependent particulate mass. Many researchers use particulate mass as an ETS marker, or indicator of ETS exposure. For example, Quant et al., 1982 based their ETS studies on PM_{10} mass fraction measurements. $PM_{2.5}$ mass concentration, and its relation to nicotine concentration, was used to characterise air quality with respect to ETS in a number of public buildings by Miesner et al., 1989; in these studies, $PM_{2.5}$ mass concentration ranged from 6.0 to 550 $\mu\text{g m}^{-3}$, with higher concentrations detected in smoking rooms or subway stations.

As demonstrated above, the greater number of ETS particles is in the submicron mode. For this reason, $PM_{2.5}$ fraction will be a better characteristic of the mass in the submicron mode. Even then, the larger mode contributes a significant fraction of the total ETS mass. Of interest for the exposure assessment would be an investigation of the distribution of toxic substances between the two modes.

4. Conclusions

The results presented here provide a good base for studies on human exposure to the presence of ETS in indoor environments. In this work, the size distribution of ETS and changes to the distribution with time were investigated under a range of indoor environmental conditions. The size distribution spectrum of ETS is a good representation of and a good marker for the ETS particulate phase. The presence and location of ETS peaks are specific to ETS in indoor environments and the peaks are clearly distinguishable from the background particle distribution. These measurements do not, however, provide information on the gas/vapour phase of the smoke. If information on a specific gas substance is required rather than on general trends of ETS behaviour, this sub-

stance should be investigated separately in addition to the investigations on the particulate phase. General conclusions from these studies are as follows:

- An initial ETS size distribution in an indoor environment about 10 min after generation by a human smoker, has a major peak in the submicron range between 60 and 90 nm. The location of the peak does not depend on the relative humidity but does depend on the way the cigarette is smoked. The values presented here are at the lower end of values reported in the literature.
- The particle size increases between 20 and 50% during the first 30 to 60 min after ETS generation and then remains unchanged for the duration of the experiment. The smaller the size of ETS, or combustion aerosol in general, the faster the particle growth and the larger the relative increase in size. A decrease in particle size (shrinkage), was not observed during these experiments. Particle shrinkage has been reported in the literature. It is possible that it takes place in a very short time after smoke generation, in the order of seconds.
- Natural ventilation which is the most common type of ventilation for residences is often not sufficient to effectively reduce human exposure to ETS. A high concentration of ETS was recorded up to 3 h after smoke generation, depending on the air exchange rate.
- Both the SS and the MS smoke reveal bimodal size distribution. In both cases the most significant, in terms of number, is the submicron peak. The peak at larger particle size, better defined for MS than for SS, has particle concentrations several orders of magnitude lower than the submicron peak. The CMD of this peak is located between 3 and 4 μm .
- The submicron ETS particles constitute potentially higher health hazard than the larger mode, due to their ability to deposit deeper in the respiratory tract and due to their large numbers. Larger particles deposit predominantly in the nasal area in normal nasal breathing. Of significance for better exposure assessment would be to investigate the distribution of toxic substances between the two modes.

– Controlled chamber experiments are useful for investigations of general trends in ETS size distribution and concentration and the results from such experiments, in most cases, correlate well with those from real indoor measurements. There are however, aspects which show certain differences in the two types of experiments. For example, an initial CMD of the submicron peak is systematically shifted towards larger values for the chamber experiments in comparison with indoor ones. Another example is the larger particle growth at higher humidities for the chamber experiments in comparison with indoor experiments. These variations indicate that chamber experiments can not fully simulate indoor measurements, and results from such experiments should be treated with caution in application to exposure assessment. The experimental findings presented here will be complemented by theoretical modelling of the interaction processes currently in progress.

Acknowledgements

The smoke generator was designed and built by Alex Zapantis as part of his MSc course. The authors are grateful to Jane Hitchins for her assistance in sample collection and data presentation.

References

- Baker, R.R., 1980. Mechanisms of smoke formation and delivery. *Recent Advances in Tobacco Smoke*, 6: 184–224.
- Chang, P.-T., L.K. Peters and Y. Ueno, 1985. Particle size distribution of mainstream cigarette smoke undergoing dilution. *Aerosol Sci. Technol.*, 4: 191–207.
- Chen, B.T., J. Namenyi, H.C. Yeh and J.L. Mauderly, 1986. *Inhal. Toxicol. Res. Inst. Annual Rep. LMF-115*.
- Davies, C.N., 1988. Cigarette smoke: Generation and properties of the aerosol. *J. Aerosol. Sci.*, 19(4): 463–469.
- Duc, T.V. and C.K. Huynh, 1987. Deposition of sidestream tobacco smoke particles in an experimental chamber. *Toxicol. Lett.*, 35: 59–65.
- First, M.W., 1985. Constituents of sidestream and mainstream tobacco smoke and markers to quantify exposure to them. In: R.B. Gammage and S.V. Kaye, (Eds.), *Indoor Air and Human Health*. Lewis Publishers, Michigan, USA.
- Guerin, M.R., C.E. Higgins and R.A. Jenkins, 1987. Measuring environmental emissions from tobacco combustion: Sidestream cigarette smoke literature review. *Atmos. Environ.*, 21(20): 291–297.
- Ishizu, Y., K. Ohta and T. Okada, 1980. The effect of moisture on the growth of cigarette smoke particles. *Beit zur Tabak Int.*, 10: 161–168.
- Johnson, W.R., R.W. Hale, J.W. Nedlock, H.J. Grubbs and D.H. Powell, 1973. The distribution of products between mainstream and sidestream smoke. *Tob. Sci.*, 17: 141–144.
- Kousaka, Y., K. Okuyama and C.-S. Wang, 1982. Response of cigarette smoke particles to change in humidity. *J. Chem. Eng. Jpn.*, 15(1): 75–76.
- Li, C.S., W.-H. Lin and F.T. Jeng, 1993. Inhaled dose assessment of submicrometer particle emissions from indoor combustion sources. *Proceedings of Indoor Air 93*, 3: 87–92.
- McAughy, J.J., A. Black, A.C. Wells, D.A. Knight and J.N. Pritchard, 1993. The fate of nicotine on ageing of environmental tobacco smoke in indoor environments. *Proceedings of Indoor Air 93, Helsinki, Finland*, 3: 97–102.
- McCusker, K., F.C. Hiller, M. Mazumder and R.C. Bone, 1980. *Clin. Res.*, 28: 841A.
- McCusker, K., F.C. Hiller, J.D. Wilson, P. McLeod, R. Sims and R.C. Bone, 1982. Dilution of cigarette smoke for real time aerodynamic sizing with a spart analyzer. *J. Aerosol. Sci.*, 13(2): 103–110.
- Miesner, E.A., S.N. Rudnick, Fu.-C. Hu, J.D. Spengler, H. Özkaynak, L. Preller and W. Nelson, 1989. Particulate and nicotine sampling in public facilities and offices. *JAPCA*, 39: 1577–1582.
- Morawska, L. and M. Jamriska, 1996. Deposition of radon progeny on indoor surfaces. *J. Aerosol. Sci.*, 27(2): 305–312.
- Nelson, P.R., D.L. Heavner, B.B. Collie, K.C. Maiolo and M.W. Ogden, 1992. Effect of ventilation and sampling time on environmental tobacco smoke component ratios. *Environ. Sci. Technol.*, 26: 1909–1915.
- Porstendörfer, J. and A. Schraub, 1972. *J. Coll. Interface Sci.*, 48: 461–469.
- Quant, F.R., P.A. Nelson and G.J. Sem, 1982. Experimental measurements of aerosol concentrations in offices. *Environ. Int.*, 8: 223–227.
- Rando, R.J., P.K. Menon, H.G. Poovey and S.B. Lehrer, 1992. Assessment of multiple markers of environmental tobacco smoke (ETS) in controlled, steady-state atmospheres in a dynamic test chamber. *Am. Ind. Assoc. J.*, 53(11): 699–704.
- Ramsey, R.S., J.H. Moneyhun and R.A. Jenkins, 1990. Generation, sampling and chromatographic analysis of particular matter in dilute sidestream tobacco smoke. *Anal. Chim. Acta*, 263: 213–220.
- Ueno Y. and L.K. Peters, 1986. Size and generation rate of sidestream cigarette smoke particles. *Aerosol Sci. Technol.*, 5: 469–476.