Modelling of indoor concentrations of ultra-fine particles based on laboratory measurements

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ABSTRACT
The objective of the present study is to apply and test a mathematical model for the determination of the strength of various indoor sources of ultra-fine particles (UFP), and the sink effect for such particles. The model is intended for further development in order to create a tool capable of predicting the concentrations of fine and ultra-fine particles in a room. Input data to the model are the ventilation rate, emission rates of ultra-fine particles from different indoor sources and properties describing sink effects. Laboratory measurements of 10 indoor sources (e.g. cigarette smoke, candles, air-freshener spray, etc.) carried out in a full-scale test chamber were utilized for model testing and verification. However, only results for burning cigarettes are presented in this paper. The source strength of such UFP was approximately 4–5 orders of magnitude greater than the source strengths of particles in several size ranges above 0.3 μm, which were also studied. Sink effects had a pronounced influence on the experimental results and were included in the model. The particle removal rate (loss rate) for UFP was 0.63 h^{-1}, while values between 0.38 and 1.89 h^{-1} were observed for the larger particle fractions. The model provides a useful tool to estimate the strength of particle sources and sinks.

INDEX TERMS
Airborne particle; Ultra-fine particulate matter; Mathematical model; Source; Sink

INTRODUCTION
Due to the contributions of indoor particle sources (such as cooking, smoking and cleaning), indoor exposures can significantly impact on short-term particulate matter exposures. Since several recent studies have demonstrated associations between such exposures and acute health effects, the characterization of short-term indoor particle concentrations is important. Humans and their activities are known to generate substantial amounts of particulate matter indoors and potentially they can have the strongest influence on short-term exposure (Garrett *et al*., 1998).

Indoor air quality models of varying complexity have been developed and verified during recent years, especially concerning volatile organic compounds. The source models can be categorized into two groups: physical models based on the mass transfer theory and empirical models based on experimental studies, partly in small test chambers and partly in full-scale test chambers (Sparks *et al*., 1996; Kraenzmer and Ekberg, 1997). In addition, experimental and theoretical research on sorption processes has been presented (Axley, 1993; Jørgensen, 1999).

More knowledge on indoor levels of small particles is needed and in particular on their generation, transport and removal. In this context, modelling can be used as a tool when analysing monitored concentration data to obtain more detailed information about the strength

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of particle sources and sinks. Such models have previously been presented to a limited extent only.

Therefore, an indoor air quality model for predicting ultra-fine particle concentrations in a full-scale chamber was applied to investigate removal and generation of particles from several indoor sources. The sink effect was calculated by comparing the decay of the measured concentration of ultra-fine particles with the decay of a tracer gas.

METHODS

Measuring Equipment

The experiments were carried out in the air quality laboratory of Danish Building and Urban Research (DBUR). The full-scale chamber has a volume of 32 m^3. The walls and the ceilings of the chamber consist of panes of glass mounted in aluminium frames and the floor is made of high-pressure laminated fibreboard. Low concentration of polluting gases and particles in the supply air are ensured by the use of a fine filter of class EU7, a charcoal filter, a fine filter of class EU7 and a HEPA filter. The air change rate in the chamber was $1.7 \pm 0.1\ h^{-1}$. Two table-fans were in operation during all experiments in order to ensure complete mixing of the chamber air. A tracer gas decay method was used to determine the air change rate and for verification of the air mixing conditions in the chamber. The tracer gas concentration decay was measured continuously over 2 h with a photo-acoustic spectroscopy (PAS) instrument from Innova, type 1302. The temperature and relative humidity were recorded during the experiments. An over-pressure of 3–11 Pa was maintained in the test chamber relative to the adjoining room throughout the experiments.

The particle concentration was continuously monitored before, during and after the various sources were activated, using three different particle counters. The results from two of these are utilized in the present paper. The concentration measurements were made in the centre of the test-chamber. One of the particle counters used (TSI model CPC 3007) was a condensation particle counter, while the second, Malvern APC 300, was an optical particle counter. The CPC 3007 enables real-time particle number concentration measurements, and data collection in the particle size range from 0.01 to greater than 1.0 μm. The sample interval for this counter was approximately 60 s. According to the manufacturer, the concentration accuracy up to 100 000 particles/cm^3 is ±20% of the reading. The concentration accuracy over 100 000 particles/cm^3 is estimated to be approximately within ±20 to ±40% of the reading.

Malvern is an optical particle counter capable of measuring different particle fractions in the size range from 0.3 to 35 μm. With the Malvern counter, the fraction between 0.3 and 1.0 μm was chosen. The sample interval for this counter was approximately 80 s. According to the manufacturer the coincidence error is less than 20% at 1 200 000 particles/ft^3.

The condensation particle counter is assumed to provide data on particles that are in close agreement with the generally accepted definition of UFP (particles smaller than 0.1 μm). This assumption was supported by the following observation: A comparison between the two instruments showed that the concentration of particles larger than 0.3 μm was negligible compared with the concentration of particles measured between 0.01 and 1 μm.

Calculation of Source Strengths

A mass balance model, previously applied for analysis of gaseous contaminant concentration was used (Kraenzmer and Ekberg, 1997). The basic assumption that govern the model are that particles are perfectly mixed within the chamber, i.e. the concentration of particles are uniform throughout the whole volume. Furthermore, the measurements showed that the concentration of particles in the supply air was close to zero $C_s = 0$.

$$\dot{V} \cdot C_s + S = \dot{V} \cdot C_I + R + V \frac{dC_I}{dt} \quad (1)$$

where

$$R = r \cdot V \cdot C_I \quad (2)$$

$$S = \left(\overline{C}_I + \frac{V}{\dot{V} + rV} \cdot \frac{C_I^{n+1} - C_I^n}{\Delta t} \right) \left(\dot{V} + rV \right) \quad (3)$$

where $\Delta t =$ time step (h), $\dot{V} =$ airflow rate to and from the chamber (m^3/h), $C_s =$ concentration in supply air (particles/m^3), $S =$ strength of indoor sources averaged over each time step Δt (particles/h), $\overline{C}_I =$ average concentration in indoor air during the time step Δt (particles/m^3), $R =$ rate of particle removal (particles/h), $V =$ chamber volume (m^3), $r =$ particle removal rate constant (h^{-1}) and $C_I^n =$ concentration indoors at the beginning of time step n (particles/m^3).

The model is able to predict the strength of indoor sources dynamically if the other variables are known. The time step (about 1 min) was determined by the particle sampling interval. The airflow rate, concentration in supply air, concentration in indoor air and volume of the chamber were experimentally determined. Decay measurements of particle concentrations were used to assess the particle decay time constant T_{PD} by regression analysis. The ventilation time constant, T_{VENT}, was determined by a tracer gas decay measurement. Comparison of the two time constants provided the particle removal rate constant, r (particle loss rate) according to Eqn (4).

$$e^{-t/T_{PD}} = e^{-t(1/T_{VENT} + r)} \quad (4)$$

Sources

The sources of the examined emissions were pure wax candles, a vacuum cleaner, scented candles, air-freshener spray, a flat iron (with and without steam) on a cotton sheet, an electric stove, radiators, a gas stove, burning cigarettes and frying meat. However, only the results from experiments with burning cigarettes are presented in this paper.

Burning cigarettes

Three cigarettes were consecutively burned for approximately 10 min each.

RESULTS

Figure 1 illustrates the level of particle concentration generated from three cigarettes. The maximum concentration of ultra-fine particles was approximately 167 000 particles/cm^3 when an approximate steady state equilibrium was obtained.

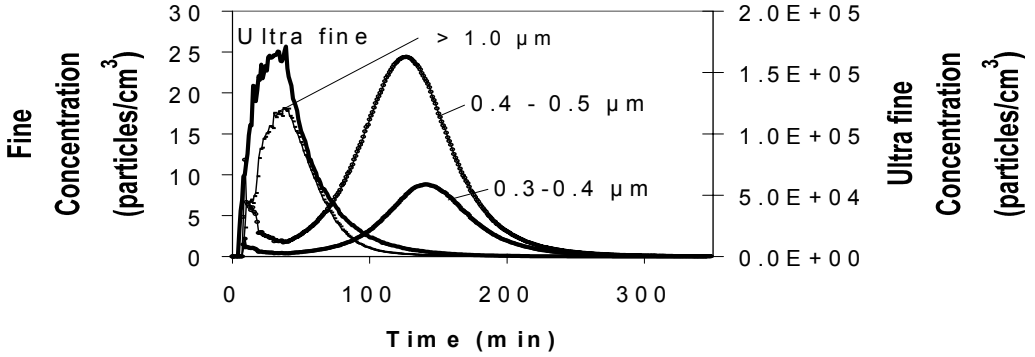

Figure 1 Concentration of particles from burning cigarettes.

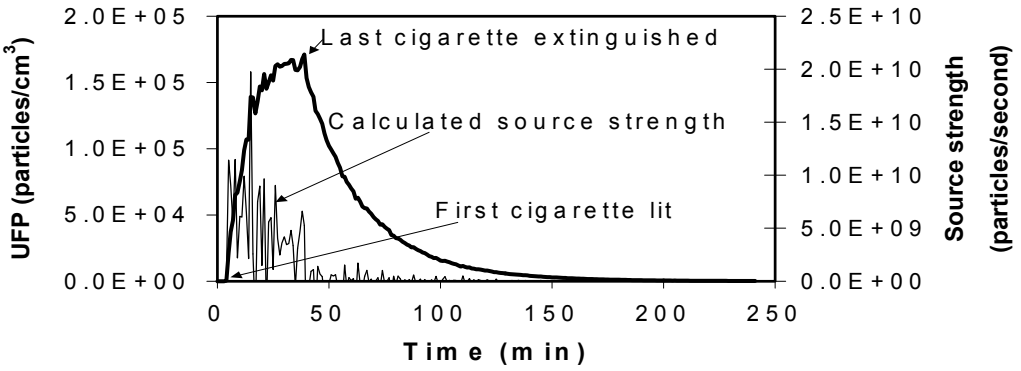

Figure 2 Calculation of source strength together with measured concentration of UFP.

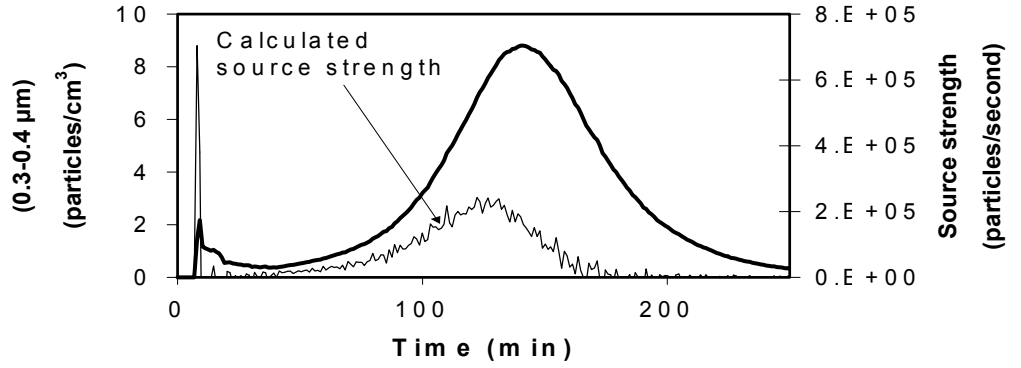

Figure 3 Calculation of source strength together with measured concentration of particles between 0.3 and 0.4 µm.

The maximum concentrations of particles in the size range of 0.3–0.4 µm, 0.4–0.5 µm, 0.5–0.6 µm and >1.0 µm were approximately 8, 24, 11 and 17 particles/cm^3, respectively. The concentrations of particles in the four size ranges between 0.6 and 1.0 µm were each approximately 3–5 particles/cm^3. The maximum concentration of particles larger than 1.0 µm was reached simultaneously with the maximum concentration of ultra-fine particles. There was a delay of 1–2 h before the maximum concentration was reached for all other fractions. The delay was longer for smaller particles. Figure 2 shows the measured ultra-fine particle concentration together with a curve representing the source strength of ultra-fine particles calculated using Eqn (3). Figure 3 shows the measured concentration of particles in the size range from 0.3–0.4 together with a curve representing the source strength calculated using Eqn (3). The measured particle concentrations together with the measured ventilation rate were used to calculate the particle loss rate for each size range. The overall dependence of loss rate on particle size can be seen in Figure 4.

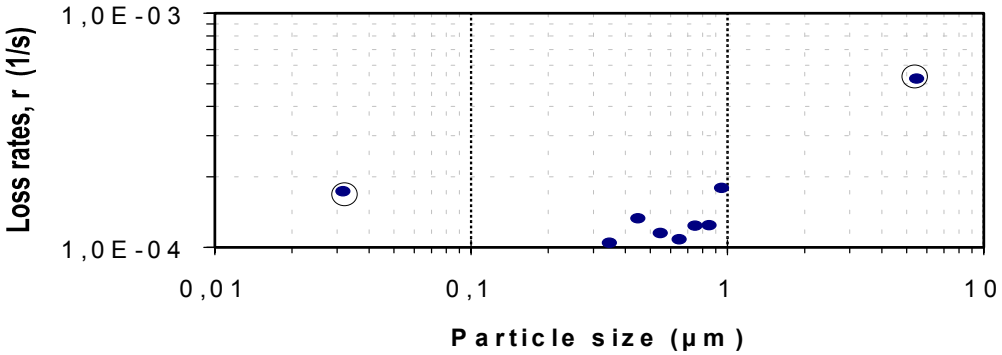

Figure 4 Particles loss rates. The smallest particles range between 0.01 and approximately 0.1 µm, while the largest particles range between 1.0 and about 30 µm. For each interval the mean particle diameter has been roughly estimated as the geometric mean.

Table 1 shows the maximum calculated source strength and the particle removal rate constants of burning cigarettes in the test chambers. The results show that the sink effect varies with the particle size.

Table 1 Particle removal rate constant, maximum concentration and calculated maximum source strength for the various particle size ranges studied

Particle size (µm)	0.01–0.1	0.3–0.4	0.4–0.5	0.5–0.6	0.6–0.7	0.7–0.8	0.8–0.9	0.9–1.0	>1.0
r (1/h)	0.63	0.38	0.48	0.41	0.39	0.45	0.45	0.65	1.89
C_{max} (p/cm^3)	1.67×10^5	8	24	11	5	5	3	3	17
S_{max} (p/s)	1.98×10^{10}	2.39×10^5	6.67×10^5	3.06×10^5	1.45×10^5	1.21×10^5	1.08×10^5	1.17×10^5	1.26×10^6

DISCUSSION

During the burning of cigarettes, ultra-fine particles dominated the number concentration in the chamber. The source strength of UFP was approximately 4–5 orders of magnitude greater than the source strengths of particles in the studied size ranges above 0.3 μm. Figure 1 shows that the concentration increase of particles in the size range 0.3–1.0 μm was delayed compared with the concentration increase for smaller particles. This indicates a particle coagulation effect. In this case ultra-fine particles may have coagulated to 0.4–0.5, 0.5–0.6 and 0.3–0.4 μm. Particles larger than 1.0 μm follow the pattern of the ultra-fine particles measured by condensation particle counters.

Sink effects had a pronounced influence on the experimental results and were considered by the model. The results in Figure 4 show that the sink varies with the particle size. The measurements for particles with diameters between 0.3 and 1.0 μm give a smaller loss rate than for the other particle sizes. Possibly this is true also for particles down to about 0.1 μm. It may depend on particle diffusivity and gravitational settling velocity. When a particle is small enough, diffusivity dominates the change of loss rate with particle size. The diffusivity increases as particle size becomes smaller, resulting in an increase in loss rate. For a large particle, however, the effect of particle settling velocity on loss rate may overwhelm the effect of diffusivity and loss rate increases as the particle size increases (Xu *et al.*, 1994). It should be noted that in this study the air velocity in the room was between 5 and 20 cm/s and thus, had an influence on the movement of particles compared with their settling velocity. Consequently, the explanation for the different time constants between the tracer gas and particles may be particle diffusivity, gravitational settling velocity, coagulation and impaction on surfaces in the room.

CONCLUSION AND IMPLICATIONS

The source strength and sink effects are two important parameters that are difficult to quantify experimentally without using an appropriate model. Modelling of the source strength and the loss rate for ultra-fine and fine particles simultaneously contribute to a more complete understanding of indoor particle sources and their size distributions. The model provides a useful tool to estimate the strength of particle sources and sinks.

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