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CIGARETTE SMOKE COAGULATION MEASUREMENT AND EMISSION FACTOR DETERMINATION

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ABSTRACT

Fine dust or PM10 is now in the public discussion as many European cities are recording an exceeding of the limit values. The main causes are combustion generated nanoparticles and resuspended dust. Concerning combustion generated particles cigarette smoke is often neglected as particle source for fine dust. This work presents typical measurements of cigarette smoke with regard to particle number concentrations over time, the apparent Smoluchowski coagulation coefficient and a resulting particle emission value for cigarette smoke for intercomparison or emission dispersion calculation purposes.

INTRODUCTION

A main topic for air pollution and health risk are combustion generated nanoparticles. A typical measured quantity is the PM10 value also called fine dust in the public discussion. It is a mass related quantity, measuring particles which are smaller then 10 micrometer on a mass related base. In the scientific community there is a great uncertainty about whether the mass is relevant concerning health issues or better other quantities like the particle number, the particle surface or chemical particle properties. Combustion generated particles – nanoparticles - are even smaller then 1 micrometer and there can be found a significant peak typically about a particle diameter of 80-100 nm (Sturm [1]), also called accumulation mode. It is also not widely known that cigarette smoke is producing fine dust, although it is recognized that it is unhealthy. It is the fine dust properties, that lead to increased influence of cigarette smoke on health often neglected or disregarded in public discussion, like long lasting air pollution in closed rooms with high particle concentrations.

It is therefore of importance to investigate the fine dust properties to make an comparison of the different fine dust sources, with respect to their influence on particle aerosol concentration. Another possibility of application is the fine dust dispersion calculation, needing as input defined particle emission sources as boundary conditions.

For these applications the kinetics of particle coagulation or growth has to be looked at. A basis description of coagulation is done with the Smoluchowski [2] equation, with the fundamental quantity of the coagulation coefficient (K), describing the development of aerosols concentration over time, when no or minor dilution occurs. With regard to the Smoluchowski equation K is derived for a monodisperse aerosol, for a polydisperse aerosol the effective K is roughly 10% higher according to Fuchs [3].

EXPERIMENTAL

Only in the 90 ties the DMA (differential mobility analyzer) technology, also known as SMPS (scanning mobility particulate seizer) was used commercial. This made possible to measure a new quantity the size distribution measurement of nanoparticles, typically in the size range between 10 and 1000 nm.

Different methods can be applied for measuring K e.g. a tube reactor or a batch reactor (Heiden [4]). The flow sheet of the experiments, performed with students in an experimental lecture, is shown in Fig. 1. A small batch reactor, consisting of a 10 liter sampling bag, was used. The cigarette smoke of one cigarette was inhaled and exhaled immediately. Inhaling into the lungs was avoided for measuring fresh smoke. During exhalation the bag was filled by natural breathing, while in the meantime the bag was closed. This way only the concentrated exhaled cigarette aerosol was used as sample. The SMPS system 3081 from TSI consisting of the classifier 3080 and the CPC 3010 (condensation particle counter) was used for getting the particle concentration over time.

The results for the measured size distributions over time are shown in Fig. 2, where measurements were done every 2 minutes in sequence. Dp denotes the particle diameter and N the total particle concentration per volume. For the measured size distributions the lognormal distributions were calculated also shown in Fig. 2. The cigarette aerosol showed a very sharp decrease in particle concentration, with a very high particle wall diffusion frequency.

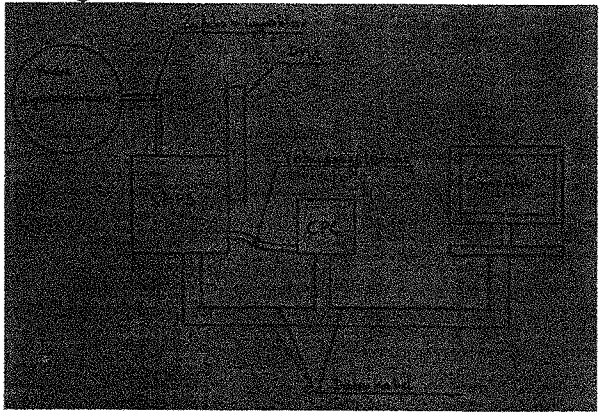

Fig. 1 Experimental setup for the particle number size distribution and the coagulation measurement.

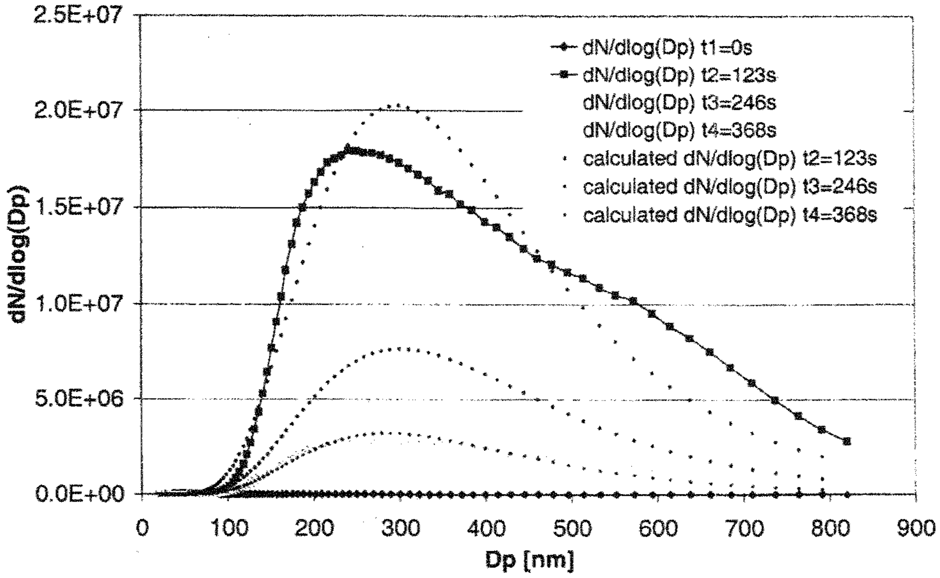

Fig. 2 Particle number size distributions over time of exhaled cigarette smoke resampled from a 10 liter sampling bag.

The calculated total particle concentrations were fitted with a theoretic equation, considering aerosol coagulation and deposition, according to Heiden [5] and Fig. 3. The resulting fit had a R^2 of 0.995 with a coagulation coefficient K of $7.2*10^{-8}$ [cm^3/s], were the wall deposition frequency is $1.7*10^{-3}$ [1/s], indicating that due to the small volume the proportion of wall deposition caused by diffusion is high compared to coagulation. A main influence on this is due to the fact, that the total volume filled with one cigarette exhaust yields a very small volume, and that the bag was very flat when filled, deviating largely from spherical shape.

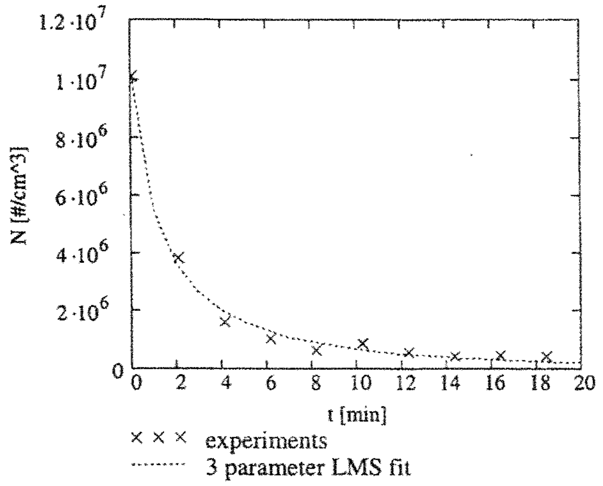

Fig. 3 Total particle concentration N over time for the cigarette experiment over time t and the corresponding 3 parameter least mean square fit according to Heiden [5], for the initial concentration N_0, the coagulation coefficient K and the particle wall diffusion frequency L.

Finally the emission factor for a cigarette with respect to the particle number concentration was determined. A cigarette lost 1.875 [mg/s] mass per second and was smoked in 4 minutes. From the dilution of the cigarette in a closed room the net particle emission rate alpha, from cigarette smoking, was calculated from the measured particle concentrations with alpha=$7.2*10^9$ [#/s] particles per second, leading to $1.72*10^{12}$ [#/cigarette] particles per cigarette. Eq. 1 and data from Tab. 1 were applied for a mass balance getting the reaction rate r [#/(s*cm^3)] from initial clean air particle concentration N_0 to the polluted particle concentration N_t.

$$\text{alpha} = \frac{N_t - N_0}{t} \cdot V = r \cdot V = Q \cdot \left(N_t - N_0\right) \quad \text{Eq. 1}$$

Eq. 1 can also be interpreted as the equation for a CSTR (continuously stirred tank reactor), e.g. according to Moser [6], where Q is then the feeding fresh air stream.

This is a first emission factor for cigarette smoke, which has to be consolidated with more cigarette measurements.

Tab. 1 Measurement parameters for the emission factor calculations. See text for description of the experiments.

symbol	value	unit	description
N_0	5.51E+03	#/cm^3	initial particle concentration
N_t	9.17E+04	#/cm^3	particle concentration in the end
V	20	m^3	volume of the room
t	4	min	time cigarette smoked
Q	0.08991	m^3/s	dilution of the room with fresh air

CONCLUSION

Cigarette smoke measurements have been performed leading to typical measurement values of the particle number size distribution, the particle number concentration and the coagulation coefficient. In addition emission factors from a simultaneous mass reduction measurement of the smoked cigarette have been determined. Future measurements can include following features like measurement in closed rooms, with different types of cigarettes and cigars, as measurements on typical places which are subject to cigarette smoke influence and hence increased health risk.

ACKNOWLEDGEMENTS

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