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VERIFICATION OF A SET-UP FOR MEASUREMENTS OF ENVIRONMENTAL FINE AND ULTRAFINE PARTICLE RESPIRATORY TRACT DEPOSITION IN HUMANS.

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INTRODUCTION

Epidemiology tells us that elevated outdoor particle concentration, e.g. PM_{2.5} or number of ultrafine particles is associated with increased prevalence of respiratory and cardio-vascular disease in susceptible sub-population (e.g. asthmatics and children). Detailed knowledge of the deposited fraction of inhaled aerosol is needed when determining the dose to the respiratory tract e.g. for different environmental aerosols, when comparing susceptible sub-groups or during controlled exposure studies. Respiratory deposition varies strongly with particle diameter in PM_{2.5}. For hydrophobic particles in the size range 100 – 1000 nm, as little as 10-20% of the inhaled particles deposits in the respiratory tract (ICRP 66) while 80-90% is exhaled. In this region the majority of the mass in rural and urban PM_{2.5} is found. However for 20 nm particles the deposited fraction is as high as 70-80 % with the major part depositing in the lower respiratory tract. The equilibrium RH in the alveolar region is ~99.5% (Anselm et al. 1990) which causes the majority of environmental particles to grow to different extent, thereby changing the deposition probability. Ammonium sulphate (an important constituent of the ambient aerosol) has a diametric growth factor (G_F) of 3-5 at RH=99.5%. Solid ultrafine particles may consist of *agglomerates* that collapses and shrinks when exposed to a high relative humidity, however the presence of liquid/semi-volatile components may inhibit this behaviour (Weingartner et al. 1997). Further the *semi-volatile components* may evaporate in the human lung or in the equipment.

A set-up for fast (~5-10 min) measurements of size fractionated respiratory tract deposition of environmental fine particles has been constructed. The aim of this work is to test the method using hygroscopic and non hygroscopic aerosols and investigate the method for some potential artefacts when used for various environmental particle sources.

METHODS

The system (fig. 1) utilises near-real-time aerosol spectrometers, alternatively sampling in inhaled and exhaled air samples. The basis of the method is the conservation of the particle diameter in dehydrated samples of inhaled and exhaled air. Environmental aerosol is inhaled, a three-way valve is used to separate inhaled and exhaled air. 10 to 20 breaths of exhaled aerosol is collected in a rebreathing bag. The breath pattern is monitored using pneumotachographs. Both the inhaled and the exhaled aerosols are *dried* to a well-defined state (RH<15%), using a Nafion drier (Perma Pure inc.). A reservoir with residence time~10 s is used to ensure recrystallisation before the samples enter a SMPS 3934 (15 - 800 nm) and an APS 3310 (0.5 –2.5 µm). The inhaled sample is analysed during the breathing manouvre, followed by the exhaled sample. From the two measurements the deposited fraction as a function of diameter is deduced. The system has been initially tested using stable di-(2-ethylhexyl) sebacate (DEHS) and hygroscopic NaCl polydisperse aerosols generated to a 18 m³ stainless steel chamber from which the aerosols were inhaled. The SMPS-system is used to quantify the losses in various parts of the system. The conservation of the dehydrated particle diameter is studied using a Tandem-DMA set-up.

RESULTS AND DISCUSSION

In figure 2 the effect of hygroscopic particle growth on respiratory tract deposition is clearly evident, suggesting a growth factor of 5. The deposited fractions presented here for a single test-subject agrees within ± 0.1 with previous experimental data based on monodisperse particles (e.g. Tu and Knutson 1984).

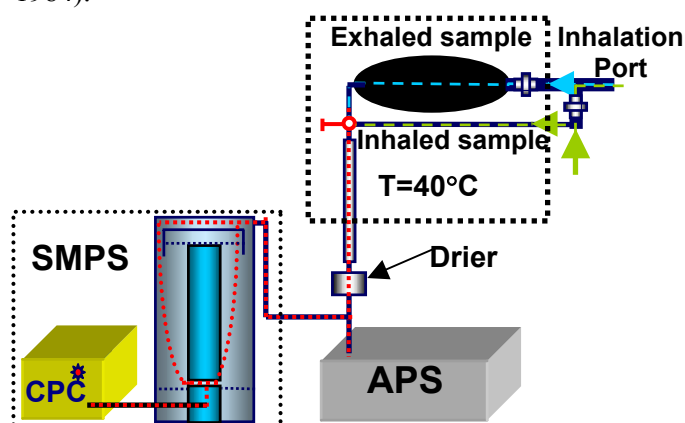


Figure 1, Schematic view of the deposition set-up.

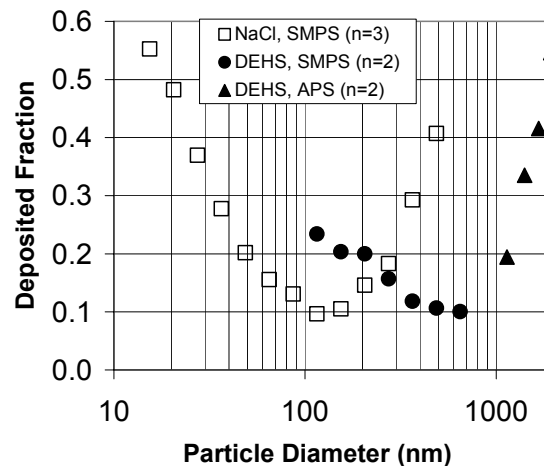


Figure 2. NaCl and DEHS, 1 test subject, male 28 years, $fp=15 \text{ min}^{-1}$, $V_t=700 \text{ cm}^3$, breathing through the nose.

The measured losses in the drier are between 1 and 10% for sub-micrometer particles and agree with diffusion theory. However the effect is minimized as both the inhaled and the exhaled sample is taken through the same drier. The breathing valves opens up with minimal obstruction, therefore the impaction losses are believed to be small. The diffusion losses in the exhalation-reservoir is minimised as the SMPS starts its scan with the smallest particles. The diffusion losses were $< 1\%$ for $d_p > 60 \text{ nm}$, however a correction is need for the smallest particles. At high concentrations coagulation might distort the measured deposition of the smallest particles. In a “bad case” of two modes at 15 and 150 nm respectively, the artefact in the deposited fraction is around ± 0.05 for the smallest particles for a concentration of 100.000 cm^{-3} . If the detection limit is defined as 0.05 uncertainty in deposited fraction for a given size, then with the present operating parameters a concentration ($dN/d\log[dp]$) of $\sim 3.000 \text{ cm}^{-3}$ is needed at 150 nm and $\sim 15.000 \text{ cm}^{-3}$ is needed in the outer SMPS channels (15 and 650 nm). In the APS-region a concentration $\sim 5 \text{ cm}^{-3}$ is needed in each size channel.

CONCLUSIONS

Results for one hygroscopic and one hydrophobic aerosol show that the method produces fast ($\sim 5\text{-}10 \text{ min}$) respiratory deposition data in the size range $15 \text{ nm} - 2.5 \mu\text{m}$. By concurrent measurements of the particle hygroscopic properties using a H-TDMA at a high RH (e.g. $\text{RH}=95\%$) and knowledge of the breathing pattern and the dimensions of the respiratory tract, the respiratory deposition models used today may be verified and if needed, optimised.

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