LUND UNIVERSITY

A Thermodesorber for Online studies of Combustion Aerosols - Influence of particle diameter, residence time and mass concentration

Dahl, Andreas; Pagels, Joakim

2003

Link to publication

Citation for published version (APA):
Dahl, A., & Pagels, J. (2003). *A Thermodesorber for Online studies of Combustion Aerosols - Influence of particle diameter, residence time and mass concentration*. Abstract from 1st International Symposium on Incomplete Combustion, University of Kuopio, Finland.

Total number of authors:
2

General rights
Unless other specific re-use rights are stated the following general rights apply:
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

Read more about Creative commons licenses: https://creativecommons.org/licenses/

Take down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

LUND UNIVERSITY

PO Box 117
221 00 Lund
+46 46-222 00 00

A THERMODESORBER FOR ONLINE STUDIES OF COMBUSTION AEROSOL PARTICLES - —INFLUENCE OF PARTICLE DIAMETER, RESIDENCE TIME AND MASS CONCENTRATION

ANDREAS DAHL and JOAKIM PAGELS

Division of Aerosol Technology (EAT), Lund Institute of Technology
Sölvegatan 26, SE-221 00 Lund, Sweden

Keywords: Thermodesorber, Volatility TDMA

INTRODUCTION

Online physical and chemical characteristics of ultrafine particles are much needed in studies related e.g. to human health and new particle formation in the atmosphere. The conditioning of aerosol particles at suitable temperatures in a thermodesorber (TD), thereby removing volatile substances is a promising tool for such studies. TD:s are usually operated in connection with a particle size spectrometer (e.g. a SMPS or an ELPI). When operated at high concentrations a desorber part filled with e.g. active carbon is sometimes connected after the heater (e.g. Burtscher *et al.*, 2001 and Wehner *et al.*, 2002). By selecting monodisperse particles with a DMA before the sample pass the TD, high-resolution spectroscopy of volatile components can be performed in a Tandem Differential Mobility Analyzer (TDMA-setup). For spherical particles, volatile volume fractions and vapour pressures can then be estimated. By combining measurements of the particle mass and mobility of the conditioned and unconditioned particles, quantitative volatile volume fractions can be determined also for fractal-like agglomerates (Park, 2003). Sakurai et al. (2003) used a TD operated in a TDMA-setup to get information about the vapour pressures of “more volatile” diesel nano-particles. Half the particle volume was evaporated at around 343 K for 30 nm particles and at around 368 K for 70 nm particles. It is desirable to compare data collected in TD:s with different residence time, for different particle diameters and mass concentrations. Therefore experiments to study the influence of these parameters are needed.

In this paper we describe the design and characterisation experiments of the recently built LUND-TD for temperatures up to 873 K. We have used this setup to study influence of particle mass concentration, particle diameter and residence time in the TD on the evaporation profile for relevant laboratory generated aerosols.

THEORY

In the free molecular regime the change in particle diameter due to evaporation can be written as:

$$\frac{dD_p}{dt} = \frac{c_i \cdot M_i \cdot K}{4 \cdot \rho_i \cdot R} \cdot \left(\frac{p_{i,\infty}}{T_\infty} - \frac{p_{s,i}}{T_p} \right) \qquad \text{Eq.1}$$

Where c_i denotes the mean molecular velocity of species i, M_i is the molar mass, K is the Kelvin factor, ρ_i is the particle material density, $p_{s,i}$ is the saturation vapour pressure of species i just above the surface and $p_{\infty,i}$ is the partial pressure of species i, far away from the particle. T_p and T_∞ are the temperature at the particle surface and far away from the particle respectively. Note that in this model, the diameter change (nm/s) is independent of particle diameter (neglecting the Kelvin effect). At low mass concentrations $p_{\infty,i}$ can be set to zero and the evaporation rate becomes independent of mass-concentration. In the continuum regime dD_p/dt is proportional to D_p^{-1}. For particle diameters smaller than 80 nm, the error of eq. 1, compared to the general formula (Flagan and Seinfeld, 1988) is less than 30% for T=293 K and less than 10% for T=773 K.

METHODS

The design of the LUND-TD is optimised for flow rates between 0.3 and 1.0 l/min, flows which are preferred for the Scanning Mobility Particle Sizers (SMPS, TSI, Inc., USA). The heater consists of an 18 mm in inner diameter, 550 mm long insulated stainless steel tube. Thermocouples are brazed on the outside of the heater, one at the middle and one 50 mm from the exit. The regulator is connected to the middle thermocouple, while the second can be used to check the exit temperature. A 500 mm long adsorber tube with activated carbon is used as adsorbent. The heater is capable of operating up to 1073 K. In order not to incinerate the carbon, a 500 mm long tube with an inner diameter of 8 mm replaces the adsorber when the TD is used at higher temperatures (see figure 1).

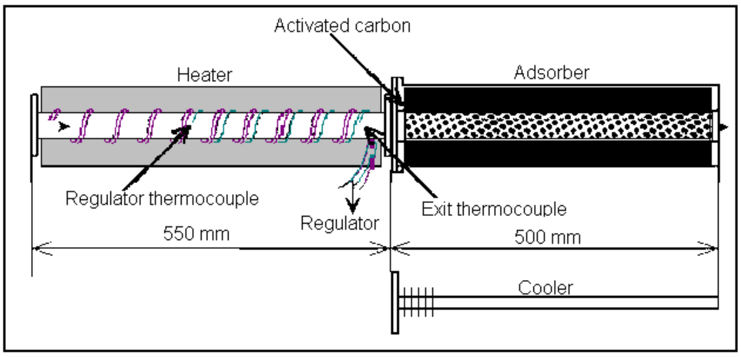

Figure 1. Schematic overview of the LUND-TD and the location of the thermocouples.

The residence time for the aerosol at the set TD temperature, T_s, was calculated as time were the aerosol was at a temperature higher than (T_s -25 K). The temperature profiles are shown in figure 2 and the calculated residence times are displayed in table 1.

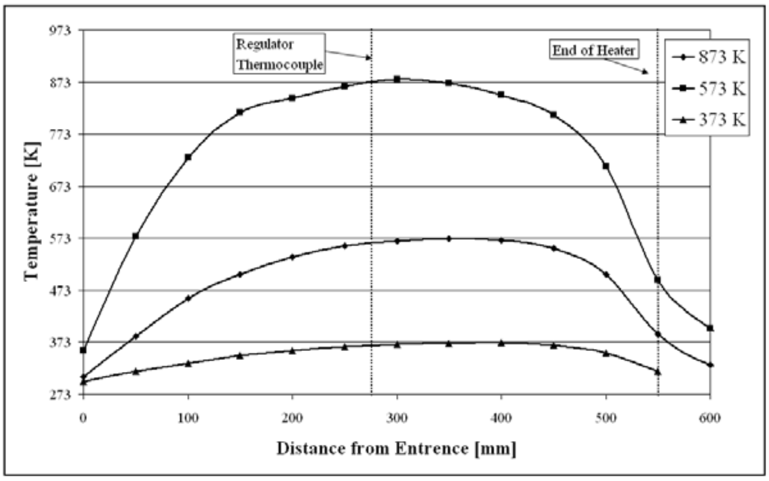

Figure 2. Temperature profiles at a flow rate of 1.0 l/min at 373, 573 and 873 K.

Table 1. Estimated residence times (Δt) for the aerosol at different T_s and flow rates

T_s [K]	Flow rate [l/min]	
	0.3	1.0
373	**15.2 s**	**4.4 s**
573	**6.8 s**	**1.9 s**
873		**1.0 s**

The penetration was measured for a monodisperse aerosols with particle diameters 20, 30, 50 and 100 nm at the flow rates 0.3, 0.6 and 1.0 l/min and at TD temperatures from room temperature up to 773 K. A nebulizer model 3076 (TSI. Inc., USA) was used to generate a K_2SO_4-aerosol (chosen because of its low vapour pressure). An electrostatic classifier (EC) model 3071 (TSI. Inc., USA) was used to select the particle sizes. A Scanning Mobility Particle Sizer (SMPS)-system, consisting of an EC model 3071 and a Condensation Particle Counter (CPC) model 3010 (TSI. Inc., USA), was used to count the particles leaving the LUND-TD.

The evaporation performance regarding ability to evaporate and remove a volatile substance from the aerosol was investigated with different aerosols at different mass concentration. The tests of the thermodesorber were made with a monodisperse KCl-aerosol and a condensation aerosol consisting of di-octyl sebacate (DOS) particles with NaCl nuclei.

The setup for the KCl evaporation was the same as for the penetration measurements. For the different TD temperatures the particle size distribution was measured. Due to the rapid particle diameter decrease in a narrow temperature range above 673 K, the TD temperature was increased with 15 to 25 K steps in that region. The DOS/NaCl-aerosol was prepared with a condensation aerosol generator (Model: SLG 270 Topas, GmbH, Germany). The generator was setup to produce an aerosol with a mean number diameter of 500 nm and a σ_g of <1.2.

RESULTS AND DISCUSSION

At room temperature and a flow rate of 0.3 l/min, the penetration of particles with a diameter of 100 nm is 90% and at higher flow rate of 1.0 l/min 96%. At 573 K the penetrations descents to 69% and 86% respectively. The main deposition processes in the TD are thermophoresis and diffusion and most of the deposition takes place in the section of the TD where they work in the same direction, i.e. where the aerosol temperature decreases. The particle penetration through the TD is less dependent of particle size (in the range 20-100 nm) and more related to the flow rate and TD-temperature.

The results from the evaporation of the KCl-aerosols are displayed in figure 3 as diameter change (ΔD_p) in nm at the set temperature. The results are from single measurements. The diameter change is independent of the initial particle diameter, as is expected from theory. From figure 3 it is also obvious that there is no influence of the mass concentration on the evaporation profile at the low mass concentrations studied. In figure 3 differences between the evaporation profiles at flow rates of 0.3 and 1.0 l/min are also displayed. Strong residence time dependence is indicated, then the particle diameter decrease seen at 0.3 l/min takes place approximately 30 K higher with a flow rate of 1.0 l/min. Comparing measurements of the two flow rates at the same temperature gives a ratio in ΔD_p of around 3-4, which according to eq. 1 (neglecting the Kelvin effect) gives a ratio of the residence times (Δt) of 3-4. This is very close to the experimentally found difference in residence time measured at 573K. A high flow rate will yield a higher particle penetration but because of the temperature-residence time relationship a higher temperature is needed to evaporate a given compound.

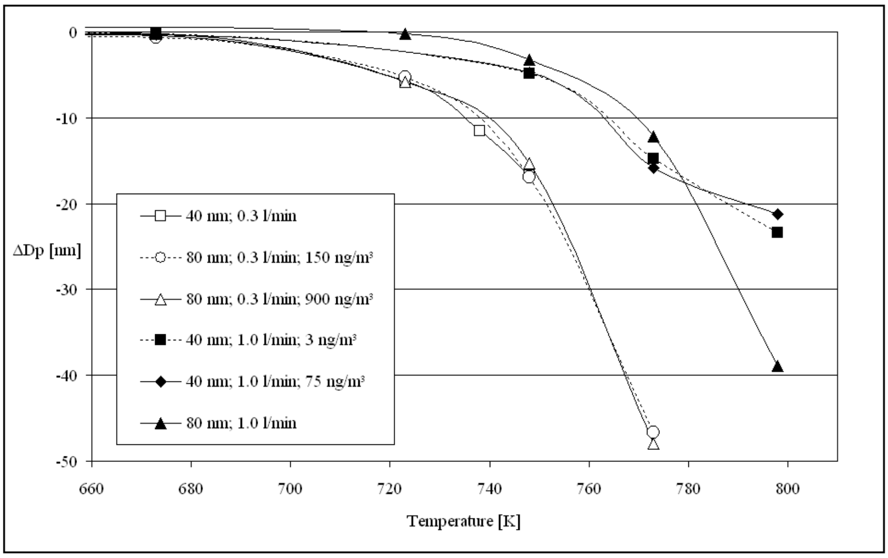

Figure 3. Particle diameter change for monodisperse KCl-aerosols with initial mean diameters of 40 and 80 nm at flow rates of 0.3 and 1.0 l/min. The measurements on 80 nm particles at the flow rate of 0.3 l/min as well as the measurements on 40 nm particles at 1.0 l/min where performed at two different initial particle mass concentrations The measurements were related to diameters at 473 K.

Figure 4 displays particle size distributions of DOS/NaCl-aerosols with initial particle mass concentrations of 70 and 400 µg/m³. At ideal evaporation only the NaCl-nucleus should remain. The aerosol with high mass concentration yields slightly larger particles exiting the thermodesorber. This is either due to insufficient evaporation in the heater, limited by the vapour pressure of DOS, or readsorbtion of DOS on the particles when the aerosol cools and enters the adsorber. The effect is not noticeable at even higher temperatures.

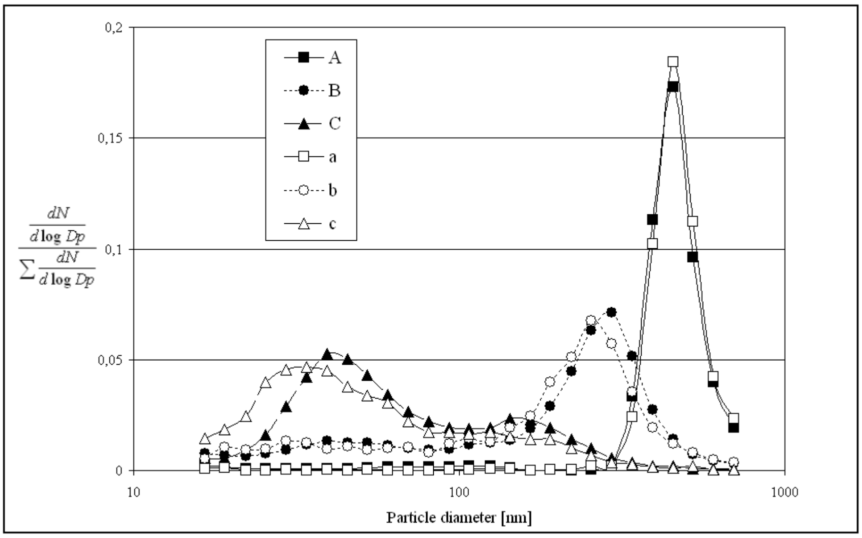

Figure 4. Particle size distribution from evaporation of DOS/NaCl-aerosol with the LUND-TD at a flow rate of 0.3 l/min. The samples A, B and C where measured on an aerosol with an initial mass concentration of 400 µg/m³ and the samples a, b and c had the initial mass concentration 70 µg/m³. The A samples where led bypass the TD and the B where measured at TD a temperatures of 353 and C at 358 K.

CONCLUSIONS

A thermodesorber suitable for online measurements of combustion aerosols has been designed and tested using relevant laboratory generated aerosols. The temperature range has been up to 873 K, but may be extended to 1073 K in future application. The particle penetration through the thermodesorber and the temperature profile are comparable to existing equipment. Evaporation experiments have been performed from 3 ng/m^3 in a TDMA setup with monodisperse particles, to 400 µg/m^3 in a single DMA-setup

The influence of residence-time, particle diameter and mass-concentration on the evaporation profile of the TD was found to be comparable to the theory in the free molecular regime. Taking the residence time in the TD into account allows for a better comparison between measurements taken with different TD designs and flow rates. More measurements are needed at high mass concentrations to investigate the performance of the adsorber and potential recondensation of volatilised material on particles.

REFERENCES

Burtscher H., Baltensperger U., Buckowiecki N., Cohn P., Hüglin C., Mohr M., Matter U., Nyeki S., Schmatloch V.,Streit N., Weingartner E. (2001). Separation of volatile and non-volatile aerosol fractions by thermodesorption: Instrumental development and applications, Journal of Aerosol Science, 32, 427–442.

Sakurai H., Tobias HJ., Parka K., Zarling D., Docherty KS., Kittelson DB., McMurry PH., Ziemann PJ., (2003) On-line measurements of diesel nanoparticle composition and volatility, Atmospheric Environment 37, 1199–1210

Wehner B., Philippin S., Wiedensohler A., (2002), Design and calibration of a thermodenuder with an improved heating unit to measure the size-dependent volatile fraction of aerosol particles, Journal of Aerosol Science, 33, 1087-1093

Park K. (2003) In-Situ Measurements of 50-500 nm particles: Mass-Mobility Relationship. *PhD-Thesis,* University of Minnesota, USA

Flagan RC., and Seinfeld JH., (1988), Fundamentals of air pollution engineering, Prentice Hall, Englewood Cliff, NJ, USA