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Urban Aerosol Studies of PM_1 Size Fraction with Reference to Ambient Conditions and Visibility

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ABSTRACT

Mass measurement of ambient PM_{10} and $PM_{2.5}$ size fractions is a rather well established technique. However there is a broad consensus in the scientific community about the importance of smaller particle size fractions regarding epidemiological and environmental aspects.

We present aerosol mass measurement performed with two different aerosol impactors operated simultaneously – a commercial real cascade impactor and a custom made cascade virtual impaction plate impactor. Both instruments have size segregation for 10, 2.5 and 1 µm in terms of an aerodynamic diameter, hence corresponding to PM_{10}, $PM_{2.5}$ and PM_1. Furthermore the Vienna Telephotometer delivers information about the extinction coefficient in the vicinity of local particle sampling.

The time-resolved size distributions of the PM_1 fraction are measured by means of the scanning differential mobility analyzer and are merged with the time-resolved aerosol extinction coefficient data in the area of interest, along with the particle surface and mass concentration measurement. Results show the importance of time resolved measurement of the PM_1 size fraction and indicate a good correlation between the ratio of PM_1 to $PM_{2.5}$ and PM_{10} with the extinction coefficient.

Keywords: PM_1; Urban aerosol; Size distribution; Aerosol extinction coefficient; Nanoparticles.

INTRODUCTION

The impact of airborne particulate matter (PM) on health and environment is strongly associated with aerosol size distributions and concentrations next to the chemical composition of particles in question. There is a large number of studies which establish links between concentration of ambient aerosols, level of air pollution and adverse health and environmental effects (e.g. Eldering and Cass, 1996; Pope III and Dockery, 2006). For the description of air quality the term particulate matter (PM) has been coined. PM_{10} particle size fraction represents the mass concentration of particles with aerodynamic diameters below 10 micrometers. Similarly, the $PM_{2.5}$ rule was established in the United States, however not yet formally in the European Union. It is a broad understanding nowadays that the PM_{10} and $PM_{2.5}$ measurement provide very important steps towards air quality assessment; however there is equally no doubt that more accurate descriptors of the actual environmental burden are still needed. There is a consensus that likely PM_1 would be a more suitable size than $PM_{2.5}$ for health related aerosol sampling (Oberdörster *et al*., 1994). However, there still is a relatively limited amount of data for the sub-micrometer ambient particle fraction available. Recently an increasing volume of scientific contributions started mirroring the enormous importance and very likeliness of health hazards one now associates with nanoparticles (Kreyling *et al*., 2006). The increasing role of ultrafine particles (UFP) (< 100 nm) is discussed in the scientific community concerning environmental and health aspects. However the $PM_{0.1}$ fraction alone seems not to be sufficient for the description of particles with regard to the deposition in the relevant lung area. There is a non-negligible contribution of particles > 100 nm to the deposition in the lung which can be calculated for tracheobronchial and alveolar region using Eq. (1) respectively Eq. (2) (ICRP, 1994).

$$DF_{TB} = \left(\frac{0.00352}{d_p}\right)\left[\exp(-0.234(\ln d_p + 3.4)^2) + 63.9\exp(-0.819\ (\ln d_p - 1.61)^2)\right] \quad (1)$$

$$DF_{AL} = \left(\frac{0.00155}{d_p}\right)\left[\exp(-0.416(\ln d_p + 2.84)^2) + 19.1\exp(-0.482\ (\ln d_p - 1.362)^2)\right] \quad (2)$$

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The search for other parameters than PM mass to characterize the environmental and health burden of submicron particles continues. Concerning health issues there already is a broad consensus in aerosol science that the total surface area of particles that might enter some regions of the human lung could be a suitable parameter for description (Wittmack, 2007). It has been found that the surface area of particles entering the lung corresponds very well to the incidence of inflammation caused by the particles (Oberdörster, 2000; Oberdörster *et al*., 2005).

To establish a reliable data base for environmental aerosol sampling the placement of the sampling inlet must be considered. Measuring the health related aerosol at a height of 25 m above street level may appear not to be significant. But it has been shown (Morawska *et al*., 1999) that the fine particle number concentration (D_p ~20–600 nm) does not vary significantly with either different heights or distances (60–200 m) from busy roads, as long as the measurement site is not located in the immediate proximity of a busy road. Also other studies (e.g. Buzorius *et al*., 1999) done on this topic did not show a high correlation between height and particle number concentration.

The recently assembled Vienna Aerosol Monitoring System (VAMS) allows us to compare different aerosol parameters of the PM_1 fraction and also delivers the environmental PM_{10} and $PM_{2.5}$ data showing a relationship between those fractions. For measuring of the detailed number size distribution below 1 μm in terms of an equivalent diameter we use a differential mobility analyzer in scan mode (SDMA) yielding time resolved data. Simultaneously the surface area of particles is monitored with the Nanoparticle Surface Area Monitor (NSAM, Mod. 3550, TSI, Inc.).

Furthermore we also use optical parameters for urban aerosol characterization obtained with an aerosol telephotometer. This device delivers information regarding the extinction properties of atmospheric aerosols in the vicinity of the sampling site. From the measured visibility (V) of the atmosphere, which can even be estimated with the naked eye (WMO, 1996), the extinction coefficient (σ_{ext}) can be determined using the well known relationship $V = 3.912/\sigma_{ext}$ (Middleton, 1952).

Comparing the acquired data in this work allows us to draw conclusions between the assessed PM burden, the easily measurable visibility (V) as well as the environmental conditions like weather or traffic situation in Vienna.

EXPERIMENTAL ARRANGEMENT AND METHODS

The urban aerosol studies are carried out in the city center of Vienna. Instruments for the current set of measurements are located on the roof of the building of the Faculty of Physics at a height of approx. 25m above street level. The location of the building is approx. 70m from a road with regular major city traffic but hardly any trucks (Fig. 1).

The roof of the faculty where the setup is assembled is the highest point in the surrounding area within a radius of about 600 m. The sampling inlets are about 1 m above roof level. The setup is shown in Fig. 2.

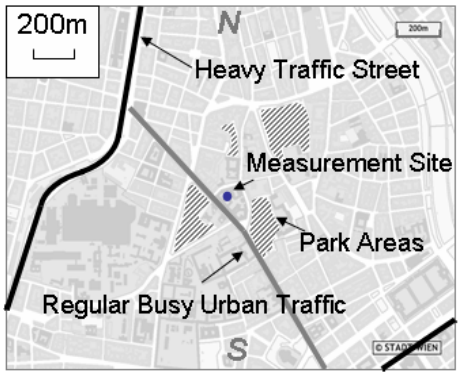

Fig. 1. Location of the sampling site in Vienna city center with indicated orientation.

For measuring the mass concentration we use two different impactors having the same size segregation. The CAscade Virtual Impactor (CAVI) (Prasserttachto *et al*., 2006) is a non-commercial in-house developed system with an omni-directional inlet with an upper cut-off size of the order of 100 μm and size separation into $PM_{10–2.5}$, $PM_{2.5–1}$ and PM_1. The total flow rate through the impactor is 12.5 liters per minute (L/min) with minor flows set at 10% of the total flow rate in the respective section. Size separated particles are collected on isopore membrane filters (0.02 μm pore size). Concurrently a commercial real cascade impactor system (ISAP 1050) with a flow rate of 38 L/min and the same size separation as the CAVI was used. The PM_1 fraction from the CAVI is taken out of the unit and sampled on an external filter. This allows the mass analysis of the sample simultaneously with operation of the Scanning Differential Mobility Analyzer (SDMA) (Reischl, 1991) combined with a CPC and the NanoSurface Aerosol Monitor (NSAM) (Fissan *et al*., 2007). The working range of SDMA spans from 10–1000 nm in terms of equivalent particle diameter.

Simultaneously the deposited particle surface area in the alveolar and tracheobronchial region of the lung is detected with the NSAM. It should be noted here that the NSAM does not measure the total particle surface area. Sampled particles are corona charged and consequently detected by means of an electrometer. By adjusting the trap voltage which eliminates ions or sub 10-nm particles and by determining the calibration coefficient the calibration of the instrument for the desired lung region can be carried out. Its response matches the lung deposition criteria as predicted by deposition models defined by the American Conference of Governmental Industrial Hygienists (Vincent, 1999).

In this work the measurement of the deposition data in two different lung regions will be discussed: the alveolar region and the tracheobronchial region. For the alveolar region the trap voltage was found to be 220 V, the calibration coefficient was determined as 0.32 (μm²/cm³)/fA and for the tracheobronchial region 100 V and 0.072 (μm²/cm³)/fA respectively. To verify the calibration, the instrument response function was compared with the calculated surface area as described in the following section.

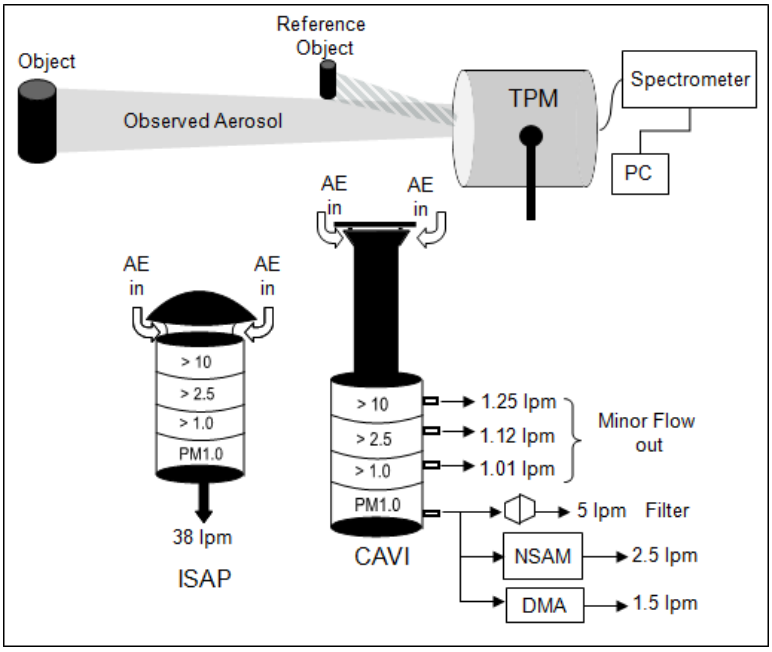

Fig. 2. Schematic diagram of the Vienna Aerosol Monitoring System showing the details of the set-up.

The significance of the NSAM data for particles with diameters above about 400 nm for most practically important situations decreases as several studies have shown, since deposition becomes density dependent above this size range due to the change of the main deposition mechanism away from diffusion towards impaction (Heyder *et al*., 1986; Fissan *et al*., 2007). Recent studies (Asbach *et al*., 2009) suggest the upper size limit for this instrument to be around 400nm as well. Since we use the CAVI as a pre-separator for particles above 1 µm diameter before they enter the NSAM and since the SDMA data do not show a significant amount of particles above 400nm in the monitored aerosol (e.g. Fig. 7) we claim that this limitation influences our results in a negligible way. Assuming that the particles in question are mostly spherical (McMurry *et al*., 1996) the number concentration obtained by the SDMA can be converted into the surface area and subsequently into the deposited surface area in the different lung regions. Based on this it can be shown, that the determined deposited surface area calculated with the measured number concentration of the SDMA, and the NSAM measured surface area show comparable results within an deviation of ~20% (e.g. 65 µm²/cm³ SDMA–58 µm²/cm³ NSAM for TB, 2.9 µm²/cm³ SDMA–3.8 µm²/cm³ NSAM for AL respectively) which could be explained with the imperfect sphericity or varying dielectric constants of ambient particles which may affect the functioning of the NSAM. Consequently, we find that the NSAM data contribute an important input to the overall assessment of the atmospheric aerosol.

The Vienna TelePhotoMeter (TPM) (Fig. 3) consists of a Schmidt-Cassegrain telescope and a spectrometer for wavelength-resolved intensity measurements. A stepper motor controlled by a PC operates the horizontal and the vertical alignment and thus allows focusing the telescope on distant objects, in our case an object in the distance *s*, the horizon and a reference object in the distance *s'* to take into account the so-called intrinsic luminosity (Horvath, 1981).

Using the Koschmieder Eq. (3) one can determine the extinction coefficient σ_{ext} and consequently the visual range V ($V = 3.912/\sigma_{ext}$). For accurate measuring of the extinction coefficient with this instrument it is necessary for the distance s to be between $1/4 - 3/4$ of the visual range as it has been shown in Horvath (1981). In case of the Vienna urban aerosol the minimum of the distance s should be approx. 2 km. The object used in this work was in a distance of 1.95 km from the measurement side and thus just within the limitation for the instrument. On the roof of the university building, in a distance of 0.05 km (s') the reference object is situated. This has to be an accurate reproduction of the object concerning illumination and reflectance.

$$\sigma_{ext} = -\frac{1}{s}\ln\frac{I_{hor}(s) - I_{obj}(s)}{I_{hor}(s) - I_{\mathrm{int}}(s')} \tag{3}$$

Ambient parameters like wind speed, wind direction, temperature and relative humidity were provided by Austria´s national weather service agency. The data were obtained hourly inside the city centre, in the proximity of the measurement site. Temperature and relative humidity were measured additionally at the sampling site during the measurements.

Calibration of the NSAM

To use the NSAM as an instrument for determining the deposited surface area in lung regions it is advisable to calibrate it. In this work it was done with Polystyrene Latex (PSL) Particles suspended in ultra pure H_20 (Merck, Inc.) and Di-Ethyl-Hexyl-Sebacat (DEHS) dissolved in

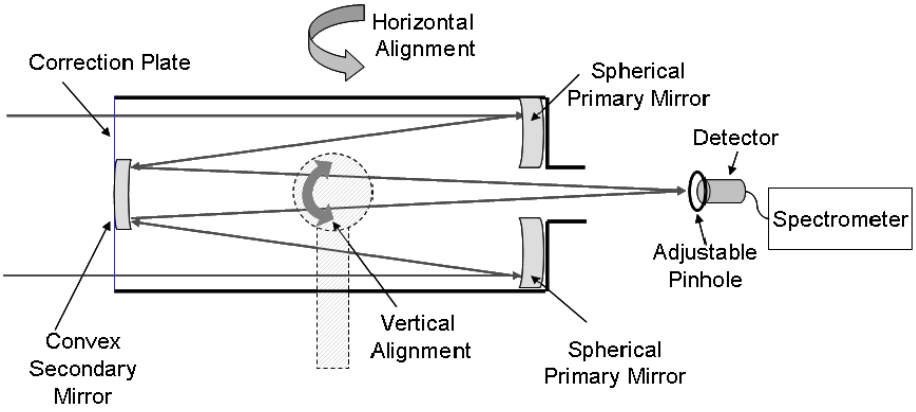

Fig. 3. Schematic diagram of the Vienna telephotometer.

Isopropanol. Both suspensions were nebulized, dried and subsequently introduced into the SDMA, used in this case as a particle size classifier. After the SDMA the aerosol was channeled simultaneously into the NSMA and the Condensation Nuclei Counter (CNC) as shown in Fig. 4.

The CNC determines the number concentration in particles per cubic meter. The NSAM measures the current resulting from the charged particles. It has been shown that this current measurement allows an assessment of the particle surface area (Shin *et al*., 2007). By adjusting the ion trap one can determine the size range of particles entering the detector. With the calibration coefficient the measured current can be converted into the surface area of the deposited particles. In Fig. 5 the deposition determined in the above described way is shown as measured deposition.

The number concentration of the SDMA size-classified aerosol is gained determined by the CNC. Because DEHS and Latex particles are spherical, the total surface area of the particles can be easily calculated. Using Eq. (1) respectively Eq. (2) one can determine the fraction of particles entering the different regions of the lung and hence the deposited particle surface area in this regions. The so calculated deposition is shown in Fig. 5 as calculated deposition.

RESULTS AND DISCUSSION

Fig. 6 shows the deposited surface area in the tracheobronchial region of the human lung. An about two-fold decrease in deposited surface area corresponds well with a nearly three-fold increase of ambient relative humidity. We believe that this is due to a humidity driven particle size increase resulting in sizes which are not monitored by the NSAM as the tracheobronchial or alveolar fraction.

Analysis of the results shows evidently the impact of relative humidity, wind speed and traffic on the particle burden in the size range below 1μm. Both particle metrics, particle numbers (Fig. 7.) and hence also the surface concentration, decrease with increasing relative humidity (Fig. 6.). The particle number concentration data from the SDMA (which means particles below 1 μm) shows an

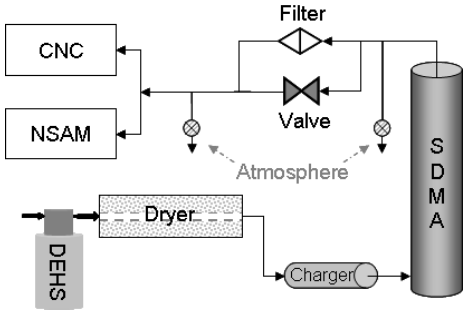

Fig. 4. Schematic diagram for the calibration arrangement of the nanosurface aerosol monitor.

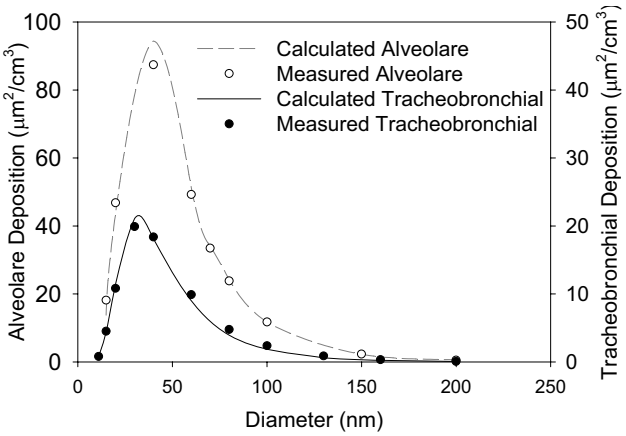

Fig. 5. Calculated and measured deposition in alveolar and tracheobronchial region for the calibration of the NSAM.

analogous behaviour to the NSAM data. Fig. 7 shows that the particle size distribution is decreasing sharply at 6 p.m. on Monday, 24 November. This corresponds well with the increase of relative humidity at the same time shown in Fig. 6. The lower number concentration during the night in Fig. 7 can be explained by the fact that traffic in the night time between weekdays is reduced to a minimum. Another effect to be seen is the impact of wind speed (plotted in Fig. 8.) to the particle number concentration (Fig. 7.). At around seven 7 a.m. on Tuesday, 25 November, during morning rush hour, one can clearly see the increase in UFP

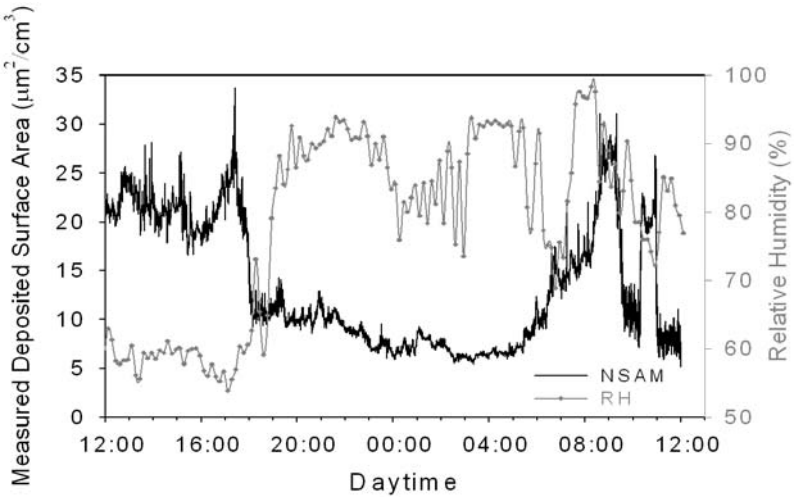

Fig. 6. Deposited particle surface area in the tracheobronchial region measured with the NSAM and the changes of the relative humidity at the same time from 24 November (Monday), noon, until 25 November 2008 (Tuesday), noon.

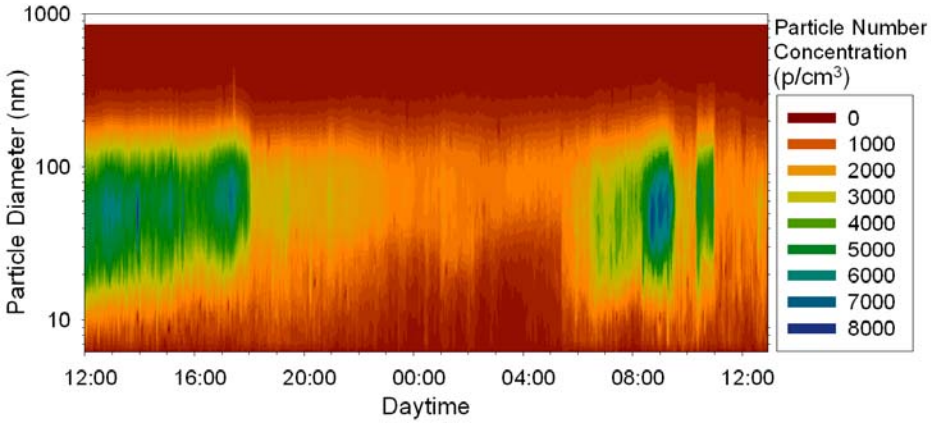

Fig. 7. Time resolved number concentrations measured with the SDMA from 24 November (Monday), noon, until 25 November 2008 (Tuesday), noon, at the same time as the data in Fig. 6.

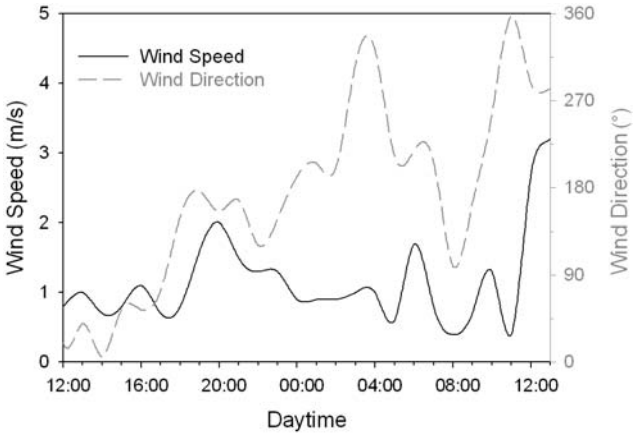

Fig. 8. Changes of wind speed and wind direction (90°-E; 180°-S, 270°-W, 360°-N), from 24 November (Monday), noon, until 25 November 2008 (Tuesday), noon, at the same time as the data in Figs. 6 and Fig. 7.

number concentration. Since the days of observation are both weekdays (Monday and Tuesday) it can be assumed based on observation that the traffic situation does not vary a lot between the two days. But unlike on the first day (left part of Fig. 7.), the particle burden on the second day reduces around 11 a.m. as the wind speed doubles at the same time as can be seen in Fig. 8. Since the relative humidity in the observed morning stays quite constant (as shown in the right part of Fig. 6.) it can be assumed that the increasing wind speed contributes to the decreasing of the particle number concentration and hence the particle surface area deposited in the lung.

A possible explanation for the decrease of particles in the PM_1 fraction during the higher relative humidity period is their humidity driven growth and coagulation meaning that they move into the $PM_{2.5}$ size fraction. Based on these findings it seems evident that the datasets provide representative information about ambient burden due to particles < 1 µm. Analysis of the PM_1 data from 24 November delivers PM_1 mass concentration from the CAVI measurement which amounts to 8.5 µg. The calculated mass concentration from the SDMA-data (Fig. 7.) before the number concentration drop (around 6 p.m.) amounts to around 10 µg/m³ and after the drop it is approximately 4

µg/m³. That corresponds to a loss of particles of about 6 µg/m³ from the PM_1 size fraction, likely due to a hygroscopic growth shifting those particles into the $PM_{2.5}$ size fraction. Obviously, PM filter sampling does not provide any insight into the actual situation and dynamics in the PM-fractions. Assuming that the particle loss of PM1 particles is due to their growth driven by the increased relative humidity their presence in the $PM_{2.5}$ fraction might not be observed due to the minute mass increase in this fraction and even possible water evaporation in the post sampling time.

A similar impact of the wind speed as described above can be observed in earlier data from 1–2 August 2008. In this case the measurement was performed from Friday (1 August) afternoon until noon of Saturday (2 August). Based on experience the traffic volume during the weekend night is not as low as during the week. Indeed there is a persistent presence of particles with a mode of around 100 nm (Fig. 10) which can also be observed for the deposited surface area (Fig. 9). This mode disappears around 3 a.m. with an increase of the wind speed by a factor of three (Fig. 11). A substantial increase of the concentration of nanoparticles down to 20 nm between 10 p.m. and 3 a.m. was detected by the SDMA and the NSAM. This higher particle burden period and the very sharp decrease around 3 a.m. shown in Fig. 10. coincidence very well with the evolution of the wind speed during this night (Fig. 11).

In both cases (Fig. 7 & Fig. 10) the wind direction does not seem to have a significant influence. In both above described datasets an impact of the wind speed is observed for velocities around 4 m/s.

Finally we investigated if there is a unique trend between the ratio of PM_{10}/PM_1, as well between $PM_{2.5}/PM_1$ with reference to the extinction coefficient σ_{ext} measured with the Vienna TPM. The experimental results are summarized in Fig. 12. It is evident that the extinction coefficient increases when the fraction of PM_1 within PM_{10} increases. An analogous effect can be observed for $PM_{2.5}/PM_1$. That shows the dominant influence of PM_1 particles on optical properties of atmosphere and the apparent correspondence between the PM_1 size fraction and the visual range.

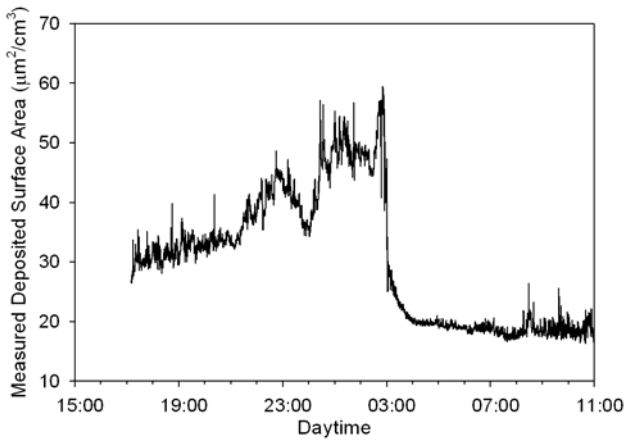

Fig. 9. Deposited particle surface area in the alveolar region measured with the NSAM from 1 August (Friday), afternoon, until 2 August 2008 (Saturday), noon.

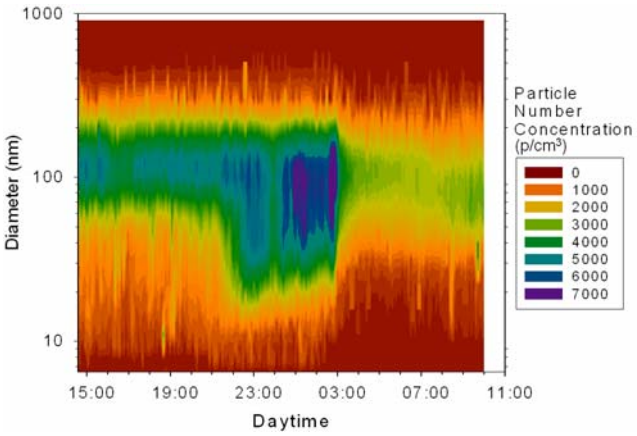

Fig. 10. Time resolved number concentrations measured with the SDMA from 1 August (Friday), afternoon, until 2 August 2008 (Saturday), noon, at the same time as the data in Fig. 9.

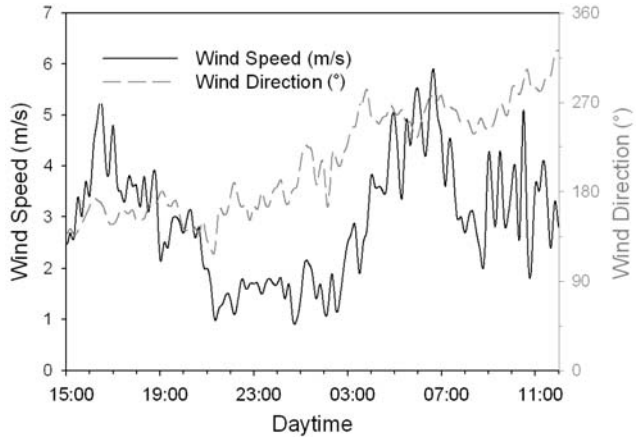

Fig. 11. Changes of wind speed and wind direction (90°-E; 180°-S, 270°-W, 360°-N), from 1 August (Friday), afternoon, until 2 August 2008 (Saturday), noon, at the same time as the data in Figs. 9 and Fig. 10.

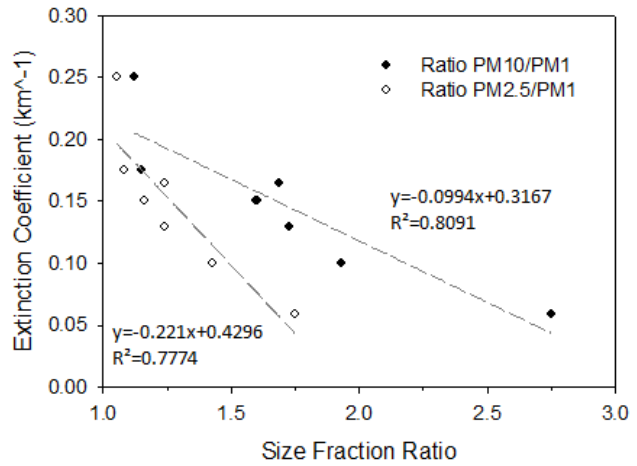

Fig. 12. Correlation between the extinction coefficient measured with the Vienna telephotometer and size fraction ratios obtained from mass concentration measurements with the CAVI.

CONCLUSIONS

Urban aerosol was investigated simultaneously by different instruments measuring various physical parameters of particles in question. The data show a strong correlation to daytime and weekdays, which can also be related to the traffic volume, hence it is clear that the motor vehicles contribute substantially to the ambient fine particles measured in Vienna urban area.

There is an evident correspondence between the observed time resolved data regarding the PM_1 fraction together with the ambient humidity and the wind speed. It was shown that an increase in wind speed decreases significantly the presence of the ultrafine particles. We found that this effect is not related to the wind direction. The changes and removal of PM_1 fraction due to ambient conditions can obviously not be determined using time integrated mass sampling. Moreover, such a measurement might lead to erroneous conclusions regarding air quality and possible health effects. The possible growth shift of PM_1 particles into the $PM_{2.5}$ size fraction due to humidity will likely contribute only minute influence on the measured $PM_{2.5}$ or PM_{10} values or maybe will even not be measured at all because of post-sampling evaporation processes. Consequently a presence of submicrometer ambient particles will not be detected or might be underestimated. Hence it is evident that not only PM_1 time integrated monitoring is of importance but also a time resolved monitoring of the PM_1 size fraction is very meaningful. The measurement of the extinction coefficient and the resulting visual range seems to deliver a good indication of the ratio of PM_1 particles to those in both larger fractions. A higher extinction coefficient corresponds to a larger PM_1 fraction within the $PM_{2.5}$ and the PM_{10} size fraction. The determination of atmospheric visibility is rather straightforward. Thus this measurement along with conventional PM measurements could represent a good possibility for the description of the actual PM_1 burden.

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