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MEASURING AEROSOL BLACK CARBON AGE WITH AETHALOMETERS

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INTRODUCTION

Aerosol mixing state is a parameter influencing optical properties of absorbing particles that is hard to measure. It was proposed that the lensing effect where transparent shell covers the absorbing particle can significantly enhance optical absorption (Bond et al., 2006). Aerosol mixing state can be assessed using advanced mass spectrometers ATOFMS (Healy et al., 2012), and with SP2 for particles with diameters above about 100 nm (Subramanian et al., 2010).

Filter-based measurements of aerosol optical absorption are widely used to determine Black Carbon (BC) concentrations in real time. Measurements at multiple wavelengths permit the separation of contributions of BC from different combustion sources (Sandradewi 2008). However, filter-based methods are influenced by the non-linear response due to the »loading« effect, caused by the increasing sample deposit on the filter (Gundel 1984, Weingartner 2003, Arnott 2005, Virkkula 2007).The filter-loading effect is the reduction of the filter based photometer sensitivity due to filter loading. It was shown that filter-loading effect differs between locations and seasons with indication that the filter loading effect in Aethalometer relates to the particle coating (Virkkula et al., 2007; Drinovec et al., 2015). Using Aethalometer model AE33 (Magee Scientific) it is possible to measure filter loading parameter k with high time resolution (Drinovec et al., 2015).Influence of coating on the parameter k was investigated both during the ambient and laboratory campaigns.

METHODS

The dual-spot Aethalometer (Magee Scientific model AE33) provides a real-time determination of the filter loading effect. The compensation parameter k is determined in real time for each of the operational wavelengths, and is used in the same manner as has been developed for off-line post-processing. This provides a time resolved specific spectral fingerprint that may be interpreted in terms of aerosol composition. Optical and chemical properties of aerosols were measured with high time resolution during summer and winter EMEP campaigns in Paris (France) and Payerne (Switzerland). An Aerosol

Chemical Speciation Monitor (ACSM) and a High Resolution Time-of-Flight Aerosol Mass Spectrometer (AMS) were used to measure quantitative chemical composition for non-refractory aerosol particles.

RESULTS

During summer we observed complex temporal variation of k, where k(880 nm)=k_6 changed from approximately 0.006 for fresh aerosols to near zero for aged aerosols as shown using Potential Source Contribution Function method for back trajectory analysis. We have combined the Aethalometer and ACSM/AMS measurements, and normalized the sum of inorganic secondary and organic aerosol mass to BC to obtain the coating factor. Values of this coating factor are expected to be high for air parcels containing aged aerosols. The ratio correlates well with the loading compensation parameter k measured by the Aethalometer at 880 nm (Figure 1). This indicates that the compensation parameter k may be used for discrimination between local (fresh) and regional (aged) air pollution aerosols.

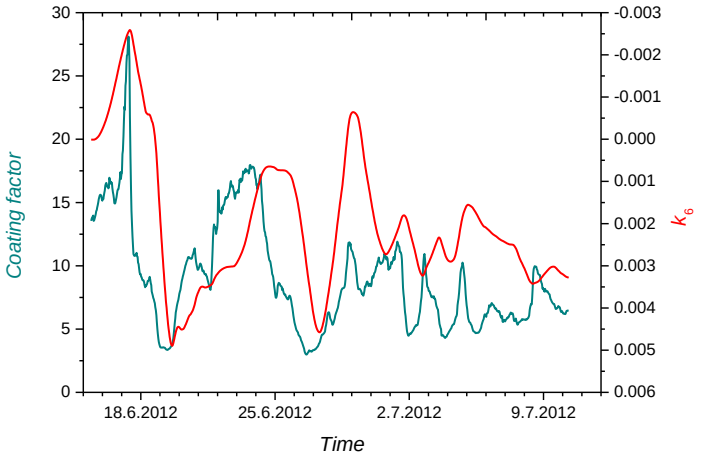

Figure 1: Coating factor and filter loading parameter k=k_6 time series during the Paris summer campaign. Note the inverted scale for k.

To investigate the influence of coating on parameter k, a drier and thermo-denuder were used to remove the coating of ambient aerosols during the campaign conducted in Ljubljana, Slovenia. Size distributions measured by SMPS and SEM images (Figure 2) have been used to determine the mixing state of the aerosols during the ambient campaign. Placing a drier and/or a thermo-denuder in the sample stream both caused the increase of parameter k at 880 nm (Figure 3), which is indicative for fresh uncoated aerosols. The effect of aerosol drying implies presence of hygroscopic coating material.

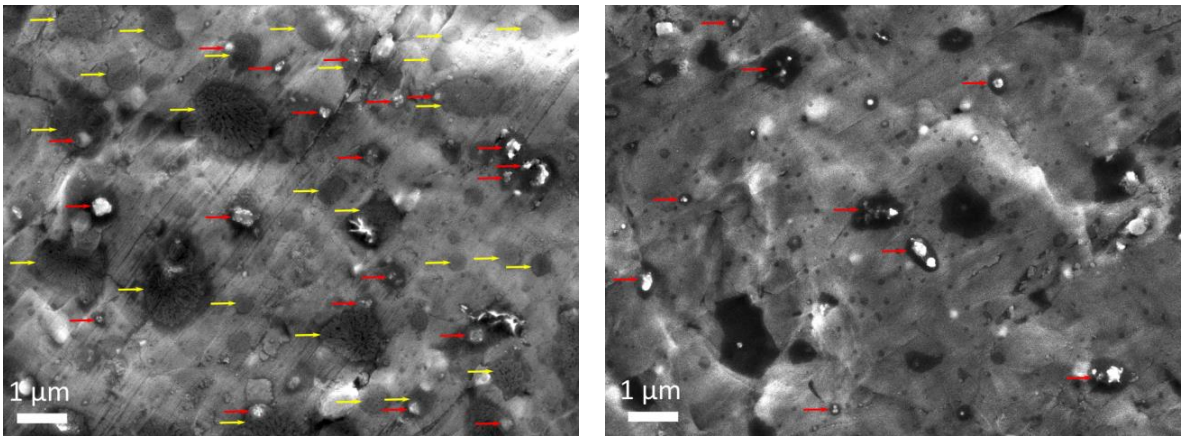

Figure 1: SEM images of ambient (left) and thermo-denuded (right) impactor samples (size range 170 – 260 nm) taken during the Ljubljana campaign. Red and yellow arrows mark soot agglomerates and secondary ammonium sulfate residue, respectively.

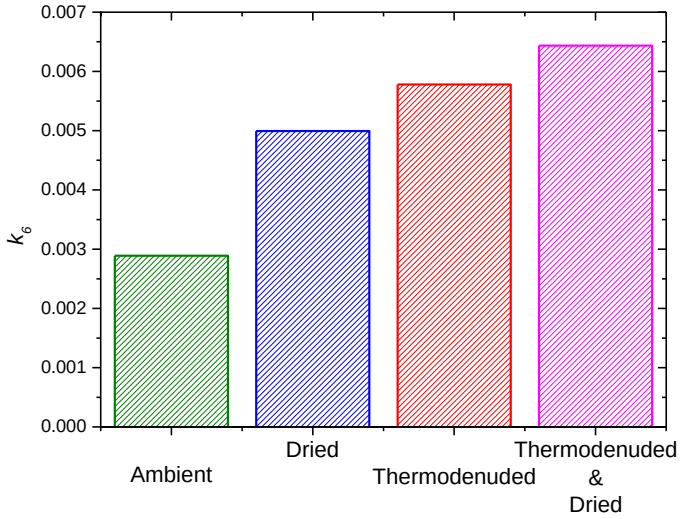

Figure 2. Influence of dried and/or thermo-denuded ambient aerosols on the filter loading parameter k6, campaign averages.

CONCLUSIONS

High time resolution measurements of the filter loading parameter in Aethalometer show daily and seasonal variations of the effect. The loading parameter values for fresh combustion products seems to be influenced by the size of the combustion products particles with an approximately 15% increase in the infrared wavelength region with for an aerosol mobility diameter increase of about 4 times. On the other hand, we observed a reduction of the filter loading effect correlated with the availability of the coating material. High coating factor coincides with low value of loading parameter and vice versa. These results show that during summer the coating material causes reduction of filter loading effect. Using a thermos-denuder to remove the coating, we show that the filter loading effect of ambient aerosols is increased, as the coating is reduced or removed. The coating composed of ammonium sulphate and secondary organics seems to be responsible for the variation of the loading effect. The potential source contribution function analysis shows that high values of the filter loading parameter in the infrared are indicative of local pollution, whereas low values of the filter loading parameter result from ageing and coating during long range transport. Results show that the filter loading parameter can be used as a proxy for determination of the particle mixing state, thus allowing to differentiate between local/fresh and transported/aged particles. Filter loading parameter is thus not only important for compensation of the Aethalometer absorption data but also provides additional information on the physical properties of aerosols.

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