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An empirical correction factor for filter-based photo-absorption black carbon measurements

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ABSTRACT

Filter-based BC measurement techniques such as the Continuous Soot Monitoring System (COSMOS) are particularly well suited to long-term observations of black carbon (BC) due to their relative robustness and reliability. However, caution is required when determining the threshold transmittance, Tr_{thresh} (roughly proportional to the time interval between filter changes), in order to ensure that acceptable measurement accuracy is maintained throughout the sampling period. We present a new, empirically derived transmittance-dependent correction factor used to interpret the response characteristics of filter-based aerosol absorption measurements performed by COSMOS. Simultaneous measurements of ambient BC aerosol mass (M_{BC}) were conducted in Tokyo, Japan, using two identical COSMOS instruments operated with different threshold transmittance, Tr_{thresh}, values, of 0.95 and 0.6. The derived values for M_{BC} were consistently underestimated by the COSMOS operating at lower Tr_{thresh}, as a function of decreasing filter transmittance. The 1-hour averaged values of M_{BC} were underestimated by around 10%, incorporating measurements across the entire range of filter transmittance (1–0.6), with a maximum underestimation at around 17% immediately preceding filter advancement (i.e. $Tr = \sim 0.6$). An empirical correction factor was derived from these ambient measurements, and was applied to M_{BC} as a function of filter transmittance, resolving the instruments to within 2%.

Further to the transmittance-based correction, the operational performance of COSMOS was tested for two types of quartz fibre filter (PALLFLEX and HEPA). Agreement in derived values of M_{BC} for two COSMOS using the same type of filter was around 2%; however, a comparison of the PALLFLEX and HEPA filters demonstrated a systematic overestimation of M_{BC} derived when using HEPA filters, of around 6–8%. A sensitivity study of a radiative transfer model indicated that this enhanced absorption was primarily a result of the increased thickness of the HEPA filter.

1. Introduction

1.1. Long-term measurements of black carbon mass concentrations

Long-term continuous on-line measurements of black carbon (BC) mass concentrations (M_{BC}) are a fundamental requirement for constraining atmospheric models, providing valuable information, such as seasonal trends, that may not be observed during short-term field deployments. However, such measurement techniques require significant labour and

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infrastructure costs, and are typically not as quantitatively detailed when compared to short-term measurements due to the methods used (e.g. either poor or zero size-resolution). Filter-based techniques are intrinsically limited by: measurement artefacts due to particle concentration, changes in morphology due to deposition, and the optical interaction of deposited particles due to their penetration depth (Moosmüller et al., 2009). There are a number of widely-used filter-based photometers commercially available, such as the Aethalometer (Magee Scientific, Berkeley, CA) (Hansen et al., 1984; Weingartner et al., 2003), Particle Soot Absorption Photometer (PSAP; Radiance Research, Seattle, WA, USA) (Bond et al., 1999; Virkkula et al., 2005), Multi-Angle Absorption Photometer (MAAP; Thermo Scientific, Waltham, MA, USA) (Petzold et al., 2005), and Continuous Soot Monitoring System (COSMOS; KANOMAX, Inc., Japan) (Kondo et al., 2009, 2011).

Aethalometer and PSAP measure the transmission of light through a loaded filter at specified wavelengths and compare it to the transmission of light through a clean filter, resulting in the derivation of M_{BC}, which requires careful correction for non-absorbing (light scattering) aerosol. Measurements of M_{BC} by the Aethalometer and PSAP require additional data (e.g. from nephelometers) for specific treatments to account for the optical influences of non-absorbing (non-BC) compounds present within the ambient aerosol, which significantly impact on their light extinction properties.

MAAP was designed to reduce the uncertainties in BC measurements caused by aerosol scattering. The optical absorption coefficient of aerosol collected on a filter is determined by radiative transfer considerations, which include multiple scattering effects and absorption enhancement due to reflections from the filter (Petzold et al., 2005). However, the accuracy of M_{BC} measured by MAAP is not well quantified (Kanaya et al., 2013).

COSMOS allows for a fully automated, high-sensitivity, continuous measurement of light absorption by BC aerosol (Miyazaki et al., 2008). COSMOS measures changes in transmittance between a clean and exposed section of automatically advancing quartz filter tape using a light-emitting diode (LED) at a wavelength of 565 nm (Miyazaki et al., 2008). The aerosol sample air stream enters COSMOS on the side of the unit, and is introduced to the surface of the quartz filter. The quartz filter will remove all aerosol particles, and the resultant particle-free sample air passes through the back of the reference portion of the quartz filter. COSMOS includes a heated inlet to effectively volatilise non-refractory aerosol components internally mixed with BC (Kondo et al., 2011), and provides automated filter changes according to a pre-determined threshold transmittance value. The measurements of M_{BC} by COSMOS have been estimated to be about 10% (Kondo et al., 2011).

A transmittance-dependent correction factor is used in deriving M_{BC} by COSMOS, based on the experiments using polydisperse nigrosine ($C_{48}N_9H_{51}$; Bond et al., 1999). However, it is not obvious if the same factor can be applied to BC particles. There has been no measurement of the correction factor for BC thus far. In this work, we have derived such a correction using ambient BC in Tokyo.

Until now, COSMOS instruments have routinely utilised PALLFLEX quartz filters (E70-2075W, hereafter simply referred to as PALLFLEX; the same filter used for PSAP), but due to a change in supplier, an alternative quartz filter (HEPA L-371M; hereafter referred to as simply HEPA) has been proposed for use in future measurements. The PALLFLEX filters were used for the original characterisation of COSMOS (Miyazaki et al., 2008), and have shown good agreement with other techniques used to measure ambient M_{BC} (e.g. Kondo et al., 2011). However, the performance of the HEPA filter is unknown, and there is a urgent need to characterise it. In this study, we also assess the applicability of the HEPA filter for use with COSMOS.

2. Measurements and methods

2.1. Experimental setup

We performed a series of experiments using ambient COSMOS measurements to (a) assess the impact of filter transmittance deterioration on BC measurements (for both PALLFLEX and HEPA filters), and (b) test the suitability of the HEPA filters for COSMOS measurements, by directly comparing the HEPA and PALLFLEX filters (resulting in the HP-factor). For these tests, two COSMOS were operated in parallel, in the configuration outlined in Fig. 1a.

The aforementioned sampling procedure takes place within the 'optical block', which is heated to a temperature of 50 °C to minimise the influence of high ambient relative humidity fluctuations on the measurement (a source of uncertainty with PSAP measurements). Another key advantage of COSMOS is that COSMOS automatically advances the quartz filter by 20 mm when the light transmission value recorded by the instrument falls below a predefined threshold value (Tr_{thresh}).

The COSMOS measurements took place downstream of an inlet heater, or thermal denuder, and a particle impactor (with a size-cut of 2 μm), as shown in Fig. 1. Following the results from detailed laboratory experiments performed by Irwin et al. (2013), an operational temperature of 300 °C was chosen for the inlet heater. COSMOS operates with a high-consistency constant flow rate of 0.7 STP LPM ± 1%, controlled by a Mass Flow Controller (MFC; SEC-E40MK3, HORIBA ESTEC, Kyoto, Japan). During these experiments, the number and mass distributions of ambient BC were periodically measured by an SP2, simultaneous with COSMOS measurements.

2.2. COSMOS operating principle

COSMOS records values of light transmission (Tr) at 1 kHz based on the transmittance of a section of blank filter (I_0), and the transmittance of the filter area bisecting the aerosol flow at time t, (I_t):

$$Tr = I_0/I_t \quad (1)$$