

Author reply to Review by Chris Cappa and Dan Lack:

We thank Chris Cappa and Dan Lack for their valuable comments which helped improving the exactitude and clarity of the manuscript.

Referee comments are repeated in black font, author replies are given in red font.

Overview

In the study of Laborde et al., absorption measurements were made using an Aethalometer at 880 nm, which is a filter based measurement technique. In reporting their mass absorption coefficient (MAC) results for different air mass types (traffic, biomass burning, aged, continental), they have assumed that the scattering “correction factor,” C , that is needed to turn the measurement (I/I_0 of light passed through the filter) into an absolute absorption coefficient is constant. They make this assumption because they do not have data available to suggest otherwise. We suggest that this is a reasonable assumption, but that the authors do not fully consider the uncertainty introduced by it.

In Section 3.3.4 they report that the observed mean mass absorption coefficient ranges from $7.8 \text{ m}^2/\text{g}$ (traffic) to $8.8 \text{ m}^2/\text{g}$ (aged) at 880 nm, with MAC values for biomass burning and continental air masses in between. They go on to compare their campaign average 880 nm value with the literature estimate of the MAC at 550 nm, extrapolating using an Angstrom exponent of 1. They find that their extrapolated MAC is $13.6 \text{ m}^2/\text{g}$, much higher than the literature value of $7.5 \pm 1.2 \text{ m}^2/\text{g}$ from Bond and Bergstrom (2006). They offer two explanations: (1) that the scattering correction factor is incorrect or (2) that the Bond and Bergstrom results were only for “fresh” emissions. However, they do not compare their “fresh” (i.e. traffic) results to Bond and Bergstrom (2006), which we suggest is the more appropriate comparison since, as Laborde et al. state, Bond and Bergstrom “reported only freshly emitted BC MAC values”. Extrapolating $7.8 \text{ m}^2/\text{g}$, the “fresh” value reported in this study at 880 nm, to 550 nm gives $12.5 \text{ m}^2/\text{g}$, which is also much higher than the literature value. Therefore, this strongly suggests that the reason for the larger mean MAC during this study is due to inadequacies in the scattering correction factor, which leads to an overestimate of (and large uncertainty in) the actual absorption at 880 nm. We strongly encourage the authors to compare both their mean values and traffic values to the Bond and Bergstrom results, as has been done above.

We fully agree that light absorption coefficients derived from filter-based measurements are tainted with experimental uncertainty. The uncertainty of MAC values further depends on the uncertainty of the EC or rBC measurement. Thermal-optical EC mass measurements are, depending on the aerosol properties, also known to be associated with substantial uncertainty. This has to be kept in mind when e.g. comparing results from different studies. The discussion of the MAC values has been revised in response to this and other referee comments. It now reads:

“The average MAC of the entire dataset is $\sim 8.6 \text{ m}^2 \text{ g}^{-1}$ at 880 nm (Fig. 12C and Table 1). This value is in agreement with previous measurements of the MAC in wintertime Paris by Sciare et al. (2011), who reported a value of $\sim 7.3 \text{ m}^2 \text{ g}^{-1}$ at 950 nm, which translates to $\sim 7.9 \text{ m}^2 \text{ g}^{-1}$ at 880 nm (using Eq. 5 and assuming $AAC=1$). On the other hand Healy et al. (2012) reported a substantially lower MAC value for the measurements that took place at city centre of Paris (LHVP site) during the same time period ($5.1 \text{ m}^2 \text{ g}^{-1}$ at 550 nm, which translates to $\sim 5.5 \text{ m}^2 \text{ g}^{-1}$ at 880 nm). This substantial difference may partly be explained with a relatively higher contribution of fresh traffic emissions and partly by experimental uncertainties of the light absorption and EC mass measurements. Bond and Bergstrom (2006) reported a MAC value of $7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$ at 550 nm ($\sim 4.7 \text{ m}^2 \text{ g}^{-1}$ at 880 nm) for fresh uncoated BC. This is substantially lower than the $7.8 \text{ m}^2 \text{ g}^{-1}$ at 880 nm reported here for strong traffic influence, and the difference to other air mass types is even larger. Part of this difference may be explained by the fact that some aged background aerosol is also present during traffic influence. However, experimental uncertainties commonly associated with light absorption and rBC/EC mass measurement may also play a role. Indeed, a constant correction factor C (see Sect. 2.4), determined from a short-time comparison with a MAAP, was used here to derive the light absorption coefficient from the aethalometer measurement. This could potentially introduce a bias of up to 40% whenever

the aerosol properties differ a lot from those at the time when the correction factor C was determined.”

If the scattering correction factor, C , is indeed insufficient to provide an accurate estimate of the absolute absorption, which seems to be the case, one needs to further ask the question of to what extent is C constant in time and independent of the aerosol composition? Implicit in the authors' comparison of MAC values between different air mass types is the assumption that C is air mass independent. This assumption may not be justified since filter based absorption measurement methods have previously been shown to suffer, at times, from biases that depend on the composition and abundance of non-BC particle components (Lack et al., 2008; Cappa et al., 2008). Although these particular studies cited used PSAP instruments, and not Aethelometers, it is reasonable to think that both PSAP and Aethelometer instruments would suffer from similar biases given that both are filter based. Given that the reported mean MAC values for the different air mass types differed by only 15%, and given the large magnitude of the biases observed in Lack et al. (2008), it would seem that the changes reported here in the MAC between different air mass types are well within the actual uncertainties of the measurement technique. We suggest that the impacts of such potential time/air mass-dependent biases in the scattering correction (and thus the absorption measurement) be discussed in much more detail and that the discussion and conclusions be revised accordingly. We have concerns that no firm conclusions can be made regarding the atmospheric variability of the MAC once the measurement uncertainties associated with the scattering correction are considered.

We agree that assuming a constant correction factor C might introduce a systematic bias for certain air mass types. This is acknowledged with the following addition to the manuscript:

“The MAC values reported above for different air mass types were derived with using a constant correction factor C (see Sect. 2.4) for the evaluation of the aethalometer measurements. However, C is known to depend on several factors including the abundance of non-BC particle components and the BC particle size (e.g. Lack et al., 2008; Cappa et al., 2008). The observed 20% difference between the MAC values under the influence of traffic emissions and aged air mass may therefore be under- or overestimated.”

References:

Bond, T. C., and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, *Aerosol Science and Technology*, 40, 27-67, 2006.

Cappa, C. D., Lack, D. A., Burkholder, J. B., and Ravishankara, A. R.: Bias in filterbased aerosol light absorption measurements due to organic aerosol loading: Evidence from laboratory measurements, *Aerosol Science and Technology*, 42, 1022-1032, doi: Doi 10.1080/02786820802389285, 2008.

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