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Drinovec et al.: The filter loading effect by ambient aerosols in filter absorption photometers depends on the mixing state of the sampled particles

REVIEW

GENERAL

In this paper the authors have analyzed the information that can be obtained from the filter loading parameter that the new aethalometer model outputs in addition to BC concentration. The paper presents field and laboratory measurements of various light-absorbing aerosols and discusses factors affecting the loading parameter. The paper is in general easy to read and the results will surely prove out to be very useful for the community measuring BC. I can recommend publishing the paper with minor revisions.

The most significant misunderstanding is already in the title of the article. The data indeed suggests that the loading parameter depends on the size and coating of the BC particles and thus their age but there is no evidence of that it would depend on the mixing state. Fig (6) shows that k_6 has very different values as a function of the coating factor defined in eq. (3) in winter and summer, that is true. On line 396 it is written that in winter aerosol is more externally mixed, which is very probably true also. The next argument is not. It is argued that since k₆ is higher in winter it is due to the state of mixing. Why would in not be due to the thinner coating of the BC particles? After all, they are the particles that affect light transmittance most in the filter medium. Let us assume that the aerosol is an external mixture of light-absorbing and scattering particles. To show that the factor that affects k₆ is really the state of mixture and not the coating of the light-absorbing (= BC, here) particles, the light-scattering particles should be removed without affecting the coating or any other property of the BC particles. So that only the state of mixing would change. Doing that would prove that it is the state of mixing that affects the k_6 . The thermodenuder field experiment, section 3.6, shows that after heating and drying the sample air the k_6 is higher. Again, this is no evidence of the effect of the mixing state on k₆. By heating the sample air the externally-mixed light-scattering particles get removed but also the coating of the BC particles gets thinner or totally removed. As a result, the size of the light-absorbing particles decreases which leads to changes in the penetration depth into the filter and also increase in backscatter fraction which is inversely proportional too the particle size. And then k₆ grows. So I claim that the change of k₆ is more probably due to the change of the coating of the BC particles and not due to the removal of the purely light-scattering particles, i.e., change of the mixing state.

Actually, I can't really find any other way to prove that k is affected by the state of mixing alone but to make an external mixture of scattering and absorbing aerosol: first measure pure BC particles from a chamber for a while, then blow in also scattering aerosol and keep measuring. If k changed just due to the addition of scattering aerosol, the title of the paper would be true. The evidence from the experiments presented in the paper rather only suggest that the loading effect depends on the coating of the light-absorbing particles. I strongly suggest changing the title accordingly. Also in the conclusions it is written "*Our 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*". Based on my above argumentation I would remove the statement about mixing state but it can be used to differentiate between local/fresh and transported/aged aerosols.

Detailed comments

P4, L111. "The absorption coefficients are calculated" How are they calculated? What C_{ref}? Based on what? Any scattering corrections?

P5, L135-143, Description of the electron microscopy. What compounds and/or elements were obtained? How many particles were analyzed?

You also present single-scattering albedo later. How was scattering measured? Describe also its measurement.

What was the time resolution of each instrument?

P6, L193: "... was routed to multiple instruments." Which instruments? The first 35 seconds of combustion was measured. Why so short? Was the burning at any kind of stable state then? I assume aerosol concentrations simply decreased. How long did you measure? Time resolution?

P10, Table 2. Why don't you also give the wavelength dependency of the k1 - k7 in the results? This applies also to the other result tables and figs.

P12, L308: "... mean mobility diameter..." which mean? Geometric? Arithmetic? Volume or number?

p12, L311 – 313. When comparing AAE you should use as similar a wavelength range as possible (cf. Lack & Cappa, ACP 10, 4207–4220, 2010, Fig 8). How would your AAE be using 370 – 880 nm?

P13, Fig 6b. Give units for y axis. Why do you use aerodynamic diameter if you measured with an SMPS? To calculate aerodynamic diameter from mobility diameter you need density. What did you use? How high was the mustard oil nucleation mode? Now the y axis is limited. True, particles smaller than 20 nm contribute little to absorption and especially scattering but if there are very many of them they may affect the aethalometer. You could calculate the contribution of the two modes to absorption. The two mustard-oil modes are probably chemically and thus also optically different. Comments on that?

P13, Table 3. What mean diameter? GMD? Give also the width as GSD.

Please make one more figure that shows time series of the lab experiment data.

P17, L439: "... the coating may also be responsible for the decrease of parameter k." And coating probably also increases SSA and decreases backscatter fraction. Consistent with Virkkula et al. (2015)

And one more comment: Reviewer 1 writes "*to my knowledge, there is no dependency of the AAE to the aerosol size (or size distribution).*" Sure particle size affects also AAE. It is easily modeled and has been published, e.g., by Gyawali &al, ACP 9, 8007–8015 (2009); Lack & Cappa, ACP 10, 4207–4220 (2010)