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Atmospheric Measurement Techniques Discussions

Interactive comment on "Comparison of co–located rBC and EC mass concentration measurements during field campaigns at several European sites" *by* Rosaria E. Pileci et al.

Anonymous Referee #1

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The manuscript reports the results of a general intercomparison of two established yet relatively distinct methods for the analysis (monitoring) of strongly light-absorbing particles (aka soot) in the atmosphere at four different locations in Europe. While the manuscript is based on a large body of measurement data using up-to-date instrumentation and integrating past knowledge into the evaluation of their output, there are a number of concerns that need to be resolved before publication.

The primary objective of the authors is to present a universally valid statistical evaluation of co-located independent measurements of BC (or its surrogates). The issue of light-absorbing carbon in the atmosphere and its measurements is probably one of the

most debated and least resolved problems in atmospheric science. While the authors give a relatively good overview of the specific problems associated with EC/BC measurements, in highlighting the general background (i.e. the nature of light-absorbing carbon in the atmosphere) they fail to account for established prior knowledge in the field. They seem to cherry-pick information from the literature, ignoring important findings that would be needed for better understanding the complexity of the problem and the difficulties with the selected methodologies. For example, the issue of light-absorbing carbon continuum is referred to in the manuscript as being newly discovered (supported by two recent papers of the authors themselves, line 80-85), though it has been on the agenda of aerosol science for nearly 20 years. In general, more focus should be put on this issue, including brown carbon, as this may greatly affect differences between the results of the measurements. Some statements such as 'brown carbon absorbs much less than EC at the red wavelength (λ = 635 nm)' (line 211-213) are largely outdated and ignore more recent findings that there are actually two types of brown carbon in aerosol, the strongly absorbing tar balls (carbon spheres) and the weakly absorbing ones (organic chromophores), the former have also been shown to absorb in the near infrared (see e.g Alexander et al. 2008, Saleh et al., 2014, Hoffer et al., 2017).

My another major concern is that even if the authors primarily make 'lump statistics' on all their measurements data, their individual measurement setup is different at each site (e.g. different cutoffs, dryers, SP2 instruments, etc.). In measuring such small-sized and adhesive species even the type and length of the tubing may introduce significant uncertainties due to wall losses, these are likely also different in the measurement sites but not reported here. My fundamental question is the following: if we take into account these uncertainties (or biases?) and the other existing uncertainties (and potential biases) correctly evaluated and reported in the manuscript (I counted 8 significant sources of uncertainties but these may not be all), correctly apply the rules of error propagation, will the overall 8 % difference between the two methods be statistically significant at all? My informed guess is that it will not. So contrary to the key statement in the manuscript that ‘median ratio between observed rBC and EC mass concentrations was 0.92’, a more realistic statement would be that ‘the two independent methods are indistinguishable within the limits of inherent uncertainties’. Overall, the general impressions from the discussions and conclusions tacitly support this latter statement as the inexplicable variations (to either directions) of campaign-wise data do not reveal any systematic difference between the two methods. This is particularly true since the individual campaigns at the four sites were markedly different in duration (14, 21, 24, 30, 54 days according to my calculations), so the ‘mean data’ reported in this manuscript refer only to this particular combination. Should the durations of the individual campaigns be different, the overall finding would have been very much different, at least within the limits of extremes. Campaign-wise discussions would have made more sense than calculating a ‘European average’ value with this fixed setup (which definitely does not exist).

My last major concern is that the authors do not exploit the possibilities of elaborating differences within the individual campaigns (though the campaigns are not too lengthy in themselves). For example, dependence of measured results on trajectory directions at the Melpitz site should have been elaborated to verify the hypothesized effect of coal burning from Eastern Europe. Although these evaluations would not involve statistical analyses, they might still be useful to imply some of the effects that are hypothesized in the manuscript.

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