Answers to referee 2

Comparison of co–located rBC and EC mass concentration measurements during field campaigns at several European sites

We thank the reviewer for the timely and constructive review of our manuscript, particularly during these difficult corona times. Please find below reviewer comments repeated in black text and our responses in blue text.

1) Some sections of the manuscript, especially the methods are excessively wordy and could be substantially tightened as too much background information is given that is neither relevant nor appropriate for a method comparison paper. It is unusual in a method section to have paragraphs explaining the basic functioning of commercial instruments (e.g. SP2).

The methods section will be shortened.

2) There is also an excessive discussion of artifacts in the thermal methods that does not really belong here as it does not seem relevant because it is not included in the results discussion. In fact a lot of the discussion is on TOR (reflectance) and the IMPROVE method, when the authors actually use TOT (transmittance). They seem to confuse themselves as in table 1 they refer to IMPROVE protocols as TOT. So this needs to be cleaned up and checked for accuracy. Also ENCan-total-900 (see Sharma et al., ACP, 2017) is neither TOT nor TOR (see your table 1) but I guess you would call it TOA as it has no pyrolysis correction neither by reflectance nor by transmittance. So less text and more accuracy. (BTW quite a few people also use CTO-375, not mentioned at all).

The discussion will be shortened retaining focus on the methods applied in this or previous rBC/EC intercomparison studies. TOT/TOR information provided in Table 1 will be corrected. Table 1 addresses all protocols applied in studies included in Fig. 6, and CTO-375 protocol was not mentioned, as it is not among these. Anyway, Table 1 and Section 2.2.2 will be shortened in response to comment 10) given below.

3) There is a 7 year delay between the first and the latest studies. With changes in Diesel emission regulations and in car/truck fleet overall in Europe and significant differences between countries France/Germany/Italy... one wonders any impact of this on observations. Still there is no discussion at all on temporal and spatial variability of diesel emissions. Same applies to other "soot" emission sources such as heating, there is only a small discussion on coal. The sites are very different and one would expect different source contributions, which will substantially impact results.

The molecular structure of BC emitted from diesel engines can vary between different engines and engine operation conditions. This variation could potentially affect either method as reactivity and optical properties of BC are related to it. In particular, BC with a lower degree of graphitization is more easily oxidized in the diesel particle filters (DPF) included in modern diesel engines (e.g Schmid et al., 2011). However, emissions from more modern cars equipped with DPF are marginal compared to older cars without DPF, resulting in a decreasing trend of traffic BC emissions and a corresponding shift towards larger relative contributions from other BC sources as a potential longer-term trend in the observed ratios. Furthermore, seasonality and regional differences are at least as important, as seen from clear differences between the BC particle properties and intercomparison results for the Melpitz summer and winter campaigns. We used back trajectory analysis and the "aethalometer model" to assess potential systematic relations between sources and

observed ratios (Sect. 3.3.3); however, the results were not conclusive for the reason stated in the conclusions: "The discrepancy between rBC and EC appears to be systematically related to the BC source, i.e. traffic versus wood and/or coal burning. However, it was not possible to identify causalities behind this trend due to potential cross–correlations between several aerosol and BC properties relevant for potential biases." - It is unrealistic to expect that the available data set would provide the basis to demonstrate a fleet modernization effect and hence we refrain from undertaking such an attempt.

Schmid, J., Grob, B., Niessner, R., and Ivleva, N. P.: Multiwavelength Raman microspectroscopy for rapid prediction of soot oxidation reactivity, Anal. Chem., 83, 1173–1179, doi:10.1021/ac102939w, 2011.

4) Related to the sites. It seems a little "odd" to refer to Paris as a European background site (L22). So I recommend streamlining the names of the sites, so for Paris call it or Paris or Palaiseau but not randomly one or the other plus the site code confuses even more as it is different form the other two. Also overall I am not convinced that it is appropriate to consider Palaiseau as a background site. The same applies to the CNR site in Bologna, which is quite central and not what one would think of as a Po Valley background site. Please be clearer in the description of the sites and the local impact Cabauw is described by its distance to the sea, which is funny when it is closer to both Rotterdam (a major port with related truck traffic) and Utrecht than the ocean.

The wording chosen in the abstract was indeed imprecise, whereas the classification provided along with the site descriptions in Sect. 2.1 is appropriate (i.e. "suburban background" and "urban background" for SIRTA and CNR Bologna, respectively). We will replace "SIRTA" by Palaiseau throughout the manuscript. As for Cabauw, we will add the distances to the Rotterdam and Utrecht to the site description.

5) A critical scientific issue is artifacts because of particle sizes. This is discussed to a certain extent. However, here again one wonders why there is not more discussion on local sources and differences between sites which will impact particle processing and association of refractory BC and EC with larger (or smaller) particles. That issue is passed upon. In particular for larger particles and the fact that Cabauw has a PM10 inlet vs a PM2.5. it misses completely that many studies documented that in processed aerosol EC and BC are associated to a significant amount with larger particles.

Several processes alter the size distribution of BC containing particles. For example condensation of secondary particulate matter or coagulation with BC-free particles will increase the aerodynamic diameter, whereas the BC mass equivalent diameter, which determines the SP2 upper cut-off, remains unaffected by condensation. Some studies indicate a shift towards smaller BC core diameters during transport with precipitation due to preferential wet-removal of larger particles. As for our study, we have provided a quite extensive discussion on upper cut-off effects in Sect. 3.3.1. Cabauw was among the sites with a small modal size in terms of rBC core mass equivalent diameter, which does not exclude a second mode of supermicron BC cores. For Melpitz, where EC measurements were available with different PM cut-off diameters, we tried to identify a relationship between supermicron EC fraction and rBC vs EC discrepancy - without a significant result. It is correct that this question is passed upon as we clearly state in the conclusions: "The discrepancy between rBC and EC appears to be systematically related to the BC source, i.e. traffic versus wood and/or coal burning. However, it was not possible to identify causalities behind this trend due to potential cross–correlations between several aerosol and BC properties relevant for potential biases.

For future intercomparison studies, it is important to constrain the upper cut–off and potential inlet losses of both methods in such a manner that these can be excluded as a source of discrepancy."

Other (details)

6) The manuscript preparation could benefit form more attention to detail and is quite careless 2 examples: 1) basic text formatting, starting with the affiliations where none of lines are really aligned in how they start. 2) Melpitz coordinates "MEL; 51◦ 320' N, 12◦ 560' E" really?

These technical edits will be implemented and typos corrected.

7) Please do not use qualitative statements that have no meaning e.g. abstract "the high correlation" what does this mean? Is it statistically significant? Is it not? "high correlation" has no intrinsic meaning. Same in the text.

We will replace "high correlation" with more precise statements along the line: "…the observed correlation between rBC and EC mass reveals a linear relationship with a constant ratio, thus providing clear evidence that both methods essentially quantify the same property of atmospheric aerosols…"

8) The abstract is too wordy and especially the second paragraph has no quantitative information is provided. You do not need to have a 3 paragraph abstract.

The abstract will be shortened.

9) Your referencing is not very up to date. Many recent papers addressed EC and BC optical properties especially relative to the aethalometer including brown carbon and how this relates to SOA and biomass burning, at the wavelengths used! Please update your referencing and include recent work insights there in your discussion.

Our manuscript does not deal with comparing optically derived equivalent black carbon, eBC, with EC and/or rBC. Spectral dependence of brown carbon optical properties come into play for the optical charring correction. This is, however, only of secondary importance for the intercomparison results of this study. More relevant is the refractoriness and reactivity, which is the primary criterion affecting the split between BC and other particulate matter in either method. "Brownness" and refractoriness are related to each other for most carbonaceous materials, whereas the latter is of greater importance in the context of this study. We will modify the text, also in response to comments by the first referee, to further clarify these aspects including additional references.

10) You cite so many thermal methods that are not really used. Hardly anybody in air pollution uses the actual NIOSH protocols, nor the Birch and Cary 96, while they are cited, people used variable timesteps or for the least longer time steps. Also the air pollution community hardly ever uses the same final temperature level than NIOSH. SO please clean this up for what is actually being used in the community (e.g. table 1)

We cited Birch and Cary 96 in the context of the introduction and basics of the NIOSH technique, but we acknowledge the reviewer's points here. The discussion of NIOSH protocols was motivated

by the fact that Fig. 6 includes a NIOSH-5040 based data point. However, in the interest of shortening the methods part, as requested above, we will shorten Table 1 and Section 2.2.2.

11) The whole discussion on "coarse" BC is misleading (late in the manuscript ~LK500). In the air quality/aerosol community, coarse tends to mean something very specific: particles between PM2.5 and PM10, sometimes particles larger than PM10 but never to my knowledge particles between PM1 and PM2.5 as it is used here. So or clearly define but better formulate this differently because it crates confusion given that you have PM10 and PM2.5 size cuts too.

The use of "coarse" and "fine" BC in Sect. 3.3.1 will be checked carefully in order to minimize potential confusion.