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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Black carbon physical properties and mixing state in the European megacity Paris" *by* M. Laborde et al.

J. SCIARE (Referee)

jean.sciare@lsce.ipsl.fr

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General comments:

Based on a large instrumental setting, this paper aims to investigate the optical and mixing state of refractory black carbon (rBC) during wintertime in the region of Paris. Among the major conclusions of this study, it was shown that rBC was moderately coated (literally no coating for freshly emitted rBC from traffic). SP2 measurements were used to investigate rBC from traffic and biomass burning and showed that biomass burning had a minor contribution to the total number of rBC particles, a medium coating thickness and showed slightly more-hygroscopic properties. By contrast, aged/continental rBC showed substantial coating and consequently larger mass absorption coefficient (MAC). These conclusions on rBC properties are crucial regard-

ing its major (but still poorly documented) impacts (climate and health). This paper is very well organized and written. A very important experimental effort has been put here to obtain a large set of quality controlled aerosol data which has been used to strengthen the general conclusions on refractory black carbon, its optical and mixing state properties. The only (minor) concern I have here is towards the proper use of the term Black Carbon (BC). The main conclusions of this paper rely on the results of the SP2 instrument which provides information on rBC which cannot be considered, to my opinion, as equivalent to EC or EBC. The authors should keep this point more often in their mind when they interpret their dataset. For that reason, I would ask the authors to keep the term "refractory black carbon" (rBC) in their paper and not use the term black carbon (BC). I would also like to see a more quantitative comparison between rBC (SP2), co-located EBC (Aethalometer with a MAC obtained for instance in Paris, Crippa et al., 2012), and co-located EC (obtained from the filter sampling).

Specific comments:

+ Line 3, page 25122: "one of the biggest European megacities". Maybe not completely exact. As far as I know there is only 2 megacities in Europe (London – the biggest one- and Paris). Should be more "one of the biggest European cities". Also, measurements were performed at a suburban site of Paris which site shows EBC concentrations lower compared to the center of Paris. This should be stated more clearly (in the paper and in the abstract).

+ Line 3, page 25122: Biomass burning rBC is found to poorly contribute to rBC. Please give a number (preferably expressed in μg/m3 which is the unit commonly used to refer to EBC or EC).

+ Line 1, page 25123: "boxdetectable". What is "boxdetectable" ?

+ Lines 7-10, page 25123: Please recall that this statement (rBC = poor CCN) is observed in urban environment. The fact that rBC makes poor CCN (i.e. weak impact on aerosol indirect effect) implies a longer lifetime for rBC (i.e. higher impact on aerosol

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direct effect). This is also to put in perspectives.

+ Line 13, page 25124: MAC is size dependent and is amplified by coatings of non-refactory matter. This is true. For that reason, it is also critically dependent on Relative Humidity (which will modify the coating and consequently the absorption properties). You may also mention this point.

+ Lines 15-25, page 25124: Reviewing the techniques investigating the mixing state of refractory aerosols, you may also mention some studies on VHTDMA.

+ Lines 15-25, page 25125: characterized or characterized. Should be the same everywhere in the paper. Please correct accordingly.

+ Line 3, page 25126: "... holds a quarter of France's population". It is closer to 20%.

+ Line 5, page 25126: You can also add Sciare et al. (JGR, 2011) which have performed a source apportionment of EBC from traffic and wood burning at LSCE (close to SIRTA) and at the same period of the year.

+ Line 5, page 25126: "in the agglomeration of Paris" instead of "in Paris" since measurements were performed at SIRTA, a suburban site located at 15km south-west from the center of Paris.

+ Line 1, page 25127: As mentioned before, I would like the authors to keep the term "rBC" along the manuscript and not make a short cut using "BC". Note that this point (use of the term "rBC" for SP2 measurements) is one of the recommendations of the Scientific Advisory Group – Aerosol of the Global Atmospheric Watch network.

+ Line 6, page 25127: Limit of quantification (LOQ) is given as a range with one order of magnitude. Can you give something more precise here? How could be translated this LOQ in μg/m3 ? I note here that the range of detection is ranging from 80 to 500nm. I believe this may represent some limitations investigating the real quantity of rBC for aged aerosols? I have here in my mind the paper from Healy et al. (2011) obtained at the same time (Paris, MEGAPOLI winter campaign) with ATOFMS size distribution of

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the EC class “ECOCNOx” which is showing a maximum around 600nm). With the only objective to have a consistent picture of black carbon properties in Paris, the author should also mention this limitation here.

+ Line 3, page 25128: Data analysis and uncertainties. I am clearly not a specialist of the SP2 instrument but I am concerned on the use of the temperature (4000K) to vaporize BC. It is stated here that this method is unbiased by the presence of non-refractory matter. Right. Besides, many papers have investigated the thermal properties of black carbon using thermal and/or thermo-optical methods. I have in my mind that biomass burning black carbon is less resistant to thermal treatment than fossil fuel black carbon. I recall some earlier study (Sciare et al., ACP, 2003) dealing with this. Also the presence of potassium in aerosols may oxidize BC particles, and thus decrease their temperature of combustion (Novakov and Corrigan, 1995). Martins et al. (1998) reached the same conclusion. There is probably some more recent studies dealing with this point. Anyway, I believe that a non negligible amount of absorbing material could have vaporized before the temperature defined threshold of 4000K given for rBC. Missing a part of wood burning black carbon could have explained the “high” MAC found at 550nm (see later on). It could have explained the relatively low contribution of wood combustion BC compared to other studies performed in Paris? (Sciare et al., 2011; Healy et al., 2012). I would like the authors to comment more on that (either in the response to the reviewers or in their paper). Is there any paper dealing with this thermal issue in the SP2 for biomass burning aerosols? Also the flaming mode of biomass burning is also leading to a clear enhancement of EC relatively to OC. Could the SP2 be sensitive to this fraction of biomass burning instead of the smoldering aerosols and detect rBC originating from this flaming mode? Then, I would not be surprised that we find a moderate coating on it (given the high EC/OC ratio reported in literature for flaming conditions).

+ Line 19, page 25131: I am not sure that you have ran a AE-7 model for the Aethalometer. I would either say AE-30, AE-31 or AE-33 ?

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+ Line 26, page 25131: PSI = acronym not introduced before in the text?

+ Line 20, page 25132: I guess the grid resolution for ECMWF is "0.18°" and not "0.18°C" (idem for 1°C).

+ line 21, page 25132: SIRTA = acronym not introduced before in the text?

+ Line 11-20, page 25135: Be careful. You are comparing rBC concentrations obtained at a suburban site of Paris with other datasets obtained using different techniques (Thermo-optical? optical?) and obtained within big cities which is not the case here (suburban site). Instead and still with the only objective to provide to the reader a consistent view of black carbon concentration levels in Paris, I would have appreciated a discussion with co-located EC data (thermo-optical method) obtained by the LCP group (N. Marchand), co-located EBC from Aethalometer data, EC from ATOFMS obtained at LHVP, and previous EC measurements reported for Paris (Sciare et al., 2010; 2011; and references therein).

+ line 16, page 25137: I would have said Fig. 6b instead of Fig. 6a.

+ line 29, page 25137: "The use of the T/B ratio can provide insight into sources of pollution as well as the photochemical age of the air mass . . ." . . .having anthropogenic influence !

+ Line 5, page 25139: The use of AAC is telling us the relative contribution of one source to the other. AAC around 2 tells you that biomass burning absorbing aerosols are dominating. It does not tell you that you are significantly impacted by high concentrations of biomass burning absorbing aerosols (an AAC of 2 can be obtained for very low concentrations of biomass burning absorbing aerosols). To make sure to isolate periods with real influence of biomass burning absorbing aerosols, I would encourage the author to check whether BC wood burning (from the Aethalometer model; Crippa et al., 2012) is consistent with the biomass periods defined in the manuscript using AAC.

+ Source apportionment of BC from traffic & biomass burning. The authors have

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decided to work with AMS organic data to estimate the relative contribution of the 2 sources. It would be more legitimate to perform this source apportionment directly with EBC mass concentrations instead of non refractory OM derived from the AMS. This could be done using the “Aethalometer model” to derive BC fossil fuel and BC biomass burning. Then, what would the final result? (compared to the AMS approach).

+ page 25142, line 11: It would say more “indicate dominating influence” instead of “indicate influence”

+ page 25142, line 23: Could HOA contain also COA that would explain the delay observed in HOA the evening ?

+ page 25144, line 6 and 14. The second period with continental/aged air masses is 7-15 Feb. or 7-9 Feb ?

+ Section 3.3.2.: Still with the only objective to provide to the reader a consistent view of black carbon coatings over the region of Paris, it would be very nice to compare (in a more quantitative way) at specific diameters being common to SP2 and ATOFMS a raw estimate of the coating (mass concentration) observed with the 2 instruments. If this requires too much work, I would put at least this comparison in the perspectives of this study.

+ page 25146, line 20: Do not forget also that aged air masses are associated at SIRTA with downwind conditions from the city of Paris bringing fresh traffic emissions.

+ page 25147, lines 22-29: I suggest to remove this paragraph which should better go in the hygroscopicity section. Also I suggest removing the first sentence “BC is insoluble in water”.

+ page 25149, end of the MAC section: Several explanations are considered to explain the difference between the obtained MAC from SP2 measurements with the values commonly reported in the literature. First, I do believe that a first comparison should be done with MAC obtained using babs (aethelomater) and co-located EC. Comparison

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could be done also with the city center site (LHVP, Healy et al., 2012). Also, the authors should also consider that SP2 may not provide a fully quantitative estimate of BC.

+ page 25150, line 8: Is it “Fig. 12b,f” or “Fig. 12c,g” ?

+ page 25150, line 17: Regarding the good agreement obtained between LHVP and SIRTA for Na+ in PM2.5 (PILS-IC data, Sciare et al.), I don’t think that local emission from de-icing salts could explain the GF of 1.8-2.

+ page 25152, end of section 3.3. : When interpreting the aerosol hygroscopicity, keep in mind that the different periods (aged, continental) are also influenced by fresh local emissions.

+ page 25153: Again here, I consider that the authors are not cautious enough regarding the conclusions they get for biomass burning with SP2 measurements. I will feel better here when they will replace BC by rBC !

+ page 25154, line 12: should be better “one of the biggest European cities”.

+ page 25155: I am not sure that investigating freshly emitted rBC at a suburban site provides the best picture of the hygroscopicity of BC for global modeling. Aged/Continental air masses should be more considered instead. Also the authors should go a little bit further in their conclusions: decreasing the wet removal of BC would enhance its lifetime and increase its direct radiative forcing.

+ Figure 1: The SIRTA site (Ecole Polytechnique, Palaiseau) is located at 19km from the city center of Paris (and not 30km)

+ Figure 3: This is a very important Figure which should be vertically extended to $\frac{1}{2}$ page. Also I don’t find relevant to present rBC data in lognormal scale. It underlines the lowest concentrations and not the highest. The same for Fig. 5.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 25121, 2012.