

Author reply to Review by Jean Sciare

We thank Jean Sciare for his valuable comments which help clarifying and improving the manuscript.

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

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, 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 regarding 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).

This request was greeted, see below for more details

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).

This request was greeted, see below for more details

+ 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”.

This has been changed to “2nd biggest European city”

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).

This has been done in the abstract, introduction and Sect. 2.5.

+ Line 3, page 25122: Biomass burning rBC is found to poorly contribute to rBC. Please give a number (preferably expressed in $\mu\text{g}/\text{m}^3$ which is the unit commonly used to refer to EBC or EC).

This conclusion is based on the modelled BC concentration calculated with a bilinear linear regression (Sect. 3.1 and using Fig. 7), assuming BC was either originating from biomass burning or traffic. The results indicate a predominance of the traffic emissions with a contribution of 99.6% of the BC concentration while the contribution of biomass burning emission was found to be close to 0. We believe the method used here is a good indicator of the predominance of traffic emission however it is a rather simplistic method and deriving an actual concentration of biomass burning BC in Paris would be highly uncertain. This is already discussed in more detail in Sect. 3.1.

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

This was a typo. It has been changed to detectable.

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

This paragraph reads now:

“The dominant fraction of the BC-containing particles in the urban area was found to have no or very little coating with non-refractory matter. The lack of coatings is consistent with the observation that the BC particles are non- or slightly hygroscopic, which makes them poor cloud condensation nuclei. It can therefore be expected that wet removal through nucleation scavenging is inefficient for fresh BC particles in urban plumes. The mixing state specific cloud droplet activation behaviour of BC particles including the effects of atmospheric aging processes are important to be considered in global simulations of atmospheric BC, as the wet removal efficiency remains a major source of uncertainty in its life-cycle.”

+ Line 13, page 25124: MAC is size dependent and is amplified by coatings of nonrefractory 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.

This reads now:

“In addition, the mass absorption efficiency (MAC) of BC is size dependent and is amplified by coatings of non-refractory matter or water at high relative humidity,…”

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

We added:

“...or by the combined volatility and hygroscopicity TDMA (VHTDMA) technique (e.g. Johnson et al., 2005).”

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

Done

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

Changed to “about 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.

Done

+ 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.

Done

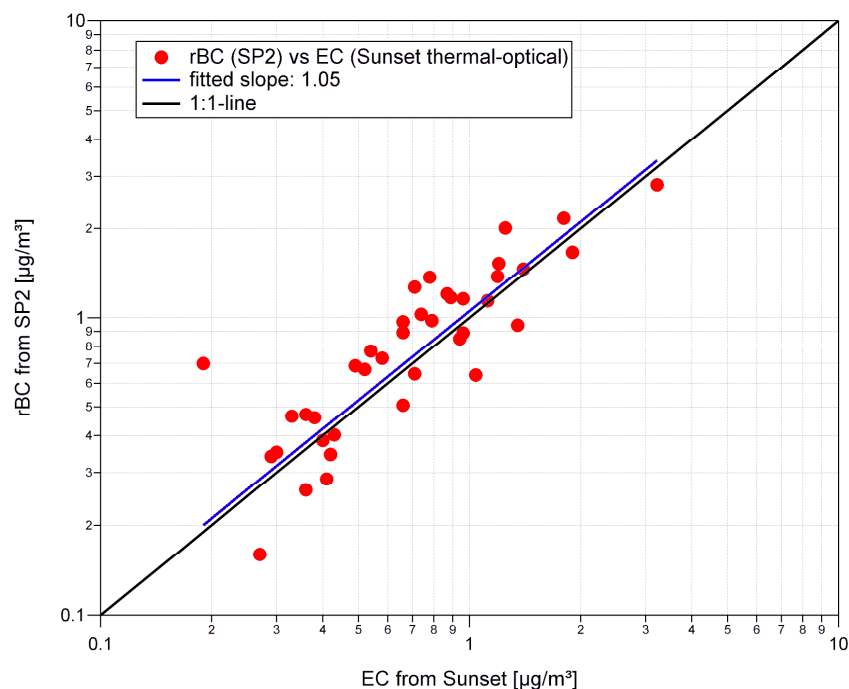
+ Line 1, page 25127: As mentioned before, I would like the authors to keep the term “rBC” along the manuscript and not make a short cut using “BC”. Note that this point (use of the term “rBC” for SP2 measurements) is one of the recommendations of the Scientific Advisory Group – Aerosol of the Global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

Done

+ 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 $\mu\text{g}/\text{m}^3$? 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 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.

The SP2 measures rBC mass concentration by quantitative measurement of BC in individual particles. Consequently it does not have a LOQ in the sense of rBC mass per volume of air (if the instrument does not have a leak). The detection range stated in the manuscript indicates the minimal and maximal amount of rBC in a single particle that can be quantified. Previous atmospheric measurements showed that the SP2's detection range covers the mass size distribution of BC well, except for potential contributions of a few BC particles with sizes in the micrometer range. Furthermore, the comparison of the rBC measurements by the SP2 with a collocated EC measurement by a thermal-optical EC measurement (Sunset ECOC analyzer behind a PM2.5 inlet) shown in below figure reveals excellent agreement between two methods, indicating that the SP2 covered the relevant part of the BC size distribution.



Healy et al. (2011) indeed reported BC particles with relatively large diameters. However, their sizing is based on the vacuum aerodynamic diameter of the full particle (BC core and coating), whereas the detection limit of the SP2 is expressed as BC core mass equivalent diameter, which is smaller for equal particles, particularly for coated BC particles in aged air mass. The discussion of these facts in Sect. 3.3.2 has been improved:

“Large BC-containing particles were also observed by Healy et al. (2012) for the continental air mass with a modal particle diameter of ~700–900 nm. However, the ATOFMS measures the vacuum aerodynamic diameter of the whole particle (BC with coating), as opposed to the SP2 which determines the BC core mass equivalent diameter. The latter quantity is smaller for equal BC particles observed by either instrument, particularly for thickly coated BC particles. The increased size of BC particles reported by Healy et al. (2012) during the continental air mass is for a good part driven by the thickest non-refractory coatings observed by the SP2 during this period (Sect. 3.1 and Sect. 3.3.3, Fig. 10a, b).”

+ 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).

The measurement principle of the SP2 is fundamentally different from thermal-optical methods and therefore the different temperature values cannot be compared. A detailed discussion of the influence of thermal reactivity of BC on its detection by the SP2 is available in the interactive discussion of a paper by Gysel et al. (2012), particularly in the following author reply:

<http://www.atmos-meas-tech-discuss.net/5/C3035/2012/amtd-5-C3035-2012-supplement.pdf>

Briefly, the time scales (in the order of μs) in the SP2 are too small for a substantial bias in the measurement of BC with different thermal reactivity (from different sources). Nevertheless, it is known that the chemical structure of the BC still has some influence on the sensitivity of the SP2 to BC mass through other effects. However, Laborde et al. (2012) have shown that the sensitivity difference of the SP2 to BC from diesel exhaust and from a log wood stove is small. Therefore, it can be expected that the SP2 is also able to reliably detect BC from biomass burning too.

The MAC value obtained in this study is actually very close to the MAC value determined for BC in Paris by Sciare et al. (2011) using a combination of aethalometer (absorption) and Sunset EC/OC analyzer (EC mass), while discrepancies to other studies remain. The discussion of the MAC values in the context of existing literature (Sect. 3.3.4) has been modified in response to this and other referee comments:

“The average MAC of the entire dataset is $\sim 8.6 \text{ m}^2 \text{ g}^{-1}$ at 880 nm (Fig. 11C 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 $\text{AAC} = 1$). 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.”

The source apportionment of atmospheric BC to its sources remains a very difficult business. Current methods such as the radiocarbon approach, the aethalometer model, and the convolution of ATOFMS or SP2 measurements with other data sets all suffer from substantial experimental deficiencies or crude assumptions. The differences in the estimated contribution of biomass burning to BC in Paris between the two methods applied in this study and the results reported in Sciare et al. (2011) and

Healy et al. (2012) are well within the uncertainty of the methods. The common finding of all is that the traffic gives the major contribution while biomass burning only gives a minor contribution. Any interpretation beyond this would be speculative. These facts are already discussed at the end of Sect. 3.1.

Gysel, M., Laborde, M., Mensah, A. A., Corbin, J. C., Keller, A., Kim, J., Petzold, A., and Sierau, B.: Technical Note: The single particle soot photometer fails to detect PALAS soot nanoparticles. *Atmos. Meas. Tech.*, **5**, 3099-3107, doi:10.5194/amt-5-3099-2012, 2012.

Laborde, M., Mertes, P., Zieger, P., Dommen, J., Baltensperger, U., and Gysel, M.: Sensitivity of the Single Particle Soot Photometer to different black carbon types. *Atmos. Meas. Tech.*, **5**, 1031-1043, doi:10.5194/amt-5-1031-2012, 2012.

+ 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 ?

Changed

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

Done

+ Line 20, page 25132: I guess the grid resolution for ECMWF is “0.18_” and not “0.18_C” (idem for 1_C).

Done

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

Done

+ 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).

This paragraph aims at comparing the measurements from SIRTA with other measurements in European cities. The comparison with other measurements in Paris is of course also interesting and has been added:

“The BC mass concentrations observed in this study are similar to previously reported values in Paris ($\sim 1.0 \mu\text{g m}^{-3}$; Sciare et al., 2011) and in various European highly populated cities, although different methods were used, i.e. $3 \mu\text{gm}^{-3}$ in Milano (Invernizzi et al., 2011), $\sim 1.7 \mu\text{gm}^{-3}$ ”

The rBC measurement cannot be directly compared with the aethalometer data because the latter only measures light absorption. Instead, the light absorption and rBC data are used in Sect. 3.3.4 to estimate the mass absorption coefficient. The rBC is also not compared with ATOFMS results because the latter is not quantitative.

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

Not changed as we really mean Fig. 6a. This paragraph aims at discussing the temporal variation of the particles growth factor. The contour plot given in Fig 6a is therefore more appropriate to do so.

+ 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 !

This point was added to the text.

+ 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.

We totally agree with the interpretation of the AAC and clarified the statement in Sect. 3.1 accordingly:

“The observed variability of the AAC is shown in Fig. 3G. Values around 2 are typically associated with dominate influence from biomass burning emissions while values around 1 are associated with dominant influence from traffic emissions (Sandradewi et al., 2008).”

In Sect. 3.3.1 we state:

“A strong influence of biomass burning was identified using the PMF results. The time periods when the BBOA mass concentration was accounting for more than 30% of the total organic mass measured by the HR-ToF-AMS were selected as “biomass burning influenced” (brown bars in Fig. 3B, D, F). These periods coincide with AAC values close to 2, which confirm strong biomass burning influence”

And in Sect. 3.2 we state (slightly adapted):

“The AAC shown in Fig. 8F - values close to 1 and 2 indicate dominating influence from traffic and biomass burning emissions, respectively - and the BBOA mass concentration shown in Fig. 8G follow a similar diurnal pattern with low values during daytime, high values during night-time and a maximum at ~10–11 p.m. LT. This pattern, which is completely different from that of traffic emissions, can most likely be attributed to a peak of domestic heating activities in the late evening hours. The number fraction of slightly hygroscopic particles ($1.1 \leq GF \leq 1.2$) also follows a similar pattern as the AAC and the BBOA, thus indicating a link between biomass burning and the emission of slightly hygroscopic aerosol which will be corroborated in Sect. 3.3.5.”

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

In the reply to an above comment we already argued that all approaches for the source apportionment of atmospheric BC have their own limitations. The discussion of result from different approaches applied during the MEGAPOLI campaign is already discussed in the following paragraph:

“...The traffic and biomass burning emissions were found to account for $Q_{\text{traff}} = 99.6\%$ and $Q_{\text{BB}} = 0.4\%$ of the total BC mass according to the simple bilinear regression model. This result is most likely biased to traffic emissions, as biomass burning is expected to give some minor contribution to BC mass, too. The relative contribution of the fragment m/z 60, a marker for biomass burning aerosol, was actually somewhat higher in the HOA factor retrieved from this data set compared to other data sets. This gives evidence that the HOA factor contains a minor biomass burning contribution, which would explain a small bias of the regression model to traffic emissions. Nevertheless, the above result indicates that traffic is the dominant source of BC in Paris, which is qualitatively consistent with 74% traffic contribution, determined by Crippa et al. (2013) for the same location and time period using the observed AAC value for the source apportionment following the approach by Sandradewi et al. (2008), and with 88% traffic contribution, determined by Healy et al. (2012) for the city centre of Paris during the same time period using data from an aerosol time-of-flight mass spectrometer (ATOFMS)...”

The analysis of diurnal patterns gives further evidence of dominating traffic influence (see Sect. 3.2):

“...The comparison of the diurnal patterns of BC mass concentration with traffic and biomass burning markers provides clear evidence that traffic emissions give the dominant contribution to BC mass concentrations and is consistent with the result of the bilinear regression model (Sect. 3.1). However, the evening peak of HOA is somewhat delayed compared to the evening peak of BC and it is close to the BBOA peak. This could indicate that the HOA factor contains some minor contribution from biomass burning, which would explain the over- and underestimation of the traffic and biomass burning contributions, respectively, by the bilinear regression model (Sect. 3.1)...”

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

Done

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

The COA and BBOA factors identified at the SIRTA have similar diurnal patterns, and it cannot be excluded that the HOA factor also contains some COA contribution. A characteristic feature of the typical COA mass spectra is a substantially higher m/z 55 signal than m/z 57 signal, while these two fragments are about equal in the typical HOA mass spectra. The HOA mass spectrum retrieved from the SIRTA site does not have an enhanced m/z 55 to m/z 57 ratio, thereby indicating that it does most likely not contain a substantial COA contribution. The m/z 60 signal, a characteristic fragment of BBOA, is enhanced in the HOA mass spectrum retrieved at SIRTA compared to typical HOA mass spectra, indicating a potential contribution from BBOA in the HOA factor. For these reasons we speculate that the delayed HOA evening peak is more likely due to a BBOA interference than a COA interference.

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

The second continental/aged air mass period is from 7 to 15 February. It has been changed.

+ 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.

The ATOFMS does provide valuable insight into the chemical composition of the coatings of BC particles, but it is, to our knowledge, not possible to quantitatively derive the coating thickness of BC particles from ATOFMS measurements.

+ 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.

Indeed, however both studies (Schwarz et al., 2008 and Shiraiwa et al., 2008) were also performed downstream of a city.

+ 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”.

This paragraph has been replaced with:

“The effect of non-refractory coatings on BC particles on their hygroscopic growth and CCN activation behaviour will be discussed in Sect. 3.4.”

Parts of the previous paragraph have been merged into Sect. 3.4.

+ 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

(aethelometer) and co-located EC. Comparison could be done also with the city center site (LHVP, Healy et al., 2012).

The collocated rBC (SP2) and EC (Sunset) measurements at the SIRTA site agree within 5% (see reply to above comment). Therefore, using EC instead of rBC would not affect the resulting MAC values. The discussion of the MAC values in the context of other studies has been expanded in response to a previous comment (see above).

Also, the authors should also consider that SP2 may not provide a fully quantitative estimate of BC. This point was answered earlier in this document.

+ page 25150, line 8: Is it “Fig. 12b,f” or “Fig. 12c,g” ?
Indeed, the text has been changed to refer to Fig. 12c, g.

+ page 25150, line 17: Regarding the good agreement obtained between LHVP and SIRTA for Na⁺ in PM_{2.5} (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.

The reference was added.

Regarding the de-icing salt hypothesis, we believe it is a possibility. We do not have further arguments at that stage apart from a few events. Since the current manuscript highlights the fact that it is only a hypothesis: “possibly indicating a short time period with influence from sea salt or de-icing salt.” we will keep it in the manuscript.

+ 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.

We try to make this point in the current manuscript page 25150, lines 4-7:

“The GF-PDFs of the aged and continental aerosols also contain, besides the most hygroscopic background mode, particles with GFs between 1–1.4 (Fig. 12e, f), which are most likely a mixture of particles from local and regional origin with varying atmospheric age.”

+ 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 !

So far there isn't evidence that the SP2 measurement should be substantially biased for BC from biomass burning (see also answer to previous comment above). Nevertheless we are now using rBC instead of BC throughout the manuscript.

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

+ 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.

We agree, a suburban site is not globally representative. However, it is the starting point of a major part of BC emissions. Understanding the initial BC properties as well as the transformation processes and time scales are some important aspects in achieving a global picture (out of many).

This paragraph reads now:

“The dominant fraction of the BC particles at the suburban site in Paris was found to be non- or slightly hygroscopic. Consequently they require a higher supersaturation for CCN activation compared to the majority of particles of equal dry size. Considering nucleation scavenging, it can therefore be expected that BC particles are enriched in the interstitial phase of liquid clouds, thereby decreasing their wet removal efficiency, increasing their lifetime and increasing the global BC burden and associated environmental impacts. This composition specific activation behaviour, as a function of atmospheric aging processes, is important to be considered in global simulations modelling atmospheric BC, as the wet scavenging efficiency remains a major source of uncertainty.”

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.

Done, see previous reply

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

This has been corrected throughout the manuscript.

+ Figure 3: This is a very important Figure which should be vertically extended to 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.

Fig. 3 is expanded to fill a full page. Both linear and logarithmic scales have their specific advantages. We prefer to use a logarithmic scale for the BC mass concentrations shown in Figs. 3 and 5.