Answer to referees

Higher absorption enhancement of black carbon in summer shown by two year measurements at the high-altitude mountain site of Pic du Midi Observatory in the French Pyrenees

We thank the three reviewers for evaluating the manuscript and providing us constructive and useful comments. Referees have common concerns which we addressed to the best of our possibilities:

- In order to better highlight the objectives and the main results of the paper (1) we modified the title, (2) clearly listed the scientific questions addressed in the introduction section, (3) modified the conclusion into a section "Summary and implications for climate models", (4) moved Figure 2 from the meteorological section to the Supplement, (5) moved Figure S7 and Figure S9 from the Supplement to the main text in Sections 3.4.1 and 3.4.2, respectively.
- We clarified and completed the section on aerosol optical properties by adding a more complete description of the observed parameters and by reworking the design of Figures 3 and 5.
- We added two elements in the Supplement describing the processing of the SP2 measurements: a Section S1 comparing BC mass concentrations obtained from our software on PYTHON and the PSI SP2 Toolkit running on IGOR and a Section S2 to explain the processing of the rBC core size distribution. A Section S5 about the discrimination of Free Tropospheric/ Planetary Boundary Layer conditions has been added in the Supplement.
- We changed the "BC" nomenclature to "rBC" to follow the recommendations by Petzold et al. (2013) for measurements performed by a SP2.

Please find below reviewer comments in black and our responses in blue. The line numbers in the responses refer to the new version of the paper.

Anonymous Referee #1

SUMMARY

The work of Tinorua et al. provides interesting dataset on black carbon properties at a high-mountain site in Europe. These sorts of data are rare and of great interest. The aim of the manuscript is to understand the variability of black carbon properties as function of season, dynamics of the boundary layer and wet removal. Although the current dataset might allow investigating these processes, the presentation and discussion of the results prevents the authors to clearly communicate their message. The language and nomenclature are often problematic to the understanding of the text. Which require a thoughtful revision. Often, the authors jump to conclusions very fast, without a proper description of the observed parameters and with a superficial use of references. As a consequence, the processes leading to the observed changes of rBC properties are often unclear. I suggest the authors to clarify their goals, reduce to a minimum the non-essential discussion and elaborate more in details their hypothesis. I also advise caution when discussing "photochemical processes" and "hygroscopicity", which cannot be investigated with the current dataset. In its current status, the manuscript is not suitable for publication. However, I invite Tinorua et co-authors

to consider the major comments and a multitude of specific, yet not minor, comments for resubmission after major changes.

MAJOR COMMENTS

First, the manuscript would benefit from a deep revision of the language, which often results non-scientific and approximative. The authors are also invited to revise the format of citations, acronyms, and units and the grammar. See specific comments.

Nomenclature is extremely important. The authors should make sure to provide the correct information especially when using abbreviations and acronyms. 1) When dealing with optical properties, it is essential to always declare the wavelength. This is not always done, in the text and especially in the figures. In some cases, it thus results difficult to understand at what wavelengths the measurements are performed. 2)Every soot-measuring technique is based on different properties of the aerosol; hence, instrument specific nomenclature must be used. Soot measured with SP2 should be named rBC (refractory black carbon). Soot measured with filter-based photometers should be named eBC (equivalent black carbon). Soot measured with thermal-optical method should be named EC (elemental carbon). This nomenclature is not applied to the data presented here and to the results of other works. Please revise all the nomenclature and resulting abbreviation following Petzold et al. (2013).

REPLY :A deep revision of the manuscript has been done in order to correct typographical and linguistic errors as best as possible. The wavelengths of each optical parameters have been added in the text and the figures. The acronym “BC” has been replaced by “rBC” throughout the entire manuscript when talking about BC measured by the SP2.

The SP2 offers the possibility to quantify the mixing state (PSD detector and time-lag) and “composition” (colour ratio) of rBC. Unfortunately, these analyses are not performed, although it might help understanding ageing process and absorption enhancement, influence of different sources and potentially wet removal. Could the author explain why mixing-state and colour-ratio were not presented in the manuscript?

REPLY : We agree with Reviewer #1 that a closure between the rBC coating and absorption enhancement would reinforce our interpretation. However, due to a technical issue on the low gain of the scattering channel of the SP2, we could not provide rBC mixing state.

Concerning the color ratio, its analysis provides information on the presence of iron oxyde contained in dust particles (Liu et al., 2018; Moteki et al., 2017; Yoshida et al., 2016). To our knowledge, this parameter can not be used to assess the rBC composition.

The discussion of results and its interpretation is often superficial. This is particularly true in Section 3.1 and 3.2, where a detailed variability of aerosol and BC properties is provided but not discussed with the appropriate literature context. The text reads like a list of numbers followed by a list of references, while the reasons causing the variability is often explained with short and generic sentences like “*It has been attributed to the seasonal variation of the continental boundary layer height, long-range transport events (e.g. Saharan dust outbreaks, coal burning from eastern Europe) and biomass burning both from forest fires in summer and domestic heating in winter.*” I suggest the author rethinking all the results section to improve their data interpretation and to set clear scientific objectives.

The figures based on time series are not particularly helpful. If the authors aim to discuss the seasonal variability it is advisable to use a longer time stamp (1 month or 2 weeks). In order to provide evidence of correlations between the various properties I also suggest using scatter plots.

REPLY: Section 3.2 has been completed to better describe the seasonal variability of aerosol optical properties and explain the cause of this variability.

The sentence quoted by the reviewer aimed to provide the reasons causing the variability mentioned in the literature. This has been reworded.

The time resolution of Figure 3 and 5 has been decrease to provide a better visualization of the seasonal variations. We preferred to use boxplot in Figures 7, 8 and 10 instead of scatterplots for clarity reasons due to the large amount of data.

SPECIFIC COMMENTS

L30: Merge the two statements, not clear what "this" refers to.

REPLY : The statements were merged as suggested the reviewer.

L36-37: please add a reference.

REPLY : The reference of Matsui et al., (2018) has been added.

L39-40: the definition is correct, but it is not described how Mac is measured. A short description of the methodology is needed since later on (L43) the instrumental influence is mentioned.

REPLY : The following sentence has been added in line 39-41 :

" The MAC_{BC} can be calculated either by dividing the measured absorption coefficient of BC by its mass concentration or by using Mie's Theory and the BC size distribution and coating thickness as input variables. "

L53: too many references, select the most relevant to deliver your message.

REPLY : We included numerous references to show the high quantity of studies highlighting the lensing effect, ie. the absorption enhancement of BC due to its coating.

L58: what it is meant with "multiplied by two"?

REPLY : The sentence has been replaced in lines 60-62 by:

" López-Moreno et al., (2014) have shown by running several regional climate models that the occurrence of winter warm events in the Spanish Pyrenees will gradually increase until 2080. This includes an increase in the number of warm days and nights and the number of snow/ice melting days at altitudes above 2000 m above sea level (asl). "

L66-76: part of this sub-paragraph can be moved into the methodology (ABL-Topoindex).

REPLY : We believe it is essential to mention in the introduction the topographic features influencing BC observations at the PDM site in order to introduce the specific scientific questions investigated in the paper.

Listing of the sections is not needed. I suggest rewriting the current paragraph focusing on the goals of your work.

A paragraph describing the main scientific questions of the paper was added at the end of the introduction:
"This paper aims to provide comprehensive picture of the seasonal and diurnal variability of rBC properties at PDM, and to explore the processes driving these properties. Specifically, in the indicated sections, the following questions are addressed:

1. What are the air mass transport pathways impacting PDM ?
2. What is the seasonal variability of aerosol optical properties and dominant aerosol types ? What is the specific contribution of rBC to aerosol absorption ?
3. How do the microphysical and optical properties of rBC vary on a seasonal and daily basis ?
4.What are the roles of wet deposition, source and transport pathway in driving rBC absorption ?"

L90: replace "sucked" with "sampled"

REPLY : This has been modified

L94: DMT is not based any longer in Boulder, but in Longmont

REPLY : This has been modified.

L93-113: Although being relatively tedious, nomenclature is important. BC measured via laser-induced incandesce technique is normally referred as rBC (refractory black carbon). I suggest reading Petzold et al. (2013) for more details. Considering this technicality, I also recommend the authors to replace "BC" with "rBC" in the text and in all abbreviations (M_{rBC},D_{rBC}, etc…) when referring to their or other SP2 measurements. BC can be used for more generic discussion in the introduction.

REPLY : The nomenclature has been modified according to Petzold et al. (2013). A text in the beginning of Section 2.2.2, lines 104-108 has been added to present the different nomenclatures relative to BC:

"BC can be measured by different methods which are based on different BC properties. Petzold et al. (2013) defined a specific nomenclature for BC according to the BC quantification method. Following the recommendation of the authors, BC quantified by laser-induced incandescence and thermal-optical analysis will be referred to as refractive black carbon (rBC) and elemental carbon (EC), respectively. More general discussion on BC without focusing on its measurement technique will be referred to as BC."

L100: out of curiosity, did the authors ever compared the results obtained with the Python code and the SP2 Toolkit?

A new Section S1 has been added in the supplement about the SP2 data processing :
"**Text S1: Data processing for retrieving SP2 mass concentrations**

The Paul Scherrer Institute's SP2 toolkit is a softwave developed using IGOR to provide quantitative analysis of rBC mass concentration. However, this software is not suitable to analyze large amounts of data. During the PDM campaign, more than 1.2 To of data has been recorded. Processing it with the PSI toolkit would be too much time-consuming. This is why we developed a

software on Python. The data analysis was validated by comparing our M_{rBC} to the one obtained by the SP2 toolkit.

Figure S1 presents a comparison between the M_{rBC} retrieved with our Python program in blue, and the M_{rBC} calculated with the PSI SP2 toolkit in red. The output M_{rBC} from the toolkit does not take into account the rBC mass fraction below and above the SP2 size detection range corresponding to 90 < D_{rBC} < 580 nm. Therefore M_{rBC} without correction of the missing mass fraction is presented here. Globally over the 3 days, the two processing yielded Mr_{BC} values in agreement taking into account the 14% of uncertainties on Mr_{BC} (shaded areas), with a mean M_{BC} of 101.1 ± 14.2 and 82.3 ± 11.5 ng m^{-3} for our method and the SP2 toolkit, respectively. The SP2 toolkit seems to generate more M_{rBC} peaks compared to our method, which smooths a bit more the time series. Such high peaks of M_{rBC} don't seem realistic, given the situation of the site (remote station, without the presence of local rBC sources). The values provided by the PSI toolkit may be more noisy that the Python software due to a different selection of invalid individual signals. This could include signals exhibiting the maximum of their incandescence peak completely off-centered in the detection window of the SP2, incorrect sample flowrate, or an underestimation of the baseline of the incandescence peak height was underestimated, leading to overestimated individual masses,…). These different possibilities have not been explored in detail."

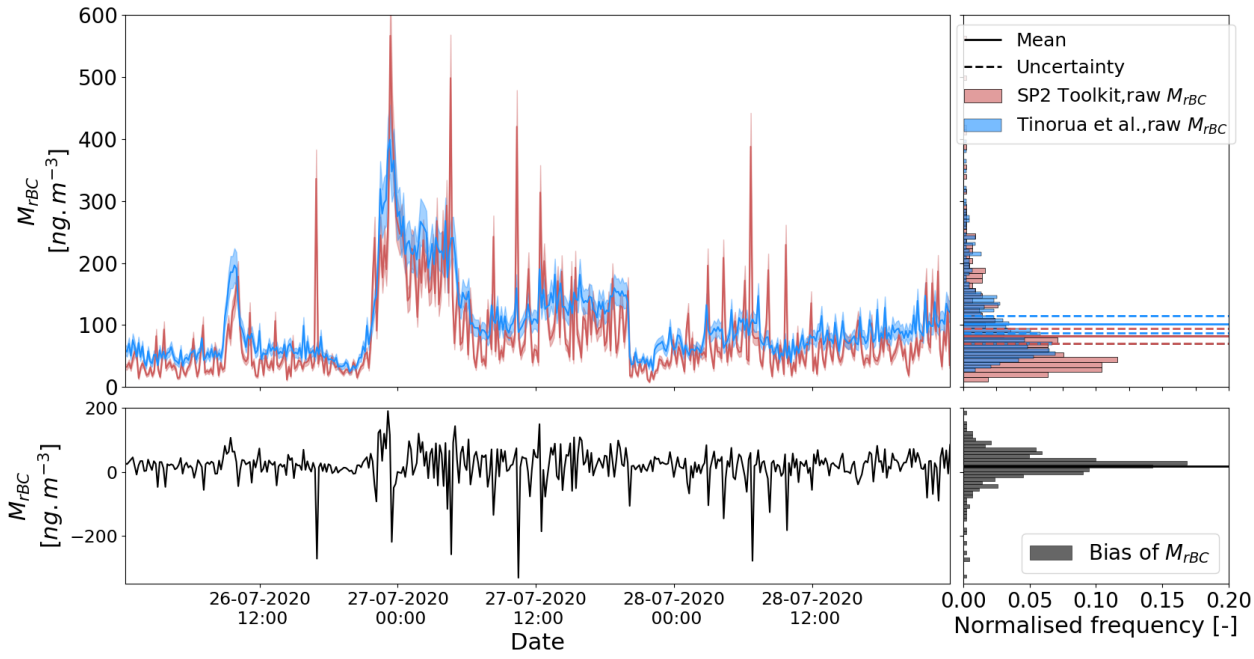

Figure S1: 72-h comparison between MBC calculated with the PSI SP2 toolkit and MBC calculated with the Python program used in this study. Data was 10-min averaged on the period from 26th to 28th July, 2020.The top left panel shows M_{BC} time series with the shaded area representing the M_{BC} uncertainties, and the associated histogram on the right-hand side with the median M_{BC} and its uncertainties represented by the solid and dashed lines, respectively. The bottom panel shows the bias (M_{BC} from our processing minus M_{BC} from the SP2 toolkit) and its associated histogram on the right-hand side. MBC data was here measured for rBC cores between 90 to 580 nm, without correction of the missing mass fraction.

L102: I do not see an increase of mass concentration at diameter smaller than 90 nm in Figure S1. Please reformulate or verify the top panel of Figure S1. Figure S1 shows both mass and number size distribution, but only mass is described.

REPLY : We agree that we did not provide sufficient elements to understand the issue. We have reformulated the sentence and added a Section S2 in the Supplement to describe the processing of rBC size distribution measured by the SP2 and the estimated missing mass fraction.

The sentence in lines 118-119 was revised as :

"However, the observed size distributions showed that an important fraction (around 12%) of M_{BC} at diameters below 90 nm is not measured by the SP2 (Figure S2 in the Supplement)"

The following text has been added to the Supplement :

"Text S2 : Information about the rBC size distribution processing

The SP2 measures rBC cores from mass equivalent diameters of 90 to 580 nm. Fig. S2 shows the two-year average of the daily rBC cores size distributions. It can be noticed in Fig. S2 that the number size distribution measured by the SP2 did not cover the full size range of rBC at PDM. This is particularly true for the rBC particles below 90 nm, where the major fraction of the M_{rBC} was missed by the SP2. In order to estimate the missing rBC mass fraction undetected by the SP2 (e.g. the mass size distribution under 90 nm and over 580 nm), the daily rBC size distributions were fitted with a sum of three lognormal functions as :

$$\frac{dM}{dln(D_p)} = \sum_{i=0}^{3} \left(\frac{M_i}{\sqrt{2\pi}\ln(\sigma_{g,i})} \exp\left[\frac{-\ln^2(D_p/D_{g,i})}{2\ln^2(\sigma_{g,i})} \right] \right)$$

with M_i, $D_{g,i}$ and $\sigma_{g,i}$ representing the rBC mass concentration, the geometric mean diameter and the geometric standard deviation of the mode i, respectively. The same function with two modes has been used to fit the number size distribution.

The fitting parameters were constrained in the following ranges : Mode 1 : $50 < D_{g,1} < 100$ nm and $1.2 < \sigma_{g,1} < 3$; Mode 2 : $150 < D_{g,2} < 250$ nm and $1.3 < \sigma_{g,2,} < 2.9$; Mode 3 : $350 < D_{g,3} < 500$ nm and $1 < \sigma_{g,3} < 3$. "

L103: "detection range", not "detection window".

REPLY : This has been modified.

L106-113: Please define what "dg" and "σg" mean. Assuming these are the geometric mean and geometric standard deviation, how these were defined, empirically? For mode 1. The SP2 lower size quantification limit was 90 nm. Does it mean that the lognormal fit is applied to the 90-100 nm diameter range to derive mode1?

REPLY : A definition of "D_g" and "σ_g" has been added in the text.

Please find details on the rBC size distribution processing in the previous answer.

Figure S1 shows the size distribution of rBC, but on what time scale? With what temporal resolution was the MBC-correction calculated? Would it change during different conditions (PBL, FT, winter, summer, etc…)?

REPLY : Figure S2 (following the new numbering) shows the two-year average rBC size distributions to illustrate the missing mass fraction of BC undetected by the SP2. However in the paper we used the daily average rBC size distribution to quantify the missing mass factor and correct the M_{rBC} measured by the SP2. The variations of the missing mass fraction with the season, dynamic conditions, etc. will be addressed in a paper in preparation for submission on AMT.

The relative standard deviation of the correction factor is approximately 90%, this lets me thing that non-negligible variability was observed during the measuring period. Could the authors have used a time dependent correction factor instead of constant one for the full dataset?

REPLY : Please refer to the previous answer. Lines 126-127 have been reformulated as follows:

"The average missing mass correction factor over the campaign was 1.2 ± 1.1 (Mean value ± STD)."

Considering the temporal variability of MBC-correction, I would like to see how MAC correlate with the correction factor.

REPLY : Figure R1 shows the MAC_{rBC} as a function of the M_{rBC} correction factor, noted $R_{fit/meas}$, equal to the ratio between the fitted mass concentration and the measured mass concentration. There is no particular correlation between the two.

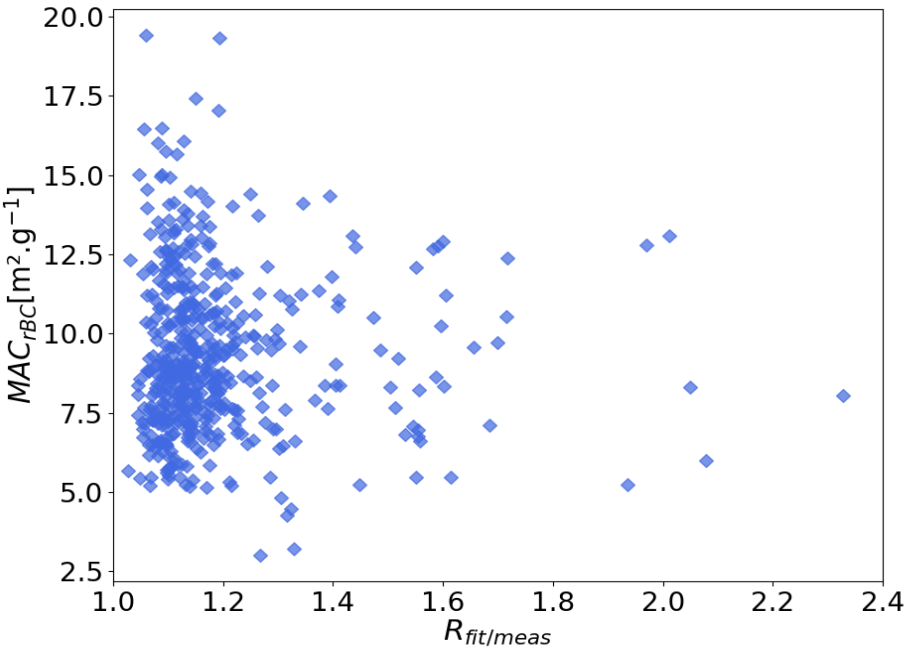

Figure 1: S8: MACBC as a function of $R_{fit/meas}$,core over the campaign. Each point represents 1 day average data.

L115: please provide the model, manufacturer, company, and country for the TSI instruments, as it is nicely done for the other instruments.

REPLY : The details for the TSI instruments have been added in the text.

L121: List the measuring wavelengths.

REPLY : This has been added.

L125: since is not yet published, the Cref value used in the present work should be described a bit better (location of the measurement, reference instrument, wavelength) and compared to previous studies. Since the manuscript is in preparation, and not submitted the year is not relevant.

REPLY : The following sentence has been added in Section 2.2.3, lines 144-146:

"The multiple scattering parameter used to correct the measured attenuation was set to 3.22, according to the value obtained at λ=880 nm by Yus-Díez et al. (2021) at the mountainous site of Montsec d'Ares located less than 200 km from the PDM ."

L126a: I strongly do not recommend the use of "MBC" for the BC mass concentration derived from the aethalometer data. First, the correct nomenclature should be equivalent black carbon (eBC; Petzold et al., 2013). Second, the mass concentration derived from SP2 measurements is also abbreviated as MBC. As a result, it become tremendously confusing to understand how MB is derived in the rest f the paper. Update the use of nomenclature.

REPLY: We changed the nomenclature used for aethalometer measurements by "mass concentration of equivalent black carbon"

L126b: Were the MeBC and σap limits corrected with Cref? At what wavelenght these values were derived, this is particoularly important (especially for σap). If I take 0.0215 Mm−1 and 0.005 μg m−3 I obtaine a MAC (or a mass attenuation coefficient) of 4.3 m2/g, please revise these values. And set the limit of AE33 based on absortion coefficient rather than MeBC, since you have a more reliable instrument (SP2) to measure the mass of rBC.

REPLY :

The corresponding line has been corrected in lines 147-148:

"The detection limit of the aethalometer is 0.039 Mm^{-1} (corresponding to an equivalent black carbon mass concentration of 0.005 μg m^{-3}). "

L130-132: when providing the information about the instruments try do be consistent with the rest of the paper and provide (model, manufacturer, company, and country), as done for the aerosol instruments

REPLY : These information have been added for all the instruments.

L133 I suggest removing ΔBC/ΔCO in this section, since it comes out of the blue without any context and it is anyway explained later in the text.

REPLY : $\Delta M_{rBC}/\Delta CO$ has been removed from this section as suggested by the reviewer.

L144: The authors should explain clearly that AAE was calculated between 450-635 nm to match the wavelength range of the Nephelometer. Since the measuring wavelengths of the Aethalometer are not listed, it becomes harder for the reader to understand why σap660 was adjusted to 635 nm.

REPLY : We agree that there were missing elements about the method used to calculate AAE between 450 and 635 nm. Therefore, we have added the measurement wavelengths of the aethalometer in Section 2.2.3 and we have completed the text in Section 2.3 as follows, lines 162-166:

"The spectral dependence of σ_{ap} was characterized by the Absorption Angstrom Exponent ($AAE_{aer,450-635}$) calculated between 450 and 635 nm as follows :

$$AAE_{aer,450-635} = \frac{-\log\left(\frac{\sigma_{ap,450}}{\sigma_{ap,635}}\right)}{\log\left(\frac{450}{635}\right)}$$

For this calculation, $\sigma_{ap,\,470}$ and $\sigma_{ap,\,660}$ from the aethalometer were adjusted at the wavelengths of 450 and 635 nm measured by the nephelometer using the AAE calculated from the aethalometer between 370-470 nm and 590-660 nm. "

L141-151: I believe a short explanation on what these optical properties represent is needed here. SSA, What SSA, AAE and SAE represent, why they are climatically relevant?

REPLY : We thank the reviewer for these suggestions and have added an explanatory sentence after formulas (1) and (2) to explain the meanings of AAE, SAE and SSA respectively :

l 166-170 : "$AAE_{aer,450-635}$, provides information about the chemical composition of atmospheric aerosols. Pure BC absorbs radiation across the whole solar spectrum with the same efficiency; thus, it is characterized by $AAE_{aer,450-635}$ around 1 (T. C. Bond et al., 2013). Conversely light-absorbing organic particles called as brown carbon (BrC) or dust particles typically have $AAE_{aer,450-635}$ greater than 2 (Bergstrom et al., 2007; Schuster et al., 2016; H. Sun et al., 2007).

L 178-180 : "$SAE_{aer,450-635}$ describes the relative contribution of fine and coarse mode particles (Clarke & Kapustin, 2010). Small values of $SAE_{aer,450-635}$ indicate a higher contribution of large aerosol particles (e.g. dust and sea salt) , while large values of SAE indicate relatively smaller aerosol particles (Cappa et al., 2016)."

L 183-184 : "$SSA_{aer,\lambda}$ describes the relative importance of scattering and absorption to the total light extinction. Thus, it indicates the potential of aerosols to cool or warm the atmosphere."

L155-157: I do not agree with the nomenclature choice. If ΔBC/ΔCO is the ratio of MrBC over ΔCO, it should be simply called MrBC/ΔCO, as done by previous studies cited in the result section (Liu et al., 2010; McMeeking et al., 2010).

REPLY : We understand the reviewer's point of view. The nomenclature of ΔBC/ΔCO has been changed to $\Delta M_{rBC}/\Delta CO$. We decided to keep the "Δ "because ΔM_{rBC} refers to the excess rBC mass concentrations, ie. above the background levels. In our particular case of a remote site without consequent rBC sources, the background levels of M_{rBC} are negligible and this why we can directly use M_{rBC} as ΔM_{rBC}.

L160: MBC under (resp. over) 160 the 5th (resp. 95th) percentile? Rephrase.

REPLY : The sentence in l. 196 has been corrected as follows :

"M_{rBC} below the 5^{th} and above the 95^{th} percentile were filtered before MAC_{rBC} calculations to reduce the influence of outliers in statistical analyses."

L161: I suggest giving more explanation about the influence of dust on absorption. Often, the authors do not provide adequate context to very specific statements, assuming that every reader has a deep knowledge of the treated topic.

REPLY : Some details has been added in Section 2.3, l. 197-199 :

"In addition, we filtered out periods when dust were sampled at PDM for the calculation of MAC_{rBC} since Yuz-Diez et al. (2021) observed significant biases in the multiple scattering correction of the aethalometer AE33 during such events."

L167-170. Provide some references for each method.

REPLY : The following references were added : Cappa et al. (2012), Healy et al. (2015), Shiraiwa et al. (2010) for the thermodenuder method, Cappa et al. (2019), Yuan et al. (2021) for the extrapolation of MAC_{BC} as a function of the BC coating mixing ratio, and Liu et al. (2017) and Zanatta et al. (2018) for Mie calculations.

L171: correct "1,95" in "1.95". Moreover, I strongly recommend reading Liu et al. (2020), who showed that, despite being widely used, 1.95-0.79i might not be representative of realistic condition. The authors are invited to verify the sensitivity of their calculated MACbare as function of different refractive index. As a matter of fact, Figure S3 showed a maximum MACbare below 5 m2/g which considerably lower than MAC of fresh and bare Bc presented by Bond (7.5m2/g). I imagine that Eabs presented here might be overestimated.

REPLY : The widely used $MAC_{bare,BC}$ value of 7.5 $m^2\ g^{-1}$ recommended by Bond & Bergstrom (2006) is provided at 550 nm. If we assume an AAE of pure BC equal to 1, this corresponds to a $MAC_{bare,BC}$ around 4.7 $m^2\ g^{-1}$ at 880 nm, which is in the range of the values shown in Fig. S3.

L183-195: for non-expert readers, this subsection might result of difficult understanding. Since the analysis is important, I suggested providing more details on how the ranking is calculated (more technical aspects could go in the supplementary). As it is, FigureS4 does not really help understanding the anabatic ranking, since zero context is provided in the supplementary.

REPLY: The text concerning the ranking method in the main body of the paper has been simplified for non-expert readers. In compensation, further detail on the ranking procedure, the determination of the threshold rank, and the interpretation of the "anabatic radon" diagnostic, is now given in the Supplement, as a text accompanying Fig.S5 (formerly Fig. S4). However the procedures are not described in full detail since this would be strictly redundant with the original description by Griffiths et al. (2014), which is given as reference.

Text part of the Section 2.4, l. 230-240:

"To discriminate FT and PBL-influenced air masses (hereafter referred as PBL/FT conditions), we followed a methodology proposed initially by Griffiths et al. (2014), assuming that the diurnal radon increase, which is typically observed at mountain sites during the daytime, is the result of transport of PBL air by thermal anabatic winds up to the summits. The method first consists in ranking the days of the sampling period by decreasing anabatic influence (details in the Supplement and in Griffiths et al. (2014)). A threshold rank (here 282, see Fig. S5 and associated text in the

Supplement) can then be determined to separate days with or without anabatic influence in the daytime.

In our study, it was necessary to select the observation hours strongly influenced by the boundary layer. To do this, we selected the first 200 anabatic days in the ranking, and from these days, all the hours with radon activity was greater than the median value for the current day.

We also needed an ensemble of observation hours with minimum influence of the PBL. In the latter case, we selected hours in the non-anabatic days (i.e. ranked after 282) with radon activity below the median value for the current day."

Text S5 in the Supplement:

"In the present study, the method used to discriminate anabatic vs. non-anabatic days follows the method by Griffiths et al. (2014) based on radon measurements, and the recognition that the anabatic influence can be measured by the amplitude of a diurnal radon cycle, properly phased with a maximum in the afternoon. The method mainly consists in ranking days by decreasing anabatic influence. All details of the ranking algorithm are given in Griffiths et al. (2014), but in overview (citing the authors) "the procedure involves computing the diurnal composite of the set of all observed days and then removing days from the set in the order which most quickly reduces the mean square amplitude of the set's composite diurnal cycle.

In our study, the procedure to compute the ranking strictly follows the steps described in Griffiths et al. (2014), except on this only point: as input data for the ranking procedure, these authors use the absolute deviation between the hourly radon data and the daily mean of the current day. In our case, we alternatively used the relative deviation (i.e. the absolute deviation normalized by the current-day mean). This considerably improved the result because the radon regional background at PDM is suspected to be much more variable than at the Jungfraujoch (see discussion below).

Then, a diagnostic value called "anabatic radon" is calculated for each day, which represents (in short) the average deviation of radon above a nocturnal background (see full detail of the calculation in Griffiths et al. (2014), which we again strictly followed). Anabatic radon mostly decreases with increasing anabatic rank (Fig. S5), at least up to a threshold rank (282) corresponding to its first minimum. Days ranked before this threshold are considered as anabatically-influenced (or more simply called "anabatic days"), and the days after this rank are considered as non-anabatic.

After this rank, anabatic radon values should expectedly be zero. This is obviously the case neither in Fig. S5, nor in the similar graph by Griffiths et al. (2014 – their Fig.3). The reason is that intraday radon variations due to any reason but anabatic transport, may occur out of phase with the thermally-driven cycle. Because of incoherent phasing, such variations contribute little to the set's composite diurnal cycle, and as a consequence, such days appear far in the ranking. But such variations may nevertheless be above the background (i.e. the minimum value) of the current day, and produce non negligible values of the "anabatic radon" diagnostic – which has thus little sense for non anabatic days.

A question arises, however, why anabatic radon appear more noisy in our Fig. S5 than in the similar graph in Griffiths et al. (their Fig.3). We have no definitive explanation to this, but may speculate that radon sources at the regional scale around Pic du Midi are more heterogeneous and intense than around the Jungfraujoch. Supporting this idea are radon exhalation maps by soils presented in

(Karstens et al. (2015) or (Quérel et al. (2022), showing radon hot spots in the western Iberian peninsula, in the French Massif Central, and (to a lesser extent) locally in the Pyrenees. In such conditions, the radon background at Pic du Midi may be much more variable than at JFJ, and other transport processes than anabatic transport may thus contribute more strongly to radon variability at PDM. This would deserve specific investigation, but is out of the scope of the present study."

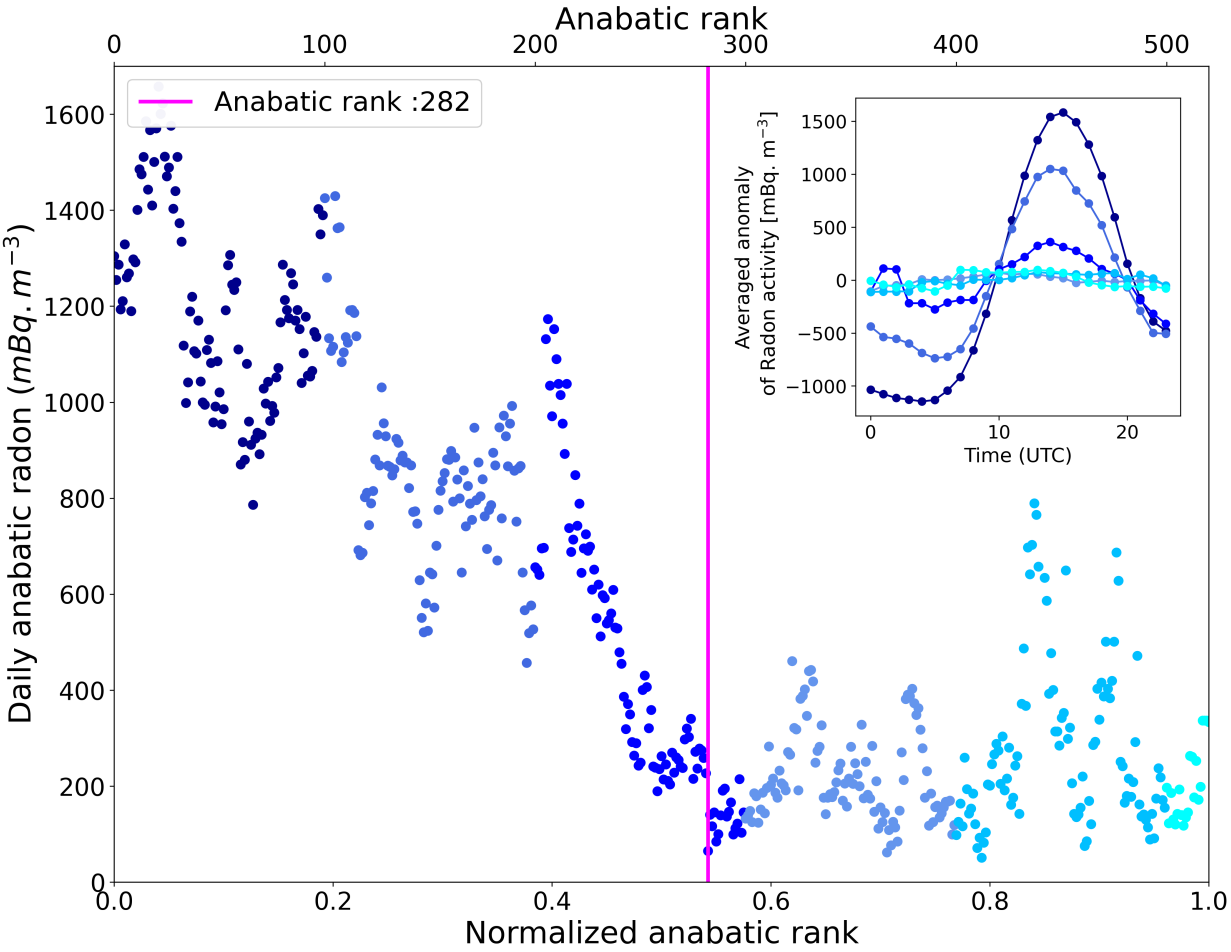

Figure S5: Daily anabatic radon as a function of the day anabatic rank (see text for details). Each dot represent an observation day ranked from the most anabatically-influenced day (left) to the least one (right). The vertical pink line represents the cut-off rank before which days can be considered as PBL-influenced. The insert shows diurnal composites of radon activity anomaly according to different ranges of ranks, using the same color code as in the main plot.

L191: I find the note particularly disturbing. Please avoid statements like "make no sense".

REPLY: This note actually brought little information but was confusing. It has been removed in the revised manuscript.

The fluctuation after rank 282 are not negligible and more noisy than in Griffiths et al. (2014). Please try to argue what might be the natural causes leading to the radon fluctuation. Could it be that these values are false negatives? Could the radon ranking be verified as function of water vapour as done in Griffiths et al. (2014)?

REPLY: A discussion on this specific point has been added in the text in the Supplement accompanying Fig. S5. This method may unlikely produce false negatives, because radon variations in phase with the thermal cycle should positively contribute to the amplitude of the set's composite

cycle. Such days should thus fall in the upper part of the ranking. But for the same reason the method is subject to false positives, when radon variation caused by any reason but anabatic transport fall in phase with the thermal cycle.

The ranking method was applied at PDM with water vapor by Hulin et al. (2019), in absence of radon data for their studied period. They obtained results (their Fig. 8) that were similar to ours in Fig. S5, in term of both (normalized) threshold rank and shape of the anabatic composite cycles. But radon at PDM has less seasonal and day-to-day variability than water vapor, and is thus less ambiguous as BL tracer than water vapor.. Griffiths et al. also pointed that the algorithm applied to water vapor is more subject to false positives.

L192-195: I am not sure to properly understand this final selection. The periods under the influence of PBL presented later are based on hourly selection and not daily selection (for ranking below 200 in the "anabatic-subset"), right? The opposite was done for FT influence. I expect the PBL-periods to occur preferentially during day-time, while FT-periods during night-time. Is the analysis only considering day-time or it does include also night-time?

REPLY : The text on the selection of either BL- or FT-influenced observation hours has been rephrased and, we hope, clarified. In summary, we made two distinct hourly selections among two previous selections of either anabatic of non-anabatic days. As BL-influenced hours are defined as hours with radon above the median of the current day among a selection of most anabatic days, these hours will clearly fall in the daytime. In contrary, FT-influenced hours may fall either during the day of the night. This is illustrated by Fig. R2.

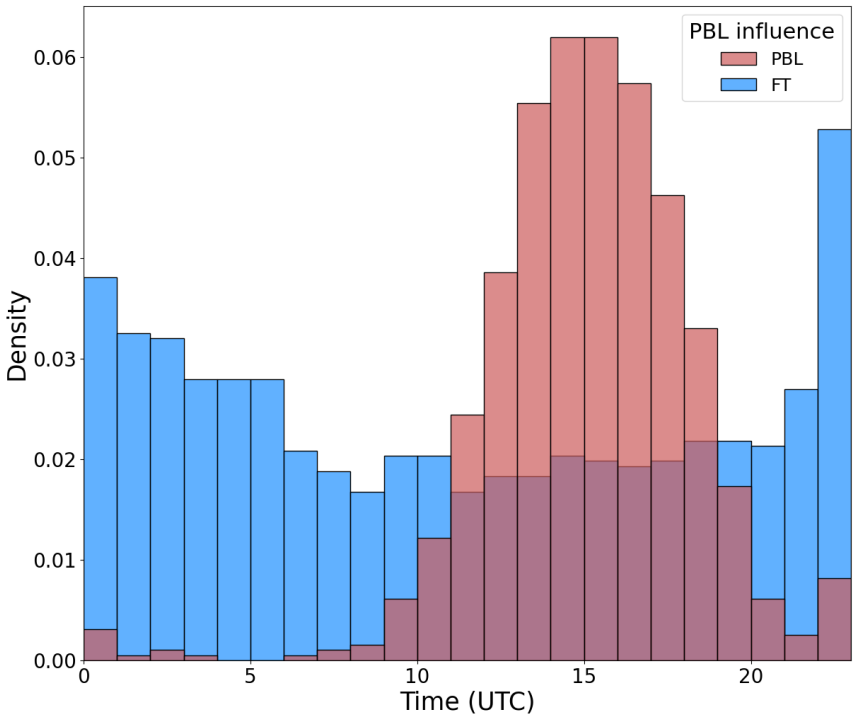

Figure 2: Density of FT/PBL classified hours

L202: m.s−1. Remove the dot.

REPLY : This has been modified.

L223-230: SSA at what wavelength? In figure 4a there are values well below 0.93. Is a monthly minimum, a season minimum? Please explain better. The simultaneous increase of SAE and absorption does not automatically indicate that absorbing particles are small in size. It must be kept in mind that BC is co-emitted with other fine aerosol species such as sulfate. I fund however interesting that the maximum peak of absorption does not correspond with a minimum of SSA. The reasons beyond the seasonal variability are actually not explained ("It has been attributed to the seasonal variation of the continental boundary layer height, long-range transport events and biomass burning both from forest fires in summer and domestic heating in winter." is a very generic statement).

REPLY : We agree with these very useful remarks which helped us for the interpretation of the aerosols optical properties. We have modified Figure 3 in order to better highlight the monthly variation of aerosol optical properties. In addition the scattering coefficient measured at 450, 525 and 635 nm have been added to Figure 3 in help in the interpretation of the SSA variations.

The text has been changed as follows, lines 271-286:

"There was a clear seasonality of aerosol optical properties. SSA at the three wavelengths exhibited the lowest monthly mean values in spring-summer (0.94 ± 0.02 at λ = 525 nm) and the highest in autumn-winter (0.99 ± 0.01 at λ = 525 nm, as shown in Fig. 3a). Simultaneously, the highest monthly mean SAE values were observed in spring-summer (1.23 ± 0.70) and reached a minimum in the winter (-0.25 ± 0.16) (Fig. 3b). This anticorrelation suggests a higher fraction of absorbing and fine particles relative to purely scattering and coarse particles at PDM during the spring-summer. Interestingly different trends can be observed between the summer and spring seasons. During spring 2019 the decrease of SSA correlated with a slight enhancement of $\sigma_{ap,880}$ (Fig. 3d) and decrease of σ_{sca} at all wavelengths. In summer the increase of $\sigma_{ap,880}$ lead to values multiplied by a factor of four, while both SSA and SAE remained rather constant. All these parameters combined indicate a similar dominant aerosol type reaching PDM but with stronger contribution in summer. This is further confirmed by the simultaneous increase of M_{rBC} in summer shown in Fig. 5.

This noteworthy seasonality of aerosol optical properties has previously been observed at other high mountain sites in Europe (Andrews et al., 2011; Collaud Coen et al., 2011; Laj et al., 2020; Pandolfi et al., 2018). The higher concentration of small and absorbing particles in summer at PDM could be attributed to a higher anthropogenic BC influence favored by strong vertical mixing and a higher PBL height, a higher occurrence of wildfires emitting large amounts of BC and Brown Carbon (BrC), or a lower precipitation rate.

In order to investigate these different hypotheses, a classification of the dominant aerosol type sampled at PDM was performed by using the spectral dependency of aerosol optical properties."

L259: check reference format

REPLY : This has been modified.

L260: why "MBCs"?

REPLY : This has been modified.

L260: Jungfraujoch name.

REPLY : It is a typo error and this has been modified.

L260-265: The seasonal variability of BC mass and absorption is opposite to background and polluted stations, where higher values are observed during winter compared to summer (among others: Yttri et al., 2007; Zanatta et al., 2016). The authors should explain this difference and potentially exploit it to introduce the analysis performed in the following sections of their works.

REPLY : In several mountainous sites of Jungfraujoch, Zugspitze-Schneefernerhaus (ZSF), the patterns of BC mass and absorption also shows minimums during winter and maximum during summer (Motos et al., 2020; Sun et al., 2021). The elevated sites presented by Zanatta et al. (2016) also exhibited maximum BC concentration and absorption during spring or summer.

The paragraph comparing M_{BC} and MAC_{rBC} has been modified and completed in Section 3.3, l. 359-379:

"The ambient MAC_{rBC} was around 9.2 ± 3.7 m^2 g^{-1} at λ=880 nm (Fig. 5d). Several studies previously reported MAC_{rBC} values between 8.9 to 13.1 for measurements at λ=637 nm in European mountain stations (Pandolfi et al., 2014; Yus-Díez et al., 2022; Zanatta et al., 2016). By using a AAE of unity, these values can be converted to MAC_{rBC} between 6.4 and 9.5 m^2 g^{-1}at λ=880 nm. These studies used different measurement techniques, analysis method and correction factors from ours for estimating MAC_{BC} that makes difficult the comparison of MAC_{rBC} derived from different instruments. Pandolfi et al. (2014) performed a linear regression between $\sigma_{ap,637}$ values measured by a Multi-Angle Absorption Photometer (MAAP) and daily M_{EC} values from off-line filter-based measurements by a SUNSET OCEC Analyzer. Yus-Díez et al. (2022) and Zanatta et al. (2016) retrieved MAC_{BC} with these instruments by calculating the ratio between the two parameters instead of a linear regression. Because of the absence of a standard method for quantifying M_{BC}, the absolute uncertainties on the MAC_{BC} obtained in the literature are very high ranging from ±30 to 70% (Zanatta et al., 2016).

In terms of seasonality we found systematically higher values of MAC_{rBC} in summer (monthly mean ± STD of 10.3± 3.3) compared to winter (8.3 ± 3.8). Similar seasonal pattern was observed in Europe at Puy de Dôme (central France) and at Jungfraujoch (Swiss Alps) mountain sites (Motos et al., 2020; J. Sun et al., 2021; Zanatta et al., 2016). An opposite trend was observed at mountain sites affected by strong precipitation during monsoon such as the Tibetan Plateau and Himalayas regions where both MAC_{BC} and M_{BC} exhibit maximum values in winter or autumn (Srivastava et al., 2022; Zhao et al., 2017). The same seasonal pattern with elevated values in winter/autumn compared to summer/spring was observed at several rural and urban sites in the PBL, which was attributed to greater emissions from residential heating combined to a lower PBL height (Kanaya et al., 2016; Yttri et al., 2007; Zanatta et al., 2016). However, maximum of MAC_{BC} and M_{BC} concentration in the PBL during cold periods is not a recurring observation even for a same measurement site. For instance J. Sun et al. (2022) showed in Beijing (China) that, due to the reduction of some predominant BC sources in winter consecutive to environmental policies, the annual cycle of M_{BC} changed over the years between 2012 and 2020."

L268: "Seasonal differences between the origin of highest MBC are thrown into relief,"…not sure what it is meant here.

REPLY : This sentence has been deleted.

L273: discussion discussed. Avoid repetitions.

REPLY : The sentence was changed and now reads:

« Further discussion on the role of PBL influence on M_{BC} will be addressed in Section 3.4.2. »

L268-272: From my point of view, Figure 7 shows that 1) the wind patterns are similar in winter and summer; 2) high MBC are associated with low wind speed; 3) and that there is a north scarred signal in winter and southern signal in summer. With the MBC scale and so many points, I cannot identify any clear correlation between wind direction and BC concentration, so I do not agree with the statement "highlighting different BC geographical sources" Similar reasoning can be done for Figure S5, where the overall origin of the air masses lays in the same western sector in both seasons. To improve the visualization and interpretation of the data, I suggest organizing the wind direction in broader classes (10-20 degrees) and normalize the MBC to its maximum. This modification might help identify a correlation between wind direction and BC concentration

REPLY : We are very grateful to the reviewer for his suggestion concerning the polar graph. Instead of plotting the 1-hour data on the windrose, we partitioned the windrose and calculated the density of the M_{rBC} data for every wind speed-direction 'bin', weighted by the M_{rBC} values. Thus, the color of each point represents the density of M_{rBC} measurements and their values.

We added a description of the representation and modified our interpretation in l. 322-332 as follows :

"Figure 6 shows bivariate polar plots obtained by combining wind analysis and M_{BC} with 1-hour time resolution in winter and summer. The densities of M_{rBC} data weighted by M_{rBC} values, and normalized by the maximum M_{rBC} were plotted as a function of wind direction and speed. The darkest areas of the wind pattern are those where the highest M_{rBC} was measured with a high occurrence, whereas lightest zones exhibit lowest measured M_{rBC} and/or a little occurrence of measurements. Note that locally emitted pollution at the measurement station was filtered before the analysis, limiting local M_{rBC} contributions emitted from the PDM station (i.e. section 2.3).

In summer, the highest M_{rBC} values were mainly associated with moderate wind speeds (above 5 m s^{-1}) and from the west and south west, suggesting a dominant regional transport (Fig. 6a). By contrast in winter (Fig 6b), the highest M_{rBC} occurred mainly under more static atmospheric conditions (ie. for wind speeds below 5 m s^{-1}) and no evident wind direction dependency. These results suggest that local-scale emissions could be a major contributor to M_{rBC} in winter unlike summer. Further discussion on the role of PBL influence on M_{rBC} will be addressed in Section 3.4.2"

L270: please define summer and winter, this applies elsewhere in the text.

REPLY : A sentence has been added at the beginning of the results sections as follows:

"In the following, seasons are defined as follows: winter (December, January, February), spring (March, April, May), summer (June, July, August), and autumn (September, October, November) "

L275-285: So, what it is the conclusion of this analysis?

REPLY : Separate paragraphs has been created in Section 3.3 to better highlight the conclusions.

L320: All paper is based on winter and summer differences. I suggest removing non-essential information like the daily cycle in autumn and spring. Considering that little to no explanation is

given about the diurnal-seasonal change, I cannot fully understand the relevance or the aim of this analysis. As said already, the authors should try to motivate the observed variability, giving context and explanation. Section 3.3 suffers, in its entirety, of this problem.

REPLY : The purpose of Section 3.3 is to describe the diurnal and season variation of BC properties, whereas Section 3.4 aims to investigate more deeply the causes of the observed variability.

As shown above, we added the main questions addressed in each section at the end of Section 1. In addition a sentence has been added at the end of section 3.3, lines 404-405, to explain the focus on winter and summer season in the following sections:

"Particular attention will be paid to winter and summer because these seasons differ greatly, whereas spring and autumn appear intermediate."

L325:Section 4 is still part of the results, right? So it should be Section 3.x. Please correct.

REPLY : We modified the name of this section by: "3.4 Investigation of factors influencing rBC properties"

L327: As it is shown in the following section, BC mass concentration and BC/CO ratio drastically change (at least in winter) due to anabatic injection from the PBL. Under the influence of PBL injection of fresh BC, wet removal has a smaller impact of BC properties compared to free tropospheric conditions. If the authors excluded periods affected by precipitation in Section 4.2, period under the influence of PBL should be excluded here. This additional filter will reduce the number of atmospheric variable and, perhaps, improve the interpretation of the results.

REPLY : We thank the reviewer for his suggestion. We agree with the reviewer that filtering periods under PBL influence is more rigorous to exclude the potential impact of PBL injections on the higher observed $\Delta M_{rBC}/\Delta CO$ when the airmass did not undergone precipitation events and with a low humidity. We applied this filter and changed the Figure 8 accordingly. The new figure is similar as the previous one, leading to the same interpretation.

L356: biomass burning influence. rBC emitted by different sources might show a difference in properties. If the biomass plumes were fresh, the authors should be able to see a difference in the size distribution, and, potentially, in the colour-ratio (ratio of BB over NB channel of the SP2).

REPLY : As explained above, the color ratio only provides information on the presence of dust particles (D. Liu et al., 2018; Moteki et al., 2017; Yoshida et al., 2016). To our knowledge, the color-ratio can not be used to assess the rBC composition.

We agree with the reviewer concerning the influence of the age of rBC from biomass burning emissions on its size distribution. However, no significant rBC size distribution variability between the seasons has been observed despite the different $\Delta M_{rBC}/\Delta CO$. This can be explain since even if the PDM was under the influence of different BC sources throughout the campaign, rBC from long-range transport appears to be the dominant source of BC and this is reflected in the globally constant rBC size distributions. Furthermore, the time resolution of the rBC size distribution (of a day) does not allow to see the impact of PBL/FT conditions (1-h of time resolution)."

L357: Be consistent with cross-references…heather is "Fig.X" or "Figure X"

REPLY : This was modified.

L362: this is most likely due to the lower concentration of BC observed in the PBL in the summer period.

REPLY : Lines 459-464 have been changed in line with the reviewer's remarks and now reads:

"Surprisingly the thermally driven PBL injection did not significantly impact M_{rBC} measured at PDM (Figure 10d). This contrasts with our winter observations and most previous surface measurements at mountain sites, where the daytime PBL development has been shown to enhance aerosol mass concentration (Herrmann et al., 2015; Venzac et al., 2009). The summer M_{rBC} values at PDM are twice as high as than those observed in winter, which indicates a massive additional regional transport of rBC in the FT and a lower contribution of rBC from PBL injection. "

L348-390: why the size distribution of rBC is not shown here? It might help with the data interpretation..

REPLY : Providing BC size distributions require extensive processing of the raw SP2 data. We have managed to analyze two years of SP2 measurements (representing more than 1.2 To of data) by using a time resolution for processing BC size distributions of one day. This time resolution do not allow to investigate the influence of PBL/FT on BC size distribution.

L369: what is "this evidence". Reduce the use of "this", it makes difficult to understand what the authors refer to.

REPLY : The sentence of lines 468-471 has been changed and now reads :

"The high rBC loading transported in the FT, coupled with the higher $\Delta M_{rBC}/\Delta CO$ observed in the summertime (Fig. 10d and e),could be due to a strong influence of biomass burning emissions on the background FT in Europe."

L371: IAGOS…Always explain every abbreviation

REPLY : The signification of IAGOS has been added.

L375-379: long unclear sentence, rephrase.

REPLY : The sentences in lines 469-472 have been modified and now reads :

"The majority of trajectories reaching PDM in summer have crossed the Iberian Peninsula and, previously, North Africa and North America (Fig. S7 in the Supplement). In these regions large fire events frequently occur, which may explain the high concentrations of strongly absorbing rBC observed at PDM during summer."

L384-390: Are the SMPS data filtered for FT and PBL conditions? Please specify. If this is not the case, PBL aerosol injection might potentially explain the concentration increase of smaller particles (PBL influence timing is exactly the same FigureS8). Overall, the statement is mostly speculative since the authors cannot prove the occurrence of coagulation and condensation on rBC cores. I thus would not call it "evidence" but rather "hypothesis". Moreover, the SP2 is capable of providing coating thickness (via the position sensitive detector) and a simpler proxy for mixing-degree (scattering- incandescence time lag). Could the authors explain why these two analyses were not applied?

REPLY : Due to a technical issue on the low gain of the scattering channel of the SP2, we could not provide BC mixing state measurements.

"Evidence" has been replaced by "hypothesis" and the end of Section 3.4.2 (lines 486-491) has been modified to focus on the shift of the dominant mode of the aerosol size distribution, which was potentially due to the condensation of preexisting particles on BC particle during the morning:

"At PDM the enhanced E_{abs} at noon was accompanied by a shift of the aerosol accumulation mode towards larger sizes, which may be due to the condensation of species on aerosol particles (Figure S10 in the Supplement). Simultaneously, a strong elevation of particle number concentration in the diameter range 10-30 nm can be observed, revealing new particle formation most likely produced by photochemical reactions at this time of the day. It is thus possible that rBC particles became more coated via condensation of species produced by photochemical reactions at noon. However, it cannot be ruled out that the evolution of aerosol size distribution is a poor indicator of the rBC mixing state."

L399-403: In the present work no evidence is provided on interaction with snow, coating thickness, lifetime, condensation rate or gaseous precursors. Only results obtained by the present study should be discussed in the conclusion section. This part is mostly speculative and I suggest removing it.

REPLY : The aim of this paragraph is to present our result on mean E_{abs} values. We think it is important to remind readers the importance of MAC_{rBC} for climate issues and put our results in the broad literature context.

L405: I would like to see if these results might change by removing the PBL periods.

REPLY : The remark has been answered above.

L406-407: avoid the use of references in the conclusions. Especially 4 in a row.

REPLY : References in the conclusion have been removed.

L415: What is the "evidence" exactly. Please elaborate.

REPLY : "evidence" has been replaced by (Lines 20-21) : "Combining $\Delta M_{rBC}/\Delta CO$ with air mass transport analysis, we observed additional sources from biomass burning in summertime leading to higher M_{rBC} and E_{abs}."

L423-427: ageing time scale and its impact on cloud activation and optical properties of BC is not treated in the present work. Saying that wet removal is independent from size and mixing state, and that hygroscopicity is not treated properly in models is a bold statement…I recommend caution. Same goes for the following statement.

REPLY : A Section 4 has been added which discuss on the implication of the results for climate models.

F1: This figure might benefit some editing. Besides the low resolution, I suggest removing the picture (although beautiful) and introduce a double map with a continental and regional scale. More info could be provided within the figure such as coordinates, altitude, managing institute, ACTRIS name, station type (mountain, background…), instrument list…

REPLY : Figure 1 has been edited. It now has a better resolution, and regional and local scales maps has been added.

F2: I do not think that Figure 2 is needed. The text in section 3.1 describes well enough the general meteorological conditions. Since day-by-day variability is not discussed (and there is no need), I suggest removing the full figure

F6: Figure 6, as Figure 2 and 4, suffers from the choice of using a daily temporal resolution. Since the authors are mostly discussing the seasonal variability, a longer time scale (month) will help visualizing the seasonal changes.

REPLY : We thank the reviewer for his suggestions.

The figure 2 has been moved in Supplementary materials and its temporal resolution has been reduced.

The temporal resolution in Figures 2, 4 and 6 has been reduced to monthly median values.

F10b-e: axis is Eabs, caption is MACbc, correct.

REPLY : This has been corrected.

FS6: I suggest plotting this graph with daily or weekly temporal resolution**.**

REPLY : The temporal resolution of Figure S6 has been changed to a daily time resolution.

FS9 Shouldn't the points have the same colour in the top and bottom panels?

REPLY : The color code is already the same for the top and bottom panels. There is a very slight difference of colors between the two panels due to different amount of E_{abs} and $\Delta M_{rBC}/\Delta CO$ data, leading to different mean wind direction for each hour .

FS10: usually nucleation mode is defined as D<10 nm

REPLY : "Nucleation mode" was replaced by "Lower Aitken mode".

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