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 #2

COMMENT #1:Instrumentation: In Sec. 2.2.1, the air was heated to keep the samplings dry. Could this measure alter the properties of BC, e.g., leading to some mass loss of volatile substances in the coating materials of BC particles?

REPLY : We followed the WMO-GAW and ACTRIS guidelines, which recommend a relative humidity below 40 % for sampling in order to avoid water uptake and be able to compare physical and optical measurements (GAW Report No. 227:WMO/GAW Aerosol Measurement Procedures, Guidelines and Recommendations, 2nd Edition2016). The Whole Air Inlet was heated to 20 °C, which is low enough to ensure that the sampling step did not affects the BC mixing state.

COMMENT #2: It will be also useful to examine the coated size of BC measured by the SP2 for the discussion about the mixing state of BC.

REPLY : We agree with the reviewers that a closure with the rBC coating measurement would reinforce our results on absorption enhancement However, due to a technical issue on the low gain of the scattering channel of the SP2, we could not provide rBC mixing state measurements.

COMMENT #3: Eq. (1) and Eq. (2) are wrong, it should be log (450/635) instead of log(450)/log(635).

REPLY : We thank the reviewer for this correction which we have taken into account.

COMMENT #4: Line 276: Based on your results, what can be the main influence that caused a higher ΔBC/ΔCO in summer than in winter when considering the larger amount of precipitation in summertime?

REPLY : Even after filtering precipitation days, we found a higher $\Delta M_{rBC}/\Delta CO$ in summer than in winter (cf. l. 333-334 :"Summer ratios were generally higher than winter emission ratios, which could reflect either lower rBC scavenging during transport or different emission sources of rBC between seasons..").

This means that the seasonal variation of $\Delta M_{rBC}/\Delta CO$ is probably due to different BC sources. The higher $\Delta M_{rBC}/\Delta CO$ in summer is most probably due to biomass burning emissions, which produce a higher $\Delta M_{rBC}/\Delta CO$ than fossil fuel emissions (Guo et al., 2017; Pan et al., 2011; Zhu et al., 2019). Further explanations on $\Delta M_{rBC/}\Delta CO$ variations was provided in lines 371-346. The argument of the biomass burning influence in summer is reinforced by the study of Dupuy et al. (2020) who found that Europe is strongly impacted by wildfires.

COMMENT #5: Line 313-314: "It is expected that the BC particles reaching PDM would be aged and relatively thickly coated." some evidence is needed here, a previous study which also conducted experiments on a mountain site influenced by the PBL intrusion (doi: 10.5194/acp-19-6749-2019) may support your results.

REPLY : For clarity reasons, the sentence in l. 389-392 was modified and references has been added:
" Given the remote mountain location and presumable distance from fresh BC sources, BC particles reaching PDM may have undergone aging and have gained a consistent coating. Previous studies found an absorption enhancement of BC due to its coating with the aging time (Peng et al., 2016; Sedlacek et al., 2022; Yus-Díez et al., 2022)."

COMMENT #6: The authors use "the size of BC core sampled at PDM was higher than other studies" to explain "BC wet scavenging did not significantly affect the size of BC-containing particles," which is not convincing. A previous study performed on a mountain site during wintertime in Beijing reported larger BC core than the present study but showed significantly lowered BC core size after wet scavenging due to larger BC cores were preferentially wet removed (doi: 10.1029/2019GL083171). Please give more explanations.

REPLY : In order to explain the impact of rBC wet scavenging on rBC size distribution, we compared the measured rBC core size distribution (see Fig. 9, which has been moved from the Supplement to the main text) of air masses affected or not by precipitation during their transport. The resulting rBC core size distribution were similar in both cases, which suggest that there was not a preferential wet removal of bigger BC cores during their transport to PDM.

The study of Ding et al. (2019) was conducted at a mountain site frequently under the influence of anthropogenic emissions coming from sources closer than the one influencing PDM. BC-containing particles may have different size and Kappa values than those measured at the PDM. Furthermore, due to different meteorological condition, supersaturation in clouds may significantly differs between the two sites. The cloud supersaturation is a key parameter impacting the critical activation diameter of Cloud Condensation Nuclei (CCN). Further simultaneous measurements of rBC mixing state and effective supersaturation in precipitating clouds are needed to test these assumptions.

COMMENT #7: The wet scavenging process of BC due to PBL cloud during its vertical transport within PBL around midday may partly explain the relationship between the diurnal variation of Eabs and ΔBC/ΔCO in summer (doi: 10.1029/2020JD033096)

REPLY : We thank Reviewer #2 for this comment. In order to eliminate the effect of rBC wet removal during PBL intrusions on E_{abs} and ΔBC/ΔCO, Fig. 8 has been modified by filtering periods when PDM was under PBL conditions. Similar results as before have been obtained with lower $\Delta M_{rBC}/\Delta CO$ obtained under precipitation, which suggests that rBC has not been removed during its vertical transport.

Minor:

COMMENT #1: Line 171: “1,95 should be 1.95”
REPLY : The notation has been corrected.

COMMENT #2: Line 173: what do you mean by “artifacts”
REPLY : An explanation has been added in lines 217-218 :
“Under precipitations some water droplet may indeed enter in the aerosol inlet and change both the inlet cut off diameter and the measured aerosol size distribution. This would bias all the measured aerosol properties. “

COMMENT #3:BL and PBL need to be uniform.
REPLY : The abbreviation PBL has been retained and harmonized in the manuscript.

COMMENT #4: Abstract, line 18: This sentence was confusing.

REPLY : The sentence in lines 18-20 has been modified and now reads :

“On the contrary, in summer, M_{rBC} showed no diurnal variation despite more frequent PBL conditions, implying that M_{rBC} fluctuations are rather dominated by regional and long-range transport in the FT.”

References:

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