

Interactive comment on “Sources and physicochemical characteristics of black carbon aerosol in the southeastern Tibetan Plateau: internal mixing enhances light absorption” by Qiyuan Wang et al.

Anonymous Referee #1

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* General comments

The authors have measured rBC mass concentrations, size distributions and mixing states at a remote site on the south-east Tibetan Plateau with a single particle soot photometer. The results are interesting but there are a number of potential issues with the measurements that don't seem to have been addressed, and a number of conclusions are reached without sufficient supporting evidence. Therefore, I have many comments that I believe must be addressed before the manuscript is considered for publication in ACP.

Conciseness is generally a problem. For example, the point that pollution can be transported to the Tibetan Plateau from the south through valleys is made multiple times in different locations with different types of supporting evidence. The conciseness of the manuscript could be improved considerably by making this point only once, with all of the supporting evidence raised at the same time. For example, Figs. 6c and the MODIS images in Fig. S5 make essentially the same point. Why are they discussed at different points in the manuscript?

The large rBC concentrations measured on 21 October is an interesting observation and perhaps worthy of further investigation. Is it possible to link this to a specific event, to see if such events might occur frequently in this region? This is potentially important since the concentrations observed on this day seem to be ~ 4 times higher than the normal daily peak concentrations.

There are still a number of grammatical errors in the manuscript and sentences that should be split into smaller parts. I have tried to point these out in my specific comments below but I cannot guarantee that this is an exhaustive list. Further proof-reading is required.

* Specific comments

P2, L2: Split the sentence in 2. E.g. '...high-elevation region. It holds ..'

P2, L23: Split the sentence. E.g. '...BC sources are strong (). The TP has become impacted...'

P2, L26: Split the sentence. E.g. '...of surrounding areas. Annually, on average, South and East...'

P3, L4: Artifact should be changed to artefact. And this sentence appears to be conflating two separate concepts. The bulk collection of particles on filters is a design choice, not a measurement artefact. E.g. particles could be classified by size before they are collected on filters. Measurement artefacts are errors arising from the measurement

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technique itself, e.g. the filter loading effect.

P3, L11: 'in' missing between 'change' and 'absorption'.

P3, L19: This is not consistent with general practice in the field. E.g. BC mass calculated from measured light absorption is called equivalent BC (eBC), not simply BC (Petzold et al., 2013).

P3, L25: This sentence requires re-wording. What sort of samples? The sentence should also be split. The free-troposphere part deserves its own sentence. Under what conditions are free tropospheric air masses sampled at the site?

P4, L6: It should also be mentioned that the SP2 detects elastically scattered light as well as thermal radiation, since the 'scattering signal' is discussed later in the discussion of how SP2 provides mixing state information.

P4, L21: It is not clear how the 20% uncertainty estimate has been arrived at. Please provide further quantitative details. Was the 'SP2 response to ambient rBC mass' determined from an independent measurement of BC mass?

P5, L4: Fig. S1 does not show that the 'vast majority' of rBC particles had VED between 70 and 300 nm. The figure shows that at least approximately half of the rBC particles had VED less than 70 nm (possibly more since the peak of the $dN/d\log D_p$ curve has not been reached by 70 nm).

P5, L8: Particle losses in such a Nafion tube (diameter 0.11 inches, length 48 inches) can be very large. Tubes of such small diameter are not typically used in aerosol sampling lines. The authors should demonstrate to what extent particle losses may have affected the measured b_{abs} values.

P5, L15: b_{ext} values calculated in this manner require very large aerosol loadings due to the short 0.354 m optical path length in the PAX. The aerosol loadings used for the calibrations should be mentioned along with the range of ambient values measured, to give a sense of where the calibrations have been extrapolated to.

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P5, L24: Please provide details of how the $\sim 15\%$ measurement uncertainty has been arrived at for the PAX. Additionally, the PAX can have difficulty measuring low, ambient absorption levels accurately (e.g. $< \sim 1 \text{ Mm}^{-1}$). Was a lower limit of detection/quantification used in this study?

P6, L1: Please provide more details on the PBL depths that were used. Are they from a model? It is not clear which data were used by clicking through to the link provided.

P6, L19: It would be useful to discuss what this parameter means physically. I.e. positive values of f indicate ...

P6, L25: Please provide further details about the actual clustering procedure that was used. Currently, only the calculation of the mean angle parameter d_{12} is discussed. No information is provided about how this parameter was used to cluster back trajectories into different groups.

P7, L10: This sentence requires more precision. E.g. change to '...the rBC sources that potentially influenced the air sampled at Lulang'

P7, L26: This is difficult to see in Fig. S3. I suggest plotting the frequency distribution on a log x-scale or maybe a reduced x-axis length to highlight this point.

P9, L6: Were data from the 21 October included in the calculation of these diurnal profiles? I'm thinking if the apparent night time peak (or any other feature in the profile) was simply a result of this one-off event of high concentrations.

P9, L18: Missing 'by' between 'accompanied' and 'deepening'.

P9, L27: Change 'south' to 'southern'.

P10, L14: While this is physically plausible, a statistical test should be conducted to determine whether it is really possible to say from this dataset that rBC concentrations were lower on moderate or heavy rain days than rBC concentrations on light rain days. Given the small sample size (only 4 moderate or heavy rain days), it may not be.

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P10, L15: Split the sentence. E.g. '... (Fast et al., 2007). Fig. 4a shows...'

P11, L30-35: While interesting, this is the 3rd time this observation has been mentioned. The conciseness of the manuscript could be improved considerably by making this point only once.

P12, L14: An 'and' is required between 'rural' and 'remote'.

P12, L31: Please provide more explanation as to how air mass transport histories might affect rBC size distributions. I'm not aware of any mechanism by which rBC core VED (i.e. rBC mass) would change during transport (e.g. due to evaporation or condensation). Size-dependent removal processes could change the rBC core size distribution as discussed in the sentences following. In any case, the cluster analysis example that is given does not demonstrate that air mass transport histories affected the rBC size distributions. The rBC from the different source regions may simply have had different initial size distributions.

P13, L10: Two things could explain the observed differences in Fig. 7: the absence of long-range transport during the rainy days or preferential wet scavenging of larger rBC cores during the rainy days. For the 2nd hypothesis, could the authors provide a reference or further theoretical argument to indicate whether this is feasible?

P13, L31: Photochemical production of coating material is just one explanation for the increased F_{rBC} during the afternoon. Mixing layer height was also high during the afternoon. Comparison of Fig. 3 and Fig. 8 suggests F_{rBC} also correlated well with mixing layer height for the periods from 10:00-19:00. Thus, another potential explanation for the high F_{rBC} values observed in the afternoon is the mixing of more aged BC particles from aloft to the surface.

P13, L32: I don't think it has been demonstrated that in situ photochemistry is completely responsible for the afternoon increase in F_{rBC} (see above comment). Therefore, I don't think it is justified to report oxidation rates and compare such rates with

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those observed at Qinghai Lake.

P14, L20: The PAX can have difficulties to measure b_{abs} values less than $\sim 1 \text{ Mm}^{-1}$. Since a considerable amount of the measurements fall in this range, could the authors provide a scatterplot of b_{abs} vs rBC concentrations from the SP2, and if relevant add it to the supplementary information? Such a plot might help to determine if a lower limit of quantification should be applied to the PAX measurements. E.g. if it shows the PAX was insensitive to changes in BC concentration below some threshold.

P14, L26: Given the very narrow nafion drier used in front of the PAX, I think it must be checked whether the b_{abs} measurements are biased low, which would mean these MAC values are also biased low.

P15, L12: Should be '... if the fraction of thickly coated rBC particles increased by one percent...'

P15, L25: More specifically, if coatings are formed by condensation, this is due to the $1/\text{Diameter}$ dependence of the condensation rate.

P16, Section 4: A number of the conclusions made in this section might need to be updated after the specific comments above have been addressed.

References: Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A. and Zhang, X.-Y.: Recommendations for reporting “black carbon” measurements, *Atmos. Chem. Phys.*, 13(16), 8365–8379, doi:10.5194/acp-13-8365-2013, 2013.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-801>, 2017.

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