Dear Dr. Töpfer

We have addressed the reviewers' comments below and made substantial changes to the manuscript, including the title and abstract. We have removed the case study–application of our empirical findings to IMPROVE network data–because of crucial differences between their measurement method and ours. We have also elaborated on how the radiative transfer modeling informed this study. With these adjustments to the scope and technical clarity of our paper, we look forward to a favorable decision on this revised submission.

Changes made to the manuscript, as well as our responses to reviewers' comments are highlighted in red.

Thank you,

Sincerely,

Rajan K. Chakrabarty and co-authors.

Reviewer 1

General comments of the "implications" discussed in our manuscript.

"The discussion paper undertakes to produce an empirical calibration for a specific filter-based measurement in terms of in-situ aerosol measurements. Its finding, that applying the same calibration to a completely different measurement system in Figure 5 produces an unsatisfactory relationship, should not surprise anyone who recalls the authors' earlier comment"

We are grateful to Dr. White for his insightful, detailed comments on our manuscript. We found these comments very helpful in refining the thought process behind this paper, leading to a substantial revision. We agree that the empirical equation developed in this work is not applicable to IMPROVE data and have entirely removed the "implications" section. We have also highlighted how the filter optical measurements used here are distinct from those in literature and provided our reasoning for defining this alternate measure of optical depth. Further, several new figures have been added to the manuscript and the supplement to better illuminate our modeling results (Figures 2, 3, 5, S2-S5).

Specific comments 1 and 2.

"The entire description of the authors' filter optics measurements is a scant two sentences: "Transmittance (T) and reflectance (R) for the filter samples were measured using a Perkin-Elmer LAMBDA 35 UV-vis spectrophotometer…… if R were measured on the side darkened by the collected sample deposit."

The filter optical measurements are now described on page 6, Line 5. The rationale behind using reflectance measurements on the "sample" side of the filter is detailed in Section 2.1. Briefly, we recognize that the quantity $\ln(\frac{1-R_s}{T_s})$, where R_s and T_s denote reflectance and transmittance measurements made on the sample side, is not equivalent to Beer-Lambert attenuation. We now explicitly define it in the manuscript as a separate measure of filter optical depth (represented by OD_s). We experimentally observed that this alternate filter optical measure is remarkably well-correlated with *in-situ* absorption optical depth. We also found that the two-layer filter model supported our findings (Fig.s 2 and 3 in the main manuscript) and indicated that the reason for the above constrained relationship is the behavior of R_s as a function of filter loading, for different values of SSA (Fig. S2 in the Supplement).

Specific comments 3 and 4.

"The experimental program described in this discussion paper was well designed to test our standard accounts of radiative transfer in filter samples, providing real data for a closure study that I have not seen attempted before. … Absent any clear link to the measurements, what was the intent of the modeling? Relatedly, in what sense is Equation 7 a "best fit" to the observed relationship between ATN and AOD?....What sort of functional form does the two-stream model predict when it is run over a wide range of sample loadings from a representative aerosol of fixed optical characteristics?"

A comparison between modeled and measured values of OD_s is now shown in the manuscript (Fig. 5). The predicted relationship between AOD and OD_s (along with other commonly used filter optical measurements) is demonstrated in the Supplement (Fig. S5) and discussed in the manuscript. Due to the quadratic dependence of R_s on particle loading, a non-linear function form is required to capture the relationship between OD_s and AOD for any SSA value, but the coefficients of such a relationship (say, power law) are not consistent across all SSA values.

Specific comment 5

"The only results I see explicitly attributed to the two-stream model are the colored wedge shapes in Figure 2. I don't understand why these don't show y-axis ATN declining toward zero, the model prediction for a blank, as the virtual absorption optical depth of the aerosol sample approaches zero on the x-axis."

This was due to a calculation error at zero aerosol optical depth which has now been fixed.

Specific comment 5

"The two-stream radiative transfer modeling framework presented here, along with its hyperbolic solutions, is widely known also as Kubelka-Munk theory, after an extensive older literature arising in paper and paint research (e.g. Kortum, 1969). Referencing this connection in passing might attract a few additional readers."

We have a reference to the Kubelka-Munk theory to the manuscript.

Reviewer 2

"In this manuscript the authors investigated the relationship between absorption optical depth of aerosol carbonaceous particles derived from in situ measurements and attenuation obtained using filter based method. The authors derived empirically an estimation of the aerosol mass absorption coefficient (MAC) from PTFE filter measurements…. Finally, I think the author could discuss and explore the results found in the case study in more details."

We thank the reviewer for their helpful and encouraging comments. Throughout the manuscript, we have now ensured technical precision and clarity in our language. Relevant experimental details have been added and uncertainties have been stated in our figure captions. Finally, because of a crucial difference between our measurements and those in the IMPROVE network, we have removed the case study section from the manuscript.

P1L15: "When you use the term "light absorption", do you mean the mass absorption coefficient or absorption optical depth or the linear absorption coefficient? The same thing for "attenuation measurements". Are you referring to extinction, absorption or something else? If something else, please define." P1L18: In the sentence "we find the ratio between in situ absorption and bulk attenuation to be inversely proportional to aerosol single scattering albedo", please clarify the term "absorption". Are you referring to absorption optical depth?

We have carefully gone through the manuscript to ensure that all optical quantities are defined and referred to more specifically.

P1L21: What are the results from the case study? You have not mentioned that in the abstract.

The case study has been removed.

P1L27: For completeness, you could add here a few examples of known carbonaceous sources with different MAC.

Typically observed ranges of carbonaceous aerosol MAC values have been added at P2L2.

P2L18: Another relevant reference here: Martins, J. V., Artaxo, P., Kaufman, Y. J., Castanho, A. D., and Remer, L.: Spectral absorption properties of aerosol particles from 350–2500nm, Geophys. Res. Lett., 36, L13810, https://doi.org/10.1029/2009GL037435, 2009.

This reference has been added to the manuscript.

P3L26: You mentioned here "sampled by the four IPNs" but I don't see any reference to these measurements later in the text. It might be interesting to comment if the average absorption coefficients was derived using those four instruments. Also, it would be interesting if you could report/discuss for each sample type.

A sentence on the range of absorption coefficient values has been added at page 6, line 5. Given the huge variability in absorption and scattering coefficients, we only discuss the dimensionless optical properties calculated from these measurements (aerosol optical depth and single scattering albedo).

P4L5: I suggest adding here a figure or table reporting the average values of the absorption coefficient in Mm-1 as well as the standard deviation for the different samples. Also, I would move table S1 that is currently in the abstract to the main manuscript.

We have moved the table of experiments to the main manuscript.

P5L5: In the sentence: "the penetration of aerosols into the filter was assumed to be 10%". Could you please clarify the meaning of that? Do you mean 10% of the mass collected? How do you estimated this number? Figure 5 shows concentrations in the range from 0.01 to near 10ugm-3. Would you expect the percentage be strongly dependent of the mass concentration collected on the filter?

We assumed that aerosols only penetrated the first 10% of the filter thickness. Membrane filters are expected to have a low penetration depth, so we assumed a reasonable value here. We expect the general trends from our model to hold even with variations in penetration depth. Our supplement now includes a figure (Fig. S3) showing the effect of this parameter on transmittance and reflectance.

P5L13: I think it is missing here an evaluation of the uncertainties in the calculation of ATN.

Figures 2, 3 and 5: These plots are missing error bars. You should also add information about the uncertainty of the measurements in the legend of each figure.
The calculation of measurement errors is described on P6L30. Errors were random and well-constrained, with little variability from sample to sample. We believe that adding error bars to Figure 6 (which contains 225 data points) does not add to the visual interpretation of these data. We did, however, add information about the measurement uncertainty to the figure caption.

P7L9: How these method compare/differ from the derivation of the mass absorption efficiency presented in (Martins et al., 2009)?
Martins et al., 2009 used a theory-based model to derive mass absorption efficiency, assuming an absence of multiple-scattering artefacts. Our goal was to link filter optical measurements to aerosol optical depth, and compare experimental findings with predictions from a radiative model of particles embedded in a multiple-scattering medium.