

Interactive comment on “Photophoretic spectroscopy in atmospheric chemistry – high sensitivity measurements of light absorption by a single particle” by Nir Bluvshstein et al.

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We would like to thank anonymous referee #2 as well, for carefully reading our submitted manuscript and for raising important questions that helped us improve the quality and readability of the manuscript.

Answer to question a:

Please refer to the answer given to question #1 from referee #1. Figure 1 in the reply to referee #1 will be added to address this question.

Answer to question b:

C1

Values of κ for BrC reported in the literature are limited by the detection limit and sensitivity of the measurement technique that is used. Therefore, a more accurate statement would be that quantifiable values of κ for BrC are typically much larger than 10^{-4} while the reality is that there is no physical lower limit for absorptivity of BrC in the atmosphere.

An alternative argument could be made that lower values of κ for aerosols are insignificant due to their low thermal effect (at least in the troposphere). It is, therefore, worth clarifying that the main motivation for the application of this technique is to measure these previously unquantifiable values and to follow the evolution processes of BrC during atmospheric aging. This could mean either ‘bleaching’ or ‘browning’ and for that reason, one should be able to retrieve κ at lower values that are commonly reported.

To answer this comment and comment number 6 from reviewer 1 we will add the following text at the end of the introduction in the revised version of the manuscript:

“With this approach, we gain from combining the advantage of light absorption sensitivity nearing that of bulk UV-vis measurements with the advantage of studying chemical processes of the particle phase (accessible supersaturated conditions) in an environmental chamber able to simulate a wide range of atmospheric conditions. This could contribute to the study of light absorption evolution during atmospheric aging of BrC aerosols.”

Answer to question c:

As mentioned above, any color-forming or degrading aging processes could be relevant candidates. Oligomerization, nitrification, acid-catalyzed dehydration have been reported as a possible mechanism for chromophore formation. Oxidation and photooxidation reactions are known bleaching processes. One particular system of interest is the browning of sulfuric acid aerosols in the presence of ketones or aldehydes. For this system, the formation of UV-Vis chromophores was reported so far only in bulk and film experiments but not for aerosols.

C2

Answer to question d:

We appreciate the reviewer's suggestion and agree that such a measurement would be beneficial to demonstrate the usefulness of this method. To produce a sample of humic or fulvic acid material with low absorptivity in the range of $\kappa \leq 10^{-3}$ at 473 nm it would need to be highly diluted or mixed with a non-absorbing material. This cannot be achieved with water because these particles are not sufficiently hygroscopic (additionally, we refer the reviewer to a short discussion about measurements at high RH in the answer to question 3 by reviewer #1). Unfortunately, this was attempted but could also not be achieved with PEG400 due to extremely low solubility. An additional option would be to simply use a longer wavelength for the excitation light source. This was, however, not available at the time of performing these experiments.

Minor comments:

-no need to use a non-standard symbol like χ for the size parameter when the standard x will do fine.

The notation for size parameter was changed from " χ " to " x " in the text as well as in all of the equations.

-Line 134: "retrieved by minimizing the difference between measured and calculated wavelength" I assume you mean minimizing the sum of squared differences?

What was actually done was to minimize the sum of the absolute value of the differences. This will be added in the text.

-use of the times symbol in many equations is unnecessary.

To improve clarity, the multiplication symbol was removed from all equations.

-Line 225: What is the viscosity of PEG400?

A range of viscosity values for PEG400 as reported by the supplier (Merck), at 293.15 K was added to the text; 105 - 130 mPa sec.

C3

-Line 269: "473-nm" to "473 nm"

This was correct in the text.

-Figure 2 caption: intensity units are italics when they should not be

This was corrected in the text.

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