Manuscript no.: Atmos. Meas. Tech. Discussion: 7/C2989/2014

Title: **Mass specific optical absorption coefficients of mineral dust components measured by a multi wavelength photoacoustic spectrometer**

Authors: **N. Utry, T. Ajtai, M. Pintér, E. Tombácz, E. Illés, Z. Bozóki, and G. Szabó**

Reviewer #3:

Reviewers' comments: This manuscript describes the use of a four-wavelength (266 nm, 355 nm, 532 nm, 1064 nm) photoacoustic spectrometer to measure the absorption of various mineral dust (MD) components in aerosol form. The measured absorption values are combined with aerosol size distribution and mass measurements to derive mass specific absorption coefficients (MAC's) for all seven components. These values could be of utility to others wishing to estimate absorption by mineral dust aerosols or, as the authors suggest, for comparing to bulk measurements. Comparisons to absorption redicted by Mie theory and based on previous (bulk) measurements of indices of refraction demonstrate general agreement between the two approaches but also indicate some discrepancy which may, or may not, be attributed to deficiencies in the methods used to make the bulk measurements.

Overall, the manuscript is fairly well written, though there are some sections and paragraphs that run on too long. Conversely, the Results section is only one paragraph long; I suggest that the authors consider moving Section 2.4 (Calculations) to Results from Experimental and methods. The authors have done a good job of describing the instrument and the methodology as well as the calculations. The scope of the results and conclusions from this work is fairly limited, but they still could be of use to others in the field and therefore I recommend publication with the following comments being considered.

Authors' response: First of all, we would like to thank the reviewer for this review. The authors completely agree that all the implementing comments and suggestions in the revised MS have really improved its scientific level and also its impact on the field.

Specific comments:

1. **Please show reproducibility of the absorption measurements for individual MD components. Also, please indicate how many data sets or measurements were averaged to obtain the results presented in Figure 3 and Table 1.**

Authors' response: We really agree with the reviewer that the concentration instability of the re-dispersed particulate matter is one of the major limitations of its investigation. Therefore, the concentration instability implemented into the parameters which are actually measured and the protocol of the applied data acquisition and evaluation need to be described in more details in this study.

Therefore we added the following to the revised MS on page 5 from line 202-207:

*"*In the measurement mode a complete 15-minute-long measurement cycle includes a 10-minute sampling and a 5-minute background measurement period. Generally, 8 measurement cycles were used for data evaluation. The initial transient period was not evaluated. Depending on the investigated mineral dust component and the applied wavelength, the fluctuation of the PA signal including concentration instability was found to be in-between 15% and 22%

2. **How appropriate is the use of Mie theory with MD particles? Specifically, how good is the assumption that the particles are homogeneous and spherical? This should be addressed quantitatively and with citations to previous work in this area since comparisons are made to Mie theory results.**

Authors' response: We definitely agree with the reviewer that using the Mie model for non-spherical particles such as dust minerals means limitation to the calculations. So according to this suggestion we explain the legacy and the limitation of Mie code for the retrieval of mass specific absorption coefficient and the imaginary part of refractive indices in the revised MS.
page 11, line 293-306:

i.e. "However, it is noteworthy, that although the application of Mie-theory for non-spherical particles limits the reliability of the computed data, many prior works used this approach to calculate the spectral responses of dust minerals (Conant et al.,2003;DeSouza-Machado et al.,2006;Moffet and Prather, 2005;Wang et al.,2002). This is because the most widely used shape sensitive models such as T-matrix or DDA (Discrete dipole approximation) are also not using the real morphology of the investigated aerosol sample but they are based on mathematically well characterized geometrical approximation (Kalashnikova and Sokolik, 2004; Tegen and Lacis, 1996). (Kalashnikova and Sokolik, 2004; Tegen and Lacis, 1996). Kalashnikova and Sokolik demonstrated that the deviation between the spherical and non-spherical approaches become significant above the approximate size parameter of 5 and only in case of the asymmetry parameter and the scattering phase function which are mainly governed by the coherent scattering physical process, while the absorption which is an incoherent physical phenomena, the deviation is much less significant even above size parameter higher than about 5."

3. **Is "AOC" a common abbreviation for "aerosol optical absorption coefficient"?**

Authors' response: We agree with the reviewer that the applied abbreviation here is not the most frequently used one for the aerosol optical absorption coefficient. We changed the "AOC" to the more common "OAC" abbreviation in the revised MS.

4. **Page 9027, lines 17-18: The work cited of Lin and Campillo, 1985 doesn't seem to have employed either the difference approach or the filter transmission approach mentioned**.

Authors' response: We agree with the reviewer in that the reference cited to this statement is not the proper one therefore we neglect it from the revised MS.

5. **Page 9028, line 27-29: Citations to previous work with photoacoustic spectroscopy of aerosols need to be added.**

Authors' response: we agree with the reviewer that citation to some previous works in which the aerosol phase photoacoustic spectroscopy is used should be added here, therefore, according to this suggestion we cited some other works related to this issue in the revised MS.
page 4, line 91-95:
i.e. "The photoacoustic (PA) measurement technique has already proved its applicability under laboratory and field conditions in case of strongly absorbing aerosol components such as black carbon, brown carbon and HULIS (HumicLike Substances) (Andreae an Gelencsér, 2008; Cappa et al., 2008; Lack et al 2002; Chakrabarty et al.2011; Moosmüller et al., 2012; Ajtai et al.,2011; Utry et al.;2013).

6. **Page 9030, line 2: What is meant by "free-floating operation"? Measurement of suspended particles? Please be more specific.**

Authors' response: we agree with the reviewer that "free-floating operation" used here instead of measurement of suspended particles is confusing. Therefore, according to this suggestion, we modified the text in the revised MS.
page 4, line 95-97:
*i.e. "*The accuracy of the method stems from both its scattering insensitivity and its filter free sampling (measurement made on the aerosol on its natural, suspended state)*."*

7. **Page 9033: With such wide size distributions, how are doubly-charged (and triply charged for that matter) particles accounted for?**

Authors' response: we agree with the reviewer that the reliability of the size distribution measurement is limited by multiple charges especially using wider size ranges (measurement with long DMA). It is noteworthy (although we did not implement it into the text) that the differences between the OPC and the SMPS data in their overlapping size domain were found to be inside the cumulative uncertainty range of the instruments, which is kind of indirect validation of the applied data evaluation protocol. Therefore, according to this suggestion, we implemented the applied data evaluation protocol including multiple charge correction in the revised MS.
page 8, 215-217:

i.e. "The multiple charge correction defined by the ISO 15900 recommendation was used in the data evaluation during the whole measurement campaign to further increase the reliability of the measured data."

8. **Page 9033, line 9: Please quantify the "Negligible differences" between the CPC and the OPC particle counting in the overlap region.**

Authors' response: we agree with the reviewer that "negligible differences" used here is laxity and the differences between the measured data in the overlapped size region should be more precisely quantified in the MS. Therefore, we implemented this suggestion into the revised MS.
page 8, line 223 – page 9, line 225.
i.e. "The differences between the measured data from SMPS and OPC in the overlapping size domain was found to be inside the cumulative uncertainty range of the instrumentations (i.e. below 10 %), therefore it was neglected during the data evaluation."

9. **Page 9036, lines 16-17: How do the data in Table 1 and Fig. 3 (I assume "Fig. 1" on line 16 is a typo) prove the MD components are volumetric absorbers. This statement needs to be explained in much more detail.**

Authors' response: we really agree with the reviewer in that the dust minerals sample presented here cannot be typified as volumetric absorber with exhaustive confidence and using this classification is confusing and might mislead the readers. Since we cannot support this statement adequately, under this suggestion, we neglect this classification in the revised MS.

10. **Page 9036, lines 17-19: What are MAC's black carbon and brown carbon for comparison to the MAC's for MD measured here?**

Authors' response: we agree with the reviewer that some literature MAC values of black carbon and brown carbon should be added here for intercomparison purposes, therefore, according to this suggestion we cited some other works related to this issue in the revised MS.
page 16, line 363-367:

"As it is expected, their MAC values are several orders of magnitude smaller than those of BC (7.5±1.2 m^2/g@550 nm (Bond and Bergstorm, 2006), 10±3.5 m^2/g@405 nm (Cross et al., 2010)) and BrC (0.5-1.2 m^2/g@404 nm (Lack et al.,2012)."

11. **Table 1: Lambda_2 should be a lowercase lamda. Also, how do these values of MD MAC's compare to any others that have been measured?**

Authors' response: We agree with the reviewer that the Lambda_2 is a typo which should be repaired in the revised MS. We also agree with the reviewer that the presented values would need to be implemented into the earlier published results to compare. Anyway, the available MAC data defined at the specific wavelengths used in this study are really limited with incomplete description in many cases in the literature. However, the presented results shown similarity with other earlier published data measured i.e. indirectly on the water suspended dust components (Babin et al., 2004). Therefore, the authors have decide to make the quantitative comparison not on the MAC values but rather on the calculated and measured OAC values as well as on the calculated and the literature κ values.

12. **Figure 1: "Teom" should be all capitals: "TEOM". Also, the dotted flask and the associated arrow are confusing. Is this meant to represent shaking of the flask?**

Authors' response: we agree with the reviewer in that although in the experimental section we describe the whole aerosol generation procedure including shaking of the vessel, in Figure 1, it is not described unambiguously. Therefore, according to this suggestion, we also modify the figure caption in the revised MS. Furthermore; we also change from "Teom" to "TEOM" in this figure.

i.e. "Figure 1: Schematic diagram of the experimental setup (The vessel with dotted contour line and the arrow indicates that it is being shaken)"

13. **Figure 3: The axis of ordinates uses the abbreviation "OAC" for the aerosol optical absorption coefficient, but "AOC" is used everywhere else in the manuscript**

including in the caption for this figure.

Authors' response: We changed the "AOC" to the more common "OAC" abbreviation in the revised MS.

Finally, we also appreciate and partly agree with the suggestion regarding to the structure of the MS. Therefore, to reduce the disproportion in between the length of sections we merged the section 3 and 4 and named this section results and discussion. Anyway, we also describe the calculation separately in the subsection of experimental and methods.