Atmos. Chem. Phys. Discuss., 11, C11681–C11686, 2011
www.atmos-chem-phys-discuss.net/11/C11681/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Complex refractive indices of Saharan dust samples at visible and near UV wavelengths: a laboratory study" *by* R. Wagner et al.

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Received and published: 9 November 2011

We thank Referee #2 for his comments to improve the quality of the paper; below, we address the individual comments and outline our suggested changes for the revised manuscript text.

COMMENT:

1) Error analysis and error bars shown in figures: Figure 14 shows frightening error bars for the measurements of dust absorption coefficients by SOAP, that is error bars covering an order of magnitude or more for most visible and near-infrared wavelengths (450 nm and above). However, fig. 13 showing retrieved spectra for the imaginary part

of the imaginary part of the refractive index k does not show corresponding error bars. If careful error analysis shows that errors in k are comparable in size to those of the absorption coefficients and exceed the spectral variation in k, the main result of this manuscript would be gone. While the authors claim "that a detailed inter-comparison of the two techniques for measuring particle absorption [i.e., SOAP and photoacoustic] which would also help to better constrain their respective measurement uncertainty is beyond the scope of the present manuscript.", I would think that this would be the most promising way for possibly reducing the excessive errors in their SOAP absorption measurements. In any case, fig. 13 needs error bars and a discussion if any spectral variations exceed these error bars.

ANSWER:

Much the same concern has been expressed by Referee #1 (see point 7) and we therefore again outline our envisaged improvements to the manuscript text with respect to the uncertainty level of the retrieved k data sets:

The error estimate of our retrieved k data sets is indeed a crucial aspect in our work. The large error bars for the SOAP retrievals (Fig. 14) reflect the weakness of any filter-based absorption measurement, namely a residual cross sensitivity to aerosol scattering. Even if this cross-sensitivity is small and a good measurement of the scattering coefficient is available, it cannot be completely corrected because it is an inherent problem of the method. This error is particularly pronounced for aerosols with a single scattering albedo above 0.95, i.e., for the spectral range above 600 nm in the case of mineral dust. The good reproducibility of the SOAP retrievals shown in Fig. 13 therefore reflects the good precision of the instrument but tells nothing about its accuracy. Unfortunately, using closure between scattering and extinction, as suggested by Referee #1, would also not help because we have a similar inherent problem in this difference method in the case of weakly absorbing aerosols.

We therefore completely agree to extend the inter-comparison between the SOAP and

the photoacoustic data in the revised manuscript text although the novel 4-wavelengths photoacoustic spectrometer has been employed for the first time to measure dust absorption and its estimated uncertainty levels at the different wavelengths (Fig. 14) were derived from former inter-comparison measurements with the difference method for strongly absorbing soot aerosols (Ajtai et al., 2010).

The comparison with the photoacoustic data shown in Fig. 14 already highlights one important finding, namely that the varying background absorption level of the dust probes above 600 nm is indeed a real physical effect because the variation in the absorption coefficients of the two dust probes is much larger than the estimated uncertainty level of the photoacoustic data.

As suggested, we will repeat at least two retrievals of the imaginary complex refractive indices also for the photoacoustic data and will add these data, together with their error estimates, to Figs. 13/15. These data will also corroborate that the variability of the magnitude of the k values for the different dust probes towards UV wavelengths depends on their iron oxide content because it is much larger than the uncertainty level of – in particular – the photoacoustic data, being the second major conclusion from this manuscript. The difference between the SOAP and photoacoustic k retrievals will then also be a more realistic estimate of the uncertainty of the retrieval results than the standard errors currently denoted in Table 5 and shown in Fig. 13 as error bars. This aspect will be discussed and corrected in the revised manuscript text.

COMMENT:

2) T-matrix modeling of optical particle properties is based on the axis ratio AR determined from SEM and TEM images. However, the use of 2-d images to determine 3-d particle properties depends critically on assumptions about particle orientation on the filter substrate. Do the particles have random orientation or are they oriented on a flat substrate minimizing the distance between their center of gravity and the substrate plane? What is the surface roughness of the filter substrate?

ANSWER:

The impaction stage (TEM grid) is smooth. We know that chain-like particles orientate in parallel to the streamlines in an impactor, meaning that they would lie flat on the surface. The dust particles, however, are not so extreme in shape; so it is not sure whether this applies because the orientating forcing should be much weaker then. After impaction, the particles might orientate on the substrate provided that they are asymmetrical enough to overcome their current adhesion forces.

As outlined in the first paragraph of Sect. 4.3, it was our initial approach to use the measured AR distribution in the retrieval of the complex refractive indices, for which we have contrained the 3D AR bins between 0.25 and 4 in our data base calculations, based on the extreme limits observed in the TEM images. When analysing the shape dependency of our retrieval approach (Fig. 11), it was an interesting result that compactly shaped spheroidal shapes (with the median AR obtained by fitting ellipses to the projected particle areas in the image analyses, Fig. 6) or spheres were not able to accurately reproduce the spectral habitus of the measured extinction spectra. Rather, more elongated spheroids with an AR corresponding to the extreme limit observed in the TEM images best fitted the measured extinction spectra. This might be partly related to deficiencies when using 2d-images to infer 3d-particle properties. More likely, however, a smooth spheroidal shape is simply not adequate to describe the extinction coefficients of an irregular dust grain even if the overall AR is the same, at least in the spectral regime governed by the interference structure.

Notwithstanding this uncertainty with respect to the accurate shape respresentation of the dust grains for modelling extinction in the regime governed by the interference structure, its quantitative impact on the accuracy of the retrieved k data sets is – fortunately – rather low, in particular in comparison with the measurement uncertainties outlined above. We have highlighted in our analysis (page 21391, line 17ff) that irrespective of the choice of the aspect ratio in the retrieval approach, the retrieved k spectra are only affected by a small uncertainty of less than 10%. We will emphasize

this aspect more clearly in the revised manuscript text, including parts of the discussion from above.

COMMENT:

3) The authors talk multiple times about “re-dispersed soil samples”. This makes perfect sense for the Burkina Faso sample consisting of “wind-blown material”. However, it is not clear if the other soil samples are “re-dispersed” or simply dispersed.

ANSWER:

Yes, since it cannot be specified how much of the other topsoil material was already wind-blown, we will only use the word “dispersed” throughout the manuscript text.

COMMENT:

4) The authors state on p. 5 “Since light absorption by the dust particles is governed by their volume concentration,. . .”. My understanding is that this is true only in the Rayleigh regime (size parameter xÂń1) where the absorption coefficient is proportional to the particle volume, while for very large particles in the geometric optics regime (xÂż1) the absorption coefficient is proportional to the projected area. The dust particles considered here include large particles (x>1), so this statement is questionable.

ANSWER:

This is absolutely correct and will be clarified in the revised manuscript text. We have discussed this issue in somewhat more detail in the context of Fig. 8 (page 21383, line 16ff) when analysing the shape dependency of the absorption cross sections: it can be seen that above a particle size of 1.5 microns, the absorption cross sections start to become sensitive to the surface area of the particles. But this threshold also depends on the magnitude of the imaginary index, which was 0.014 in the presented example. Due to the manuscript re-organisation recommended by Referee #1 and in order to avoid duplications, the text passage under discussion will be shifted to our new Sect. 4.2 “Retrieval approach: Basic considerations” because the uncertainties

associated with sizing of the dust particles are an important reason for our modified retrieval approach and will be concisely discussed in this new chapter.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21363, 2011.