Author's Response to review comments of Referee #3

by Friederike Höpner on behalf of the authors
December 12, 2015

We thank referee#3 for her/his constructive comments which we will account for in the revised manuscript. Below is a point-by-point response to the comments. A marked-up manuscript is provided as well.

1. *Single-scattering albedo (SSA) profiles: Profiles of SSA have been constructed from few UAV-borne measurements of aerosol light absorption, and profiles of aerosol scattering coefficients calculated from measured size distributions and Mie theory. Measured relative humidity was then used to convert the calculated dry values at ambient conditions. This approach contains a number of assumptions and potential sources of uncertainties which require further explanation or examination. This concern in particular:*

   (a) *uncertainties of the measured light absorption by the Aethalometer (how was it corrected for scattering, filter loading etc.?)*

   Please not that the uncertainties of the Aethalometer measurements are already mentioned in section 2.1 and the correction method in 2.3. The correction method is based on Arnott et al. (2005) and Corrigan et al. (2006). For clarification, we have extended the description of the data correction and uncertainties of calculated light absorption in section 2.1 and 2.3.

   (b) *uncertainties in the calculated scattering coefficient (What is the error when using PSL refractive index for size distribution inversion? How variable is the humidity growth factor and the resulting hygroscopic enhancement of the scattering coefficient when probing air masses of different origins and thus aerosol chemical composition?)*

   Particle sizes from the OPC measurements underestimate in general the real particle diameter the most common atmospheric particles when calibrated with PSL (e.g. Liu& Daum (2000)). Typically the particle diameter is underestimated by approximately 10%. Hence, scattering coefficients calculated by Mie-Theory will be lower for the probed ambient particle population.
   When we use the ambient refractive index, the scattering will to some extend be wrong. Mie-scattering was calculated from OPC measurements close to the

ground for an average refractive index of $m = 1.54 + 0.022i$ from INDOEX (Müller et al., 2003) and for the PSL refractive index $m = 1.59 + 0.0i$. The results were compared to the particle scattering measurements at the surface and the best fit was found for the PSL refractive index (see figures 1 and 2). That is why we decided to use the PSL refractive index. The mean difference between the surface particle scattering and the calculated Mie-scattering was 25% and is considered as uncertainty for Mie-scattering.

Possible uncertainties in the calculated scattering coefficient are now discussed in more detail in Section 2.5, including errors when using PSL refractive index and the possible hygroscopic enhancement.

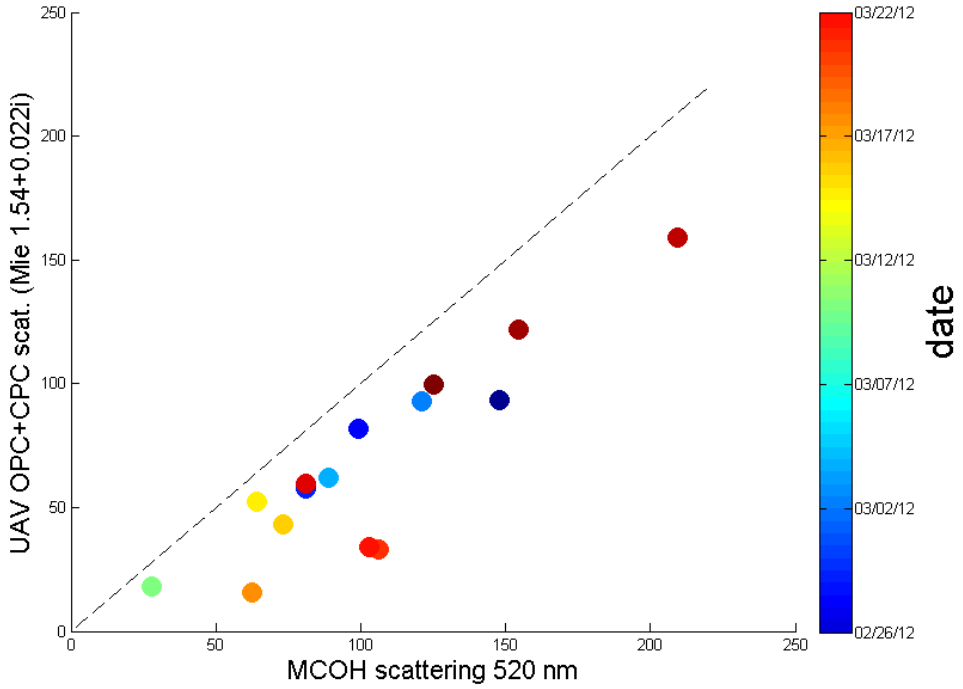

Figure 1: Mie-Scattering with refractive index of $1.54 \pm 0.022i$ vs. surface particle scattering.

(c) *Uncertainties in the vertical profiles of considered properties when inferring them from the lidar measurements and few collocated measurements by the UAV (Which lidar ratios have been applied for the different aerosol types?). In general, the entire method is reasonable, but the single steps require detailed discussion of assumptions and uncertainties.*

The Lidar ratios used are stated in Section 2.6 but are now for clarity repeated

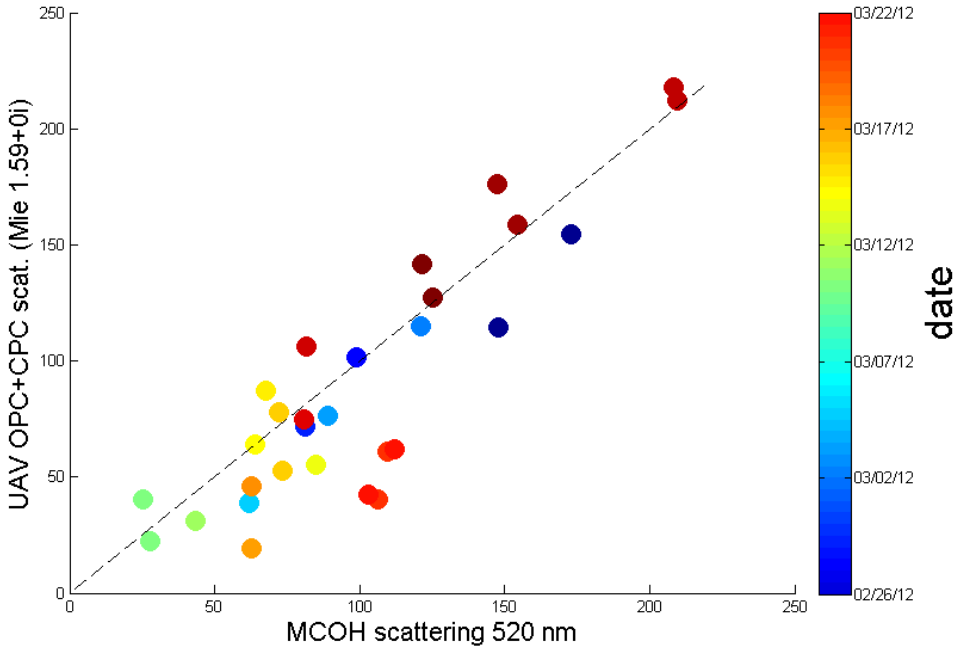

Figure 2: Mie-Scattering with refractive index of $1.59 \pm 0.0i$ vs. surface particle scattering.

in section 3.2.
The possible uncertainties are described in section 2.6.

2. *Evaluation of absorption profiles: The construction and evaluation of absorption profiles inferred from lidar measurements is based on collocated UAV measurements combined with calculating lidar absorption coefficients from particle number concentrations. The correlation used here is derived from surface measurements. However, it is well known that aerosol in the free troposphere is decoupled from the surface. Thus this step requires an in-depth discussion of the approach and of related uncertainties. In particular, statement is expected whether or not the proposed methodology for constructing vertical profiles of absorption coefficients have worked. Looking at Fig. 8 a discussion of statistical significance of the regression analyses is required.*

   The problem of a decoupled boundary layer is already mentioned in the manuscript (Section 3.1.3, 3.2.3). However, we now emphasize this issue in various sections of the revised version.
   A discussion of statistical significance and a statement regarding the robustness of the methodology has been added to section 3.2.3.

3. *The determination of mass absorption efficiency requires a detailed analysis of systematic errors. In the presented approach the MAE values have been determined against NIOSH as the thermal-optical reference method for EC dtermination. How-*

ever, IMPROVE and EUSAAR2 protocols are widely agreed for determining EC from filter samples, which differ signifcantly from NIOSH. The presented MAE values have to be discussed in this context. What MAE values could be expected when applying other thermal protocols than NIOSH?

Generally we agree with the reviewer that there may be discrepancies among the different OC/EC isolation techniques. There has been a continuous development of these methods, trying to overcome the inherent artifact of pyrolysis during the initial phases of the program, with new procedures presented with regular intervals (e.g., Hadley et al., 2008; Boparai et al., 2008). There is thus no final version of this approach.
However, we respectfully disagree with the reviewer's notion that the Birch and Cary (1996) NIOSH protocol is somehow obsolete as one of the standard protocols for OC/EC analysis. This is for instance evidenced by the 100+ citations of this reference since January 2014 (800+ citations in total, source: Web of Science). However, we do acknowledge the relevance in comparing different approaches. For the present study, the EC used for the MAE-calculations is taken from Bosch et al. (2014). Unfortunately, re-analysis using other techniques (e.g., IMPROVE or EUSAAR2) is not possible, since the filter samples have been used up for filter area-intensive isotopic analysis. It is therefore difficult to assess the associated uncertainties propagated into the MAE values for these samples.

4. *Presentation of results in its abstract, the manuscript promises vertical profiles of aerosol optical properties over the Indian Ocean for different air masses or aerosol types, respectively. Although the material is available the presentation of the results makes it difficult for the reader to extract the key pieces of information and to assess respective assumptions and uncertainties. The following structure for the results section starts with methodological part and finishes with air mass specific results. This structure may improve the presentation of the material:*
Section 3.1 Evaluation of vertical profiles, including in-depth discussion of uncertainties
Section 3.2 Absorption values and MAE
Section 3.3 Air mass classification
Section 3.4 Aerosol optical properties for probed air mass types, including comparison to earlier observations

We appreciate the reviewer's concerns of insufficient clarity of the results section but we think the presentation of the meteorological conditions and air mass sources of the field campaign combined with general results fits better in the beginning of the results section to get a good overview. Therefore, we have chosen to leave the main structure of section 3 unchanged.
However, we have clarified the key points, including assumptions and uncertainties, in the beginning of Section 3.2.

Minor issues:

1. *Please add instrument models to Table 1*

   Instrument model numbers have been added to Table 1.

2. *In Table 3, units of properties should be given*

   The unit for the absorption coefficient has been added to Table 3 and Table 4 as well.

3. *In Fig. 3 harmonized color-coding should be used for all displayed data*

   Displaying the AOD without color-coding was intentional since the AOD gives information about the total atmospheric column but the air mass sources were usually different in the boundary layer and free troposphere. Hence, an explicit color-coding for AOD is not possible.

References:

Birch, M.E. and Cary, R.A. (1996) Elemental Carbon-Based Method for Monitoring Occupational Exposures to Particulate Diesel Exhaust. Aerosol Science and Technol. 25, 221-241.

Boparai, P., Lee, J., Bond T.C. (2008) Revisiting Thermal-Optical Analyses of Carbonaceous Aerosol Using a Physical Model. Aerosol Science and Technol. 42, 930-948.

Bosch C, Andersson A, Kirillova EN, Budhavant K, Tiwari S, Praveen PS, Russell LM, Beres N, Ramanathan V, Gustafsson (2014) Source-diagnostic dual-isotope composition and optical properties of water-soluble organic carbon and elemental carbon in the South Asian outflow intercepted over the Indian Ocean. J. Geophys. Res. DOI: 10.1002/2014JD022127

Hadley, O.L., Corrigan, C.E., Kirchstetter, T.W. (2008) Modified Thermal-Optical Analysis Using Spectral Absorption Selectivity To Distinguish Black Carbon from Pyrolized Organic Carbon. Environ. Sci. Technol. 42, 8459-8464.

Liu, Y. and Daum, P. H. (2000): The effect of refractive index on size distributions and light scattering coefficients derived from optical particle counters, J. Aerosol. Sci., 31(8), 945–957, 2000.
Müller, D. Franke, K. Ansmann, A., Althausen, D. (2003): Indo-Asian pollution during INDOEX: Microphysical particle properties and single-scattering albedo inferred from multiwavelength lidar observations. J. Geophys. Res. 108, DOI: 10.1029/2003JD003538.