

## ***Interactive comment on “Optical properties of humic-like substances (HULIS) in biomass-burning aerosols” by A. Hoffer et al.***

**A. Hoffer et al.**

Received and published: 31 July 2006

Comment by reviewer: Section 2.2. The size distributions of the HULIS particles were measured by a SMPS system. The authors give a literature-based sizing accuracy of about 3 % and a particle number concentration uncertainty of about 10 %. Here a figure showing the quality of the SMPS measurements would be helpful. The size distribution measurements were used for the iterative determination of the refractive index based on Mie calculations (Sec. 2.3). Since Mie calculations are very sensitive to the actual particle size, the question raises how the uncertainties in the SMPS measurements evolve in the determination of the refractive index. The authors give in Table 1 the results of the real and imaginary part of the refractive index with uncertainties which are astonishingly small in this respect.

Reply: The parameters necessary for the calculation of the optical and physical prop-

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erties of the generated HULIS particles were measured 5 times for the daytime HULIS and 4 times for the nighttime HULIS. Each measurement includes the determination of one size distribution and of the averaged values of the nephelometer, PAS and TEOM signal. Table 1 shows the average values of the index of refraction calculated for the measurement points. In order to estimate the uncertainty of the calculation of the index of refraction we performed a sensitivity study where different input parameters (scattering and absorption coefficient, the number concentration and sizing accuracy) were individually varied, and the so obtained index of refraction was compared with the original one. For these estimations we used 5% uncertainty in the measurement of the absorption and scattering coefficient, and 10% uncertainty in the determination of the number concentration, furthermore 3% uncertainty in the sizing accuracy. The measurement errors in the real part of the index of refraction were 3.7 % and that in the imaginary part were 12.5 %. Table 1 has been corrected according the error estimations.

A figure showing the SMPS size distributions is included in the revised manuscript.

Comment by reviewer: Section 2.2. The nephelometer results were corrected for angular truncation errors based on the Mie calculations. Now, for the small sizes of the HULIS particles the angular truncation can be neglected, but the systematic error induced by nonlambertian light distribution within the nephelometer might be significant (at least for the TSI3563 nephelometer as discussed in Anderson et al. 1996). The authors should address to this point.

Reply: Assuming that the angular radiation characteristics of the light source of the Radiance Research 903 nephelometer is similar to the TSI 3563 nephelometer we can estimate the non-Lambertian error to be about -4% for the HULIS particle size distributions encountered here (geometric mean volume diameter ~140 nm; Anderson and Ogren, 1998) and this uncertainty results 1 % error in the calculation of the real part of the refractive index. Therefore the non-Lambertian effects are neglected for the purpose of this study

Comment by reviewer: Section 3.1. The authors speculate on pages 6 and 7 that the discrepancy between the imaginary refractive indices measured by the photospectroscopic and the on-line method might be due to water uptake by the nigrosin particles. Since water uptake might also change the real refractive index, the explanation seems to be inconsistent with the good agreement found for the real refractive indices. Some Mie calculations might be helpful in this context.

Reply: If water absorbed on the particles the real part of the particles slightly decreases since the index of refraction of water is  $1.33+0i$ , but the imaginary part decreases dramatically due to the 0 imaginary part. Here we estimate the effect of residual water on the real part: Assuming volume mixing it can be written:  $(1-f)0.26=0.18$ , where 0.26 is the imaginary part of the nigrosin according to the literature, 0.18 is the measured imaginary part. The volume fraction of water is  $f=0.31$ . Again assuming volume mixing we can now estimate the effect of water on the real part from  $n=(1-f)1.67+1.33f=1.565$ . The difference between the estimated (1.565) and measured (1.608) value is 2.7 %, which is less than the estimated error (3.7%) for the real part calculation.

Comment by reviewer: Section 3.2, first sentence: "Figure 3 shows the absorption spectra of HULIS isolated from day and night samples.". This seems to be inconsistent with Section 3.1 where only one nighttime sample was analysed with the spectrophotometric method. If indeed more samples have been analysed what is the reason for deducing the imaginary refractive index only for one nighttime sample?

Reply: For the HULIS isolation we combined several filter-samples in order to have sufficient HULIS mass for the planned analysis. We obtained 6 daytime (labelled as A1, B1, C1 and A2, B2, C2) and 6 night-time HULIS samples (labelled as D1, E1, F1, D2, E2, and F2). The samples A1, B1, C1 and D1, E1, F1 were combined into one daytime and one nighttime HULIS sample, respectively. These samples were used for particle generation and the subsequent measurement of the optical parameters, size distribution and particulate mass (on-line method). The samples labelled with 2 were planned to use in other experiments.

To measure the absorption with the spectrophotometric method, we used the sample B2 and E2, in 4 cm-cell. To calculate the imaginary part of the index of refraction from the spectrophotometric method, the concentration of the HULIS solution should be known. The concentration of the HULIS solution was known only for the E2 sample (containing the HULIS extracted from 7 different night-time filter samples). Since the HULIS concentration was not known in the solution labelled as B2, the calculation of the index of refraction was not possible.

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 7341, 2005.

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