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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling the optical properties of fresh biomass burning aerosol produced in a smoke chamber: results from the EFEU campaign" *by* K. Hungershöfer et al.

K. Hungershöfer et al.

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We are very thankful for the thoughtful and comprehensive comments by the reviewer. Our replies to the comments are given below. Text changes compared to the ACPD version are also indicated.

Comment 1: 12660, lines 3-10 It would be helpful if the authors could provide more information regarding the two fuels they focus on, such as their proper taxonomical names, specific location of their origin, and components of the plant that were burned. For example, was the 'African savannah grass' a single species, or a mixture of several, and if so, which ones? Also regarding fuels, the manuscript also discusses several other fuels besides the two that the majority of the results concern. Either additional

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results/figures/information should be provided for these other fuels, or the discussion in this manuscript should be strictly limited to the two African fuels. If the authors wish to retain these additional fuels, they should explicitly give the reasons why the optical closure method they describe could not be attempted (or did not work?) for those fuels, rather then simply stating that they are limiting their discussion to those experiments with a 'maximum of information'. Finally, please clarify what is meant by the phrase 'largest source of biomass burning' in S5842 line 9 of the same page. Is this based on mass of carbon emitted?...area burned?...etc.

Response: We decided to limit the discussion to the two African fuels and provide additional information for these two cases. The revised manuscript now reads:

Here we concentrate on two EFEU experiments, namely the combustion of savanna grass (Poaceae: Heteropogon spec.) from Etosha, Namibia and the combustion of musasa (Brachystegia spiciformis) hardwood from Zimbabwe. During the combustion of the savanna grass, a few acacia twigs (Acacia spec.) were added for enduring combustion. The two African fuels were chosen, because (1) not all of the experiments provided data on all of the parameters needed for the analysis presented here, (2) and tropical Africa contains about two thirds of the savanna regions worldwide, and (3) savanna fires are the largest contributors to biomass burning in terms of burned biomass (Hao and Liu, 1994; Andreae et al., 1996).

Comment 2: 12661, line 19 The term biofuel may be a bit misleading as it tends to be applied to biomass burned for specific heating or cooking purposes. Using the term 'fuel' would be consistent with the language used elsewhere in the manuscript.

Response: The term 'biofuel'; was replaced by 'fuel'.

Comment 3: 12661-12663 The description of the experimental setup would be greatly aided by including a figure or schematic. Also, section 2.1.1 could be combined with the introductory paragraph that begins section 2.1 to eliminate the need for the sub-subsection and shorten the manuscript.

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Response: A figure showing the experimental setup was added and the introductory paragraph at the beginning of Section 2.1 was merged with the introduction to Section 2 so that the use of a sub-subsection became obsolete.

Comment 4: 12662, line 8 How were the SMPS and APS data merged? (chop off the APS below 0.81? or was some standard or reference method used?)

Response: Text was added to explain this issue in more details as shown below.

old: For the two experiments considered here, the SMPS and APS data were merged without any adjustments.

new: To merge the SMPS and APS measurements, the SMPS data was used up to 0.81 um and extended to larger diameters by the APS measurements without any adjustment. This is justified by the fact that no significant shift was observed between both size distributions indicating effective particle densities near unity (Schmid et al., 2007).

Comment 5: 12662, line 20 Were nephelometer non-ideality corrections made based on an assumed distribution or measured size distributions?

Response: The measured size distribution was used and this fact was included in the text.

Comment 6: 12664, line 17 The justification for assertion that the sample contained 'compact particles' is given much later in the manuscript (use of SEM images) and should at least be mentioned here.

Response Text was changed.

old: We note that application of Mie theory requires particle sphericity, which is a reasonably good assumption for the compact particles encountered here.

new: The assumptions of particle sphericity (here: compact particle shape) andinternally mixed particles were justified by scanning electron microscopy (not shown). In

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addition, Schneider et al. (2006) found dynamic shape factors of less than 1.2 for similar burns, which also indicates compact particle shape.

Comment 7: 12665, line 4 Please provide some quantification width of the distribution (such as geometric standard deviation) to put the term 'very broad' in context.

Response: We added the characteristic parameters for the trimodal lognormal fit (including the geometric standard deviation) both in the text and in Table 1.

Comment 8: 12665, line 9 Could these coarse particles possibly originate from dust or dirt present on the fuels? Were they cleaned or rinsed prior to combustion?

Response: The text was changed as indicated below.

old: Since coarse mode particles of crustal origin can be excluded, these large particles might be un-burnt parts of the biomass.

new: Although no attempts were made to remove dust or dirt from the fuel prior to combustion, we consider the coarse mode particles to be unburned parts of the biomass (char), since the carbon content of the coarse mode particles was similar to the fine mode particles (Iinuma et al., 2007).

Comment 9: 12665, lines 16-18 I was confused by this section. The Rissler et al. (2006) data shown in Fig. 1 extend to about 2 um, not 0.85 as stated in the text? Is this a fit based on data measured up to a smaller value? If this is the case the Rissler data do not necessarily prove that these coarse mode particles were not present in Brazil.

Response: For the SMOCC size distribution shown in Figure 1, the lognormal parameters for the Aitken and accumulation mode given in Table 2 of Rissler et al. (2006) were used. Based on their table caption, we assume that this fit is based on DMPS measurements that cover a particle diameter range from 0.003 um to 0.85 um. It is not our intention to claim that no coarse mode particles were present in Brazil, but that a comparison between our data and the data presented by Rissler is not possible for the coarse mode particles since the APS measurements of Rissler were not included in

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the lognormal fit used for the comparison.

In order to avoid the problem regarding the comparison of the coarse mode particles we adopted the suggestion of the anonymous referee #3 (Comment 4) and used a size distribution obtained from measurements in Africa (where the fuels for the considered EFEU experiments came from) instead of the size distribution from Brazil given in Rissler et al. (2006) by the size distribution of the Otavi fire investigated during SAFARI2000. Otavi is located in Northern Namibia and close to Etosha, the origin of the savanna grass investigated here. Hence, the whole paragraph was modified.

new text version: For comparison, the size distribution of fresh biomass burning aerosol (less than one minute old) sampled during the Southern African Regional Science Initiative (SAFARI2000) in a plume over a biomass burning fire near Otavi (Northern Namibia) is also shown in Fig. 1 (Haywood et al., 2003). The distributions were normalized to their peak values to facilitate visual comparison. The Otavi fire consisted of both flaming and smouldering combustion and the area is close to Etosha, the origin of the savanna grass that was used for the EFEU experiment. The tri-modal Otavi size distribution is similar to the EFEU data showing an accumulation and a coarse mode at diameters of about 0.2 and 2 um, respectively, with relative contributions to the total particle concentration that are comparable to the EFEU data. However, in contrast to our results the coarse mode is likely to contain a significant amount of dust (Haywood et al., 2003). In case of SAVA20a and MUSA23a there is an elevated abundance of large accumulation mode particles (D $>$ 0.2um) compared to the Otavi measurements which is possibly due to reduced coagulation in the Otavi data as a result of plume dilution.

Comment 10: 12666, line 3 What is the motivation for using the 11 percent dCO/dCO2 value to discriminate between smoldering and flaming combustion phases? Was this based on observations from this experiment or previous work?

Response: In the review article by Reid et al. (2005) (Part II) a modified combustion

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efficiency (MCE) of 0.9 is used as a threshold value between a flaming/smoldering dominated combustion. This MCE of 0.9 corresponds to a dCO/dCO2 value of 11 percent.

In the revised version of the manuscript we included Reid et al. (2005) as a reference for the 11 percent value.

Comment 11: 12666 general comment Please give the standard error of the mean when presenting the properties averaged over the course of the two experiments, as is done for the mass scattering efficiencies.

The errors were added in the text as well as in Table 1.

Comment 12:12666, lines 23-25 and 28-30 As mentioned earlier, data from other EFEU burns should not be presented unless the authors provide as complete a description of the fuels, locations, and combustion conditions as is done for the SAVA20a and MUSA23a samples.

Response: The results from the other EFEU experiments were removed.

Comment 13: 12667 general comment It would probably be easier on the reader to convert all the times from time of day to the time elapsed since the start of the burn as that is the more relevant time reference

Response: It is true that the time elapsed from the start of the burn (or the measurements) is the more relevant time. Nevertheless, we prefer using the absolute time in order to avoid unnecessary confusion, since the start of the burn and the start of the measurements are not identical and the discussion of the results occurs only in one section.

Comment 14: 12667, line 26-27 Please provide some quantification of the agreement (statistics of the fit, or at least a percent error between modeled and measured optical properties). How sensitive was the fitting procedure used?

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Response: The mean percent difference between the calculated and the measured optical properties and the standard deviation about the mean was added to the text in Section 3.2. For instance, in case of the SAVA20a experiment, the mean percent deviation for the mass scattering efficiency is -1.3% with a standard deviation of ś 1.1%. For the mass absorption efficiency and the single scattering albedo the mean percent deviation (standard deviation) is 1.4 (ś 4)% and 0.5 (ś 0.4)%, respectively. Similar good agreement between modelled and measured optical parameters was found for the second data set, where the first scattering measurement was neglected, because it seems to be erroneous.

It is not quite clear to us, what the reviewer means by 'sensitivity of the fitting routine', but we assume that the reviewer refers to the sensitivity with respect to changes in the fit parameters? The refractive indices represent the best-fit values for increments of 0.01 and 0.005 in the real and imaginary part of the refractive index, respectively.

Comment 15: 12667, general comment Are the values of 1.60 - 0.01i and 1.56 - 0.01i means of the retrieved refractive indices, or the refractive indices retrieved using the mean values of the relevant optical properties? Lines 24-26 make it sound like the later, but the use of the term 'mean effective refractive index' makes it sound more like the former method is used.

Response The values given are the refractive indices retrieved from the mean values of the relevant optical properties. In order to avoid this confusion, we dropped the word 'mean' in this context.

Comment 16: 12669, lines 25-26 Please provide more information regarding the uncertainties (or a reference discussing the uncertainties) of the nephelometer used in the experiments.

The information was added and the sentence was moved to Section 2 as suggested by the referee #4 (see Comment 19).

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old: Concerning the nephelometer measurements, the reliability of the instrument was confirmed with CO2 as calibration gas.

new: The general uncertainties of an integrating nephelometer are discussed in Anderson and Ogren (1996). During the EFEU experiments, the reliability of the instrument was confirmed with CO2 as calibration gas.

Comment 17: 12669, lines 8-10 The fact that TEOM and impactor masses agree does not necessarily prove that there was no loss of semi-volatile material, but points 2 and 3 are strong. The SMPS/APS system may also confirm the validity of the mass measurement. What density was needed to make SMPS/APS volume agree with either of the mass measurements?

Response: Since the reviewer is right by saying that the agreement of the TEOM and impactor masses does not necessarily prove that there was no loss of semi-volatile material, we removed this argument from the text. From the ratio of the TEOM-mass and the SMPS-APS-volume we obtain an effective particle density of 0.72 and 0.80 g/cm3 for SAVA20a and MUSA23a, respectively. This is considerably lower than typically reported effective densities for biomass burning aerosol (1.35 g/cm3; e.g. Rissler et al., 2006). In the modified text we now argue, that we cannot rule out negative biases in TEOM/impactor mass.

Comment 18: 12672 , line 24-29 Please provide some evidence of this either from this study or previous work

Response: In order to find an explanation for the high ECa mass fractions determined for the EFEU samples, test burns were carried out using lignin and humic acid as surrogates for higher molecular weight compounds. These tests showed that only approximately 30% (instead of 100) were classified as OC (Iinuma et al., 2007). It seems that these higher molecular weight compounds simply needed the much harsher second step of oxidation to evolve. Similar results were found from the analysis of biomass burning aerosol from the Amazon region where on average 53% of the EC

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seemed to be high molecular weight organic material with thermal properties similar to BC (Mayol-Bracero et al., 2002).

We changed the text and included the paper by Iinuma et al. (2007) as reference.

For urban aerosol samples, the applied thermographic method yields ECa fractions within the range of variation of various methods, but tends to overestimate the (actual) EC fraction due to e.g. charring of organic matter (see results for lab \#6/method VDI 2 in (Schmid et al., 2001). The reason for the observed inconsistency obtained for the biomass burning aerosol considered here is the fact that high molecular weight substances can be classified by this method as ECa instead of OC (Iinuma et al., 2007). Chemical analysis using capillary electrophoresis/electrospray ionization mass spectrometry (CE/ESI-MS) showed that the high molecular weight organic compounds that caused the high ECa fractions were probably lignin decomposition products emitted directly from the combustion source. Hence, it is possible that the determined apparent elemental carbon fraction consists of the strongly absorbing black carbon as well as weakly absorbing organic matter (i.e. brown carbon). The application of the ECa fraction determined with the thermographic analysis would therefore result in an overestimation of the black carbon fraction and an underestimation of the OC fraction of the model particles.

Comment 19: 12673, line 13-16 Is it possible that ECa is overestimated by the thermal (optical) technique? Some discussion of the uncertainty in ECa values is needed to strengthen the case the authors are making.

Response: Per definition it is not possible that the ECa is overestimated by the thermal optical techniques, since ECa is operationally defined, i.e. it is defined by the method used. However, the ECa methods are known to overestimate the (actual) EC due to e.g. charring effects.

Regarding the uncertainty of the thermographic technique used: An international round-robin test on the analysis of urban aerosol on quartz fiber filters has shown that

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the method applied for the analysis of the EFEU samples yielded ECa fractions that were within the range of variations of the various methods, but overestimated the average ECa value (see results for lab #6/method VDI2 in Schmid et al., Atmos. Env., 35, 2111-2121, 2001). Thus, the determined ECa fraction of the EFEU combustion aerosol might be an upper limit of possible values. Besides, an additional problem arised with the analysis of samples from biomass burning as higher molecular weight compounds, possibly lignin decomposition products, have such a high charring temperature, that they are not pyrolized during the first step. Instead they are classified as EC (Iinuma et al., 2007) - see comment 18.

Comment 20: 12674, line 11 If possible, replace the term 'brownish' with just 'brown&', or, more ideally, with a more quantitative measure of the filter color, such as the absorption exponent.

Response: In the updated version the term brown is used.

Technical corrections: 12662, line 19: data were corrected, not was 12662, line 26: in the case of the not in case of the 12663, line 24: change of OC to 0 to of the OC refractive index to 0 12663, line 26: might be more clear if 'various BC volume fractions'; was replaced with 'various assumed BC volume fractions' 12663, line 27: also change 'the refractive index' to 'the effective refractive index' to be consistent with terms given earlier

Response: Modified accordingly in the revised version of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 12657, 2007.

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