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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sensitivity studies on the photolysis rates calculation in Amazonian atmospheric chemistry – Part I: The impact of the direct radiative effect of biomass burning aerosol particles" *by* L. M. M. Albuquerque et al.

Anonymous Referee #1

Received and published: 8 November 2005

Review of Albuquerque et al., 2005

General Comments:

The manuscript by Albuquerque et al. presents a study on the impact of smoke aerosol on photolysis rates and subsequently on ozone photochemistry in Amazonia. Based on measurements obtained on 19 September 2002 within the LBA-RACCI-SMOCC field experiment the authors use the TUV radiative transfer model to simulate the impact

of smoke aerosol on the actinic flux. AERONET measurements of the total aerosol optical depth, model derived vertical aerosol profiles, and aerosol optical properties derived from AERONET measurements are used as input parameters for the model. The simulated diurnal variation of the PAR irradiance at the surface is compared to corresponding measurements, and good agreement between the measurements and the model results was obtained. Using a trajectory model, the impact of the smoke-modified photolysis frequencies on the surface ozone concentration is studied. The model results are compared to ozone measurements with good agreement between the model and the observations.

This manuscript deals with an important issue in current atmospheric research and employs an innovative combination of model simulations constrained and evaluated with ground based observations. The topic is well suited for ACP and I recommend publication of the manuscript after the following comments are incorporated.

Specifc Comments:

More references should be added in the introduction, especially in the paragraph on page 9327, between lines 5 and 29. These can include (but are certainly not limited to): Crutzen and Andreae, 1990 (effects of biomass burning); Chatfield et al., 2002 (long-range transport); Moraes et al., 2004 (climatic effect of Amazonia biomass burning); Guyon et al., 2004 (biomass burning as the main contributor to aerosol in Amazonia during the dry season).

page 9328, line1 ff: some references should be included for the statement that atmospheric chemistry is modified by the presence of aerosol particles; these can include: Jacobson 1998; He and Carmichael, 1999; Tie et al., 2005.

page 9327, line 6: 'top of canopy' appears twice in this sentence.

page 9327, line 11: please state what is meant by 'mean photolysis rate value', and add a reference for a model, that employs such a treatment of photolysis.

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page 9330, line 4: the actinic radiation is not obtained by expression (1), but rather used in (1), please move 'actinic radiation F(lambda) and' after expression (1).

page 9330, line 14: maybe add the TUV website to the list of references for TUV: http://www.acd.ucar.edu/Science/Models/TUV/index.shtml (even though I do have currently some problems accessing it).

page 9330, line 17: the reference for the solar spectrum is inconsistent. In the text it is given as 'van Hoosier et al., 1998', while the list of reference includes 'van Hoosier, 1996'. I could not access the given ftp-site, so I was not able to decide which one is correct. Please check.

page 9331, line 5 ff: I do not quite understand the construction of the aerosol profiles used for the radiative transfer simulations. As far as I understand, the sun photometers from the AERONET network do not measure the 'smoke aerosol optical depth', but rather the 'total aerosol optical depth'. Certainly, under the conditions discussed in the manuscript, the total aerosol optical depth (AOD) is more or less equal to the smoke aerosol optical depth. Still, it appears as if the total aerosol optical depth used in the model is the sum of the AERONET derived AOD (i.e., the total AOD) and the AOD derived from the other aerosols presented in Table 1. Please clarify.

Figure 4, 5: maybe limit the wavelength range to values relevant for the present study, i.e., wavelengths between 290 and 700 nm. In the article by Procopio et al., 2003, the aerosol optical properties was reported for 12 different situations that vary as a function of AOD. Is this information used in the present work?

page 9331, line 17 ff: Figure 5 does not show the aerosol optical properties as a function of AOD.

Figure 6: maybe add the corresponding value of MODIS derived AOD to the temporal evolution of the AERONET AOD.

page 9332, line 10: change 'or zero.' into 'of zero.'

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Interactive Comment

page 9332, line 11 ff: Are aerosol vertical profiles also available from the aircraft measurements? If so, how do the modeled profiles compare with the measurements?

page 9332: line 17: The reference 'Tonnesen, 2000' should read 'Tonnesen and Dennis, 2000'.

page 9332, line 20 ff: please include some more information on the initial conditions for the chemical simulations. A table containing the initial mixing ratios in and above the boundary layer should be added.

Figure 8: If Figure 8b shows the ratio between the simulated actinic flux with and without consideration of the aerosol, as stated in the figure caption, it should not be negative. Please check.

page 9334, line 1 ff: Please give some information and/or add a reference about the PAR measurement.

page 9333, line 20 ff: Which AOD values are used for the radiative transfer simulations? Are the average values presented in Figure 6 used? This is of particular importance for the comparison of the model results with local observations (Figure 9), when the use average AOD values might not be appropriate.

Page 9334, line 5 ff, Figure 9: The authors explain rapid variations of the measured PAR with the occurence of water clouds, leading to sharp increases and decreases, which are not reproduced by the model, since all simulations are conducted under clear sky conditions. A fully overcast sky leads to a reduced radiation at the surface compared to clear sky conditions. Therefore, it can be assumed that the model overpredicts the measured surface radiation. A broken cloud field leads to strong variations in the surface radiation (as were observed here) and can result in brief periods with enhanced surface radiation. The effect of enhanced surface radiation due to a broken cloud field has been reported e.g. by Lantz et al., JGR, 1996. For most of the day, the comparison between the model results and the measurements reported here is con-

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sistent with the presence of a broken cloud field (Figure 9). Between 18:30 and 19:30 UTC, however, the model underestimates the measured PAR quite substantially over a comparable long time period. It seems unlikely that this is an effect of the broken cloud field. Is there any explanation for this? Please comment.

Page 9334, line 20 ff: The authors conclude that the coupling of the aerosol model to the TUV radiative transfer model is a 'promising strategy for evaluating the influence of biomass burning aerosol on photolysis rates'. Another important piece of information in the modeling effort presented here is the use of measured AOD. I suspect that the agreement between the modeled and the simulated PAR degrades if unrealistic AODs are used in the simulation. The results are probably more sensitive to the use of realistic AODs than to the correct aerosol optical properties. Please comment.

Figure 10: I suggest to reduce the number of photolysis rates shown here. Maybe four photolysis rates are sufficient here, e.g., photolysis of O3 -> O(1D), NO2, HNO3, CH2O -> H + HCO.

Page 9335, line 1: The references for the updates of molecular data '(Orlando et al., 1999a, b, c)' is confusing. In the list of references there is one article with Orlando as a single author (no 'et al.' required), and two references 'Orlando et al.'. The Orlando, 1999, reference deals with reactions of chlorine, are those included in the chemical mechanism?

Page 9335, line 15 ff: the presentation and discussion of the results from the chemical simulations should be extended. Open questions include: How is the ozone mixing ratio measured? Are there other measurements available, e.g., from passive tracers like CO, and if so, how does the model compare with these observations? How are the results from a trajectory model compared with in situ measurements at a fixed location?

Page 9335, line 19 ff: It is suggested that the initial increase in the ozone mixing ratio shortly after 11:30 UTC is due to photochemical production. To me it seems more likely that high ozone concentrations from higher atmospheric layers are entrained into the

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boundary layer when the height of the boundary layer increases, leading to the rise in the ozone mixing ratio. Please comment.

Technical Comments:

The list of references is not well edited. Numerous papers appear in the list of references, but are not mentioned in the manuscript, e.g., Bais et al., 2003, Früh et al., 2000a/b, ... In total I found 16 references that were not cited in the text, but appear in the list.

The reference Graham et al., 2003, cited on oage 9327, line 12, is not clear, there are two corresponding references in the list.

The references for Andreae and Merlet, 2001 (cited page 9333, line 13) and Orlando and Tyndall, 2002 (cited page 9332, line 26) are missing in the list of references.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 9325, 2005.