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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characteristics, impacts and direct radiative forcing of aerosols at the ARM Southern Great Plains Central Facility" *by* M. G. Iziomon and U. Lohmann

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We thank both Referees for their detailed comments on our manuscript. We will improve the paper by implementing their recommendations and integrating additional aspects of ARM data sets. In particular, we will utilize aerosol vertical profile information in the radiative forcing calculations. In what follows, we address the various issues raised by the reviewers.

In response to Referee #1:

General comments- The data set used in Iziomon and Lohmann (2003) is a subset of the data stream utilized in the present study. However, while our past study investigates the optical and meteorological properties of smoke-dominated haze, our new study examines aerosols at the ARM site in their entirety. In particular, the objectives of the

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present study (which are different from our past study) are to (a) characterize and attempt to understand the factors driving the variability of submicron aerosols on various time scales at the site, (b) Examine the properties of dust aerosols about which there is still much uncertainty to date and compare these properties with those of smoke aerosols previously studied and,(c) estimate the net diabatic impact and direct radiative forcing of aerosols at the ARM site. As suggested by the Referee, we will mention Iziomon and Lohmann (2003) in the introduction of our revised manuscript and make the objectives of the present study clearer.

Detailed comments:

1. We will include a discussion about the aerosol measurement errors and uncertainties in the revised version. The plus/minus signs in the abstract as well as pages 2359 and 2364 refer to the standard deviation of measurements (see for example page 2361, line 8). We will make this distinction clearer in the revised version of the manuscript.

2. We will shorten our discussion on aerosol variation (where necessary) and include additional sub-titles to improve readability. As indicated in the manuscript (page 2363), the maritime aerosols sampled during ACE-1 were indeed well-aged as evidenced by a relatively large accumulation mode surface mean diameter (see Quinn et al. 1998, page 16,558). As referenced in our manuscript, the statement that biomass burning and fossil fuels are linked to the degradation of air quality and acid deposition was taken from the IPCC report [see IPCC, 2001: WG1-Summary for Policymakers, p9]. We cited this IPCC statement because it is relevant to the subject discussed in the paper. We reported the case study examined in section 4.2 (Figs. 8 and 9) because it reveals some interesting and unexpected features about the moisture conditions within the boundary layer for a smoky atmosphere. In particular, we observed (in contrast to our expectation) increased water vapor mixing ratio and relative humidity within the lowest 1 km during the smoky period. However, since this case study appears to be out of place here, we will omit it from the revised manuscript.

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3. As suggested, we will extend our analysis on the connection between aerosol concentration and precipitation, and examine the diurnal cycle in aerosol concentrations on rainy and non-rainy days.

4. As noted on p. 2365, we acknowledged that a longer time series would be necessary to conduct a more qualitative trend analysis. However, our emphasis here is to attempt to understand the observed aerosol temporal variability in the light of other measured atmospheric variables.

5. Although the box model utilized here is a simplified expression, it has been updated in many respects, adopted in IPCC (2001) [see p. 322] and has been widely used in literature (e.g. Haywood and Shine, 1995; Kaufmann and Holben, 1996; Hobbs et al. 1997; Sheridan and Ogren, 1999; Lesins et al., 2002). The model has been shown to produce reasonable estimates for the direct shortwave radiative forcing DFR at the top of the atmosphere by both sulfate (Charlson et al., 1992) and absorbing smoke aerosols (Chylek and Wong, 1995). One advantage of using this form of an analytical solution is an explicit dependence on the individual variables determining the forcing. Our approach here is to use local measurements as much as possible in place of the global estimates used by some authors (e.g. Haywood and Shine, 1995; Chylek and Wong, 1995). As suggested, we will provide more details about the radiative calculation. Yes, "Beta" is the fraction of radiation scattered upwards by aerosols (p 2368). It is an integral of the scattering phase function over the upward hemisphere in the plane parallel coordinates and on the illumination directions expressed by the cosine of the solar zenith angle. Wiscombe and Grams (1976) reported the relationship between the hemispheric backscatter fraction and the asymmetry parameter for the Henyey-Greenstein phase function. In this study, we determine the angular dependent upscatter fraction from hemispheric backscatter fraction measurements (see Sheridan and Ogren, 1999). The range of values of the upscatter fraction obtained here is in good agreement with those reported by Charlson et al. (1992), Chylek and Wong (1995), and Hobbs et al. (1997).

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6. The DFR expressed here is the direct aerosol forcing at the top of the atmosphere. The aerosol optical parameters used in the DFR calculation are those measured at 550 nm, similar to Hobbs et al. 1997. Thus the DFR refers to 550 nm. We will reflect this in the unit.

7. We agree with Reviewer #1 that the aerosol optical thickness depends on the height of the boundary layer. The aerosol optical thickness used here is a function of the hygroscopic growth factor, the extinction coefficient and the height of the well-mixed surface layer. We assumed here that the aerosols are primarily present in the near surface mixed layer (Bergin et al., 1996). However, in the revised manuscript, we will utilize vertical profile of aerosols obtained from Lidar and aircraft measurements.

8. The reviewer comment on comparing IPCC global mean anthropogenic aerosol radiative forcing with the total aerosol radiative forcing estimated here is taken. Consequently, we will compare our local estimate with other regional or local radiative forcing numbers. We also wish to add here, that there are papers (such as Schult et al., 1997; Hess et al. 1998), which report on total aerosol radiative forcing.

9. We will ensure that the wavelengths at which aerosol optical properties are measured are stated where currently missing in the text. The wavelenghts used in determining the Ångström exponents are 450 nm and 750 nm (see page 2364, line 22).

10. Page 2371, line 15-24: We will attempt to substantiate this speculation or otherwise omit it from the revised version.

11. We will address the minor comments and improve the readability of Tables 1, 2 and 4 (the problem with the layout of these Tables is editorial as the Tables in the submitted manuscript were well formatted) and Figures 2, 3, 5 and 6.

In response to Referee #2:

General comments- We will incorporate additional segments of ARM measurements. In particular, we plan to include a new sub-section on aerosol chemical composition at

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the ARM site to substantiate our analysis on aerosol variation. We shall also employ LIDAR and aircraft vertical profiles in estimating DFR as suggested. We will shorten section 4.1 and add further subtitles to improve readability.

Detailed comments:

1. Page 2354, line 23: We will include nitrate and ammonium here.

2. Page 2356: The SGP site is influenced by continental polar air from source regions in Canada, pacific maritime polar air that is greatly modified in traversing the mountainous western United States, and maritime tropical air passing over the warm waters of the Gulf of Mexico. As suggested, we will add this information to the site description.

3. Page 2357: We specified the instruments used in measuring Na and Np (see page 2358, lines 14 and 15). The lower diameter for Na is 0.1 micrometer. As stated in the abstract and on page 2358, the optical particle counter also measures size distribution in the 0.1-10 micrometer range.

4. The aerosol sample stream is conditioned to be less than 40% relative humidity and lower than 40°C before it enters the sampling lines, and subsequently into the instruments. A relative humidity control unit conditions the air stream. This sample conditioning provides a reference point where the properties of ambient aerosols are measured.

5. With respect to the optical properties of smoke and dust aerosols, we limited only the size range for smoke aerosols to 1 micrometer (since smoke aerosols are mainly within the size range) but obtained dust aerosols properties from the full size range measured {up to 10 micrometer} (see page 2368 lines 3-4). However, in the revised manuscript, we will consider the full size range for both smoke and dust, with the properties of sub- and supermicron aerosols discussed seperately.

6. As noted on page 2358, line 2, the aircraft measurements of aerosols commenced at the ARM site in 2000. Although our focus here is mainly on surface aerosols, for

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which there is a relatively long-term data, we will look at more cases of aerosol vertical distribution in the revised manuscript.

7. Page 2360, line 3: Convective transport is an issue at the SGP CF (see Barr and Sisterson, 2000).

8. Lines 5, 13, 22: The refereeŠs points on aerosol number concentration, photochemical production of low vapor pressure products and ambient relative humidity are taken.

9. Line 18: As noted above (in response to Referee #1), we will investigate further the diurnal dependence of aerosol concentration on the boundary layer evolution and precipitation.

10. Page 2362: As indicated on page 2361 line 1, the low albedo in autumn can be attributed to absorbing aerosols in relation to agricultural activities. Although biomass burning activities are most pronounced over the site in the summer, agricultural field burning takes place several times a year, both prior to planting sessions and subsequent to harvesting periods. We will substantiate the diurnal variation of the single scattering albedo further. The discrepancy in the values of the single scattering albedo in Table 1 and Fig. 3, where a clear difference is seen between summer and autumn, lies in the fact that Table 1 features the median of daily-averaged values, while Fig. 3 presents the mean hourly values from 1997 to 2001.

11. Line 11-15: The referee remark is noted. We will re-word these statements.

12. Line 20: We will include estimated visibility trend over the years of study at the site.

13. On page 2362, lines 17-20, we report on the spectral dependence of aeorosol total light scattering coefficient, submicrometer scattering fraction and hemispheric backscatter fraction. One application of this is aerosol modeling. Modeling aerosol effects on atmospheric radiation, by solving the radiative transfer equation, requires spectrally resolved aerosol optical thickness, phase function and single scattering albedo.

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In particular the spectral dependence of the single scattering albedo is driven by the spectral dependence of both absorption and scattering. Another relevance of this finding is with respect to sunphotometers multi-channel measurements. For instance, if the backscatter fraction is derived from sun photometer measurements, reasonable estimates could be made of the physcial and optical parameters of the particles.

14. Page 2364, line 7; The referee comment on the agreement between our finding and previous observations on marine and continental aerosols is taken.

15. Line 16-29; this paragraph complements the earlier discussions on aerosol variation at the ARM site and presents representative values for aerosol optical properties and number concentration over the five years period. We will stress this point.

16. Page 2365, line 12-17; Here we highlight biomass burning and fossil fuel as primary sources of light-absorbing aerosols, which show a positive trend at the site.

17. Line 19-29, As indicated above (in response to referee #1), our emphasis here is to attempt to understand the observed aerosol temporal variability in the light of other measured atmospheric variables. Our proposition that the La Nina-imposed drought conditions could have been enhanced by black carbon aerosols is supported by previous studies e.g. Hansen et al. (1997), Ackerman et al. (2000).

18. Page 2367: This case study reveals some unexpected but interesting features (such that the relative humidity in the boundary layer is higher during the smoky period. The essence of this case study is to gain more insight into specific processes. However (as stated above), we shall omit this case study from the revised manuscript, as it seems to be out of place here.

19. Page 2368, line 4: As indicated on page 2358, line 16, the aerosol sample stream is conditioned to be lower than 40 % relative humidity before it enters the sampling lines, and subsequently into the instruments. Consequently, the statements here are not made for ambient conditions.

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20. We used a size cut of 1.0 micrometer for smoke aerosols, since particles in the accumulation mode dominate biomass burning aerosol mass and have the greatest radiative impact on climate in the visible wavelengths. However, as indicated in (6) above, we will consider the full size range in the revised manuscript, with the properties of sub- and supermicron aerosols discussed seperately.

21. Line 10: The referee comment is noted. We will integrate vertical profile information into the radiative transfer calculations.

22. Page 2371, line 1: The high single scattering albedo (0.93) observed in the spring season for smoky conditions coincides with a relatively low å (1.9) and b (0.11), both of which are indicative of the aged character of the smoke aerosols {sometimes originating from Central America, (Peppler et al., 2000)} reaching the SGP CF. Mie theory predicts that as aerosols age and grow by coagulation, their mass scattering efficiency will increase (due to increased particle size and mass). This will in turn enhance the total optical depth and the radiative forcing .

23. Here, we estimate the total aerosol radiative forcing (natural and anthropogenic). Some authors have also reported total aerosol radiative forcing (Schult et al., 1997; Hess et al., 1998). In order to gain more insight into the aerosol burden at the ARM site, we recently examined the frequency distribution of hourly averaged sa at the ARM site. Hourly occurrences of aerosol light absorbption coefficient greater than 5 Mm-1 are indicative of strong anthropogenic pollution. About 11 % of hourly absorption measurements at the ARM site indicate very strong pollution. We will attempt to use this kind of information to contrast natural and anthropogenic forcing.

24. Page 2371, line 7-12 and minor comments: The referee remarks are noted.

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