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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The unique properties of agricultural aerosols measured at a cattle feeding operation" *by* N. Hiranuma et al.

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Response to Reviewer:

General comments: 1) The authors mention "visibility, radiative properties, climate..." in addition to air quality, as motivations for their study -throughout the manuscript- but most of the discussion is focused on the air quality aspects of it and no discussion seem to have been developed for quantifying the potential effects on visibility, radiative properties and climate. Although I agree this might be beyond the scope of such paper, the data-set would be quite useful for estimating the impact on climate and visibility, especially having chemical composition and mixing information in addition to size distributions. Maybe the authors should explain at the beginning that, although this has implication also for visibility, radiative properties and climate, the discussion in this

paper is focused on air quality.

Response: Thank you for this suggestion, the introduction has now been revised as suggested including the statement "....While there are implications for visibility, radiative properties and climate, the discussion in this paper is focused on air quality." Additional changes are specified below.

2) The compositional data and the mixing state information are extremely interesting, but it seems to me that there is little discussion on the implications of these data and findings for either air quality, radiative properties or climate (with the exception of the briefly mentioned effect on hygroscopicity). Maybe the authors could consider expanding a little bit this discussion and explain the possible implications.

Response: The discussion of these data has now been expanded as follows, beginning page 14,435 Line 12:

"...Specific classification of carbonaceous materials in ambient aerosols is essential to assessing the optical properties of atmospheric aerosols, since carbon is the strongest absorber of light in the troposphere [Andreae and Gelencser, 2006; Haywood and Shine, 1995, 1997; Mertes, et al., 2004]. The calculated absorption cross section of particulate carbon ranges from less than 4 to greater than 20 m2/g depending on assumed particle composition and morphology [Fuller, et al., 1999]. In this study, a significant percentage, 33 to 61 percent, of the particles contained brown or black carbon which strongly absorb visible and UV light. The Raman measurements also provide an indication of mixing state which influences direct aerosol properties, optical properties and hygroscopicity, as well as indirect abilities of the aerosol to act as cloud condensation nuclei [Cubison, et al., 2008; Ervens, et al., 2007]. Both scattering and absorption efficiency change with the addition of minor amounts of material to soot particles, and the results are complex and non linear with added quantities [Martins, et al., 1998; Martins, et al., 1998; Mertes, et al., 2004]. Interestingly, Jacobson showed that soot particles coated with purely scattering chemical components can become more absorbing than the already highly absorbing fresh soot [Jacobson, 2000]. In our study, more than half of the particles classified as brown or black carbon by the Raman were internally mixed with other organics or salts. This suggests that particles of this type are even more effective at absorbing sunlight than pure BBC and may have a significant impact on the radiative budget."

3) Although, this might be beyond the scope of the study, additional electron microscope analysis (e.g. SEM) of the collected particles could have helped elucidating the distinction between black and brown carbon and provide useful additional information.

Response: This is an interesting suggestion. In fact we have analyzed a large number of environmental SEM images of particles collected at the feedlot (See Hiranuma et al., 2008, Atmospheric Environment 42 (2008) 1983–1994. That analysis proved useful for visibly differentiating between some particle types, such as crystalline salts and amorphous carbon particles. It was also useful in determining the particulate hygroscopicity. However, that technique did not provide a reliable means for differentiating between black and brown carbon.

More specific comments: 1) Abstract - Line 7: “nominally” what does “nominally” really indicate here? -

Response: We have removed "nominally" from the abstract and have moved our explanation forward in the text to the first use of "nominally". Pg 14,420, line 25 now reads, "At this feedlot, wind direction is consistently southerly, with the rare exceptions during this project noted below. Thus we refer to the southern and northern edges of the facility as the nominally upwind and downwind edges, respectively."

Lines 17-18: “A significant percentage of the organic particles, up to 28 percent, were composed of internally mixed with salts”. Something seems missing before “with” or “internally”.

Response: We thank the reviewer for pointing out this error. It should read as: “A

significant percentage of the organic particles, up to 28 percent, were internally mixed with salts.”

2) Page 14422 - Lines 23-24: “For a spherical particle, the optical diameter is identical to the volume equivalent diameter”. I would guess this would significantly depend on the optical properties of the atmospheric particles with respect to those of the particles used for the calibration of the optical sizer (e.g. polystyrene latex spheres or Arizona dust) even for perfectly spherical particles. Therefore, the use of the term “identical” might be a bit too strong.

Response: The original text was factually correct, but apparently misleading. This sentence has been reworded to clarify. "The optical diameter of a sphere is identical to its volume equivalent diameter."

3) - Line 24: “If we assume the particles are spherical”. I agree with the authors that using this approximation is probably the best one can do, but if the authors have any evidence of the quasi-sphericity of the particles, it might be good to mention it here; if the contrary it true, as it seems to be the case from other parts of the manuscript, maybe the authors should be more candid about the “roughness” of this assumption. Page 14424 Line 14: “. . .irregular shape and density of agricultural dust” see the previous comments; this for example provides some evidence that particle sphericity is only a rough approximation.

Response: As the reviewer notes, assuming the particles are spherical is a typical assumption to make and is simply the best we can do. For emphasis, the text now states that this is a rough assumption.

Lines 21-22: “Nd:YVO_4 diode pumped solid state laser was used for excitation at 532 nm.” Is there a reason for using a green laser? Using a red laser could have reduced florescence interferences. A sentence explaining why the 532nm wavelength was chosen (maybe it was the only available, and that’s fine), might help.

Response: This was the only available laser at the time. As noted in Table 2, we saw very few instances (at most 2%) in which fluorescence was a problem.

5) Page 14433 Lines4-5: change "…Six spectra could not be classified to due high fluorescence signal…" to "…Six spectra could not be classified due to high fluorescence signal…"

Response: Done.

6) Page 14437 Lines 21-25: "While on-site levels of PM10 were extremely high, a large fraction of the coarse particles was rapidly deposited, and thus the impacts of the coarse mode were lessened at the regional level. However, the significance of the fine and coarse modes emitted from cattle feeding operations should be included for accurate assessments at the regional scale." These sentences seem a repetition of what just said a few lines earlier at the beginning of the page, consider removing the repetitions.

Response: This is a good suggestion. Repetition in the text has been removed.

7) Figure 3: The very last point in panel 3 for the urban PAS data (grey full circles) seems oddly low, maybe check this more carefully.

Response: The data in the figure is correct in that it accurately portrays the PAS measured values. Particles larger than 20 μm diameter were not measured at our sampling location in Houston, TX on September 26, 2006. We note that the average particle counts (i.e., in dN/dlogDp) for adjacent days (from September 25 to 29) were also very low, <9x10 x-6 cm-3.

Secondly, while the ability of the PAS to count particles in the largest size bin is subject to measurements uncertainties, the relative differences in large particle concentrations observed at the urban, semiurban, and agricultural sites is clearly demonstrated by the figure.

8) Figure 4: What is the meaning of the bottom part of the bottom panel is not immedi-

ately clear.

Response: At the reviewer's suggestion, we have updated the figure and caption for clarification.

A revised figure is attached. The caption now reads: Figure 4. In 4A, hourly averaged volume concentrations of PM measured by the PAS and the SMPS at the nominal downwind site and by the PAS at the upwind site are shown as solid squares, open triangles, and open grey circles, respectively. In 4B, background corrected feedlot particle concentrations (volume concentrations) of the total particles sampled by the PAS, of particles less than or equal to 10 micrometer diameter, and of particles less than or equal to 2.0 diameter are shown as open squares, solid grey circles, and solid triangles, respectively. Error bars represent measurement uncertainties of plus or minus 17 percent and plus or minus 3 percent for the SMPS and PAS, respectively. Note that 4B is plotted with a break in the y-axis to emphasis the fact that at low concentrations, variability spans several orders of magnitude.

9) In general consider revising the use of acronyms; often acronyms are defined and then used only here and there and not used in other parts of the manuscript; a consistency check might help.

Response: Okay. Acronyms used 3 times or less have been removed and replaced with full words.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14417, 2011.

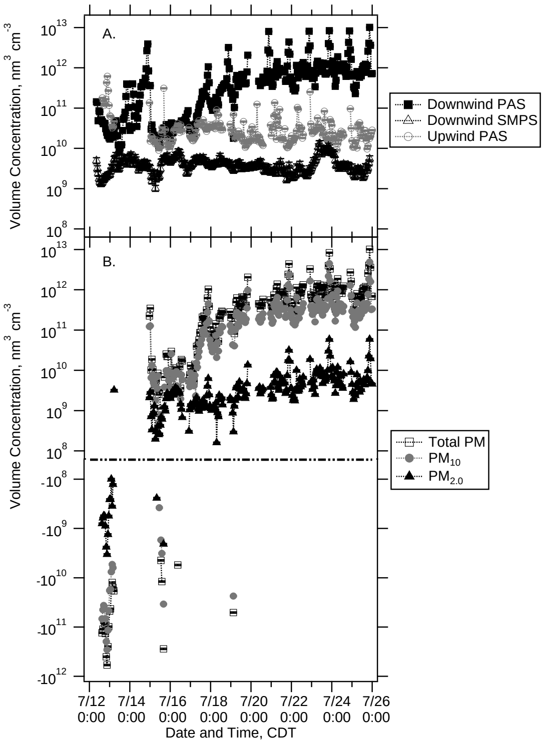

Figure 4.

Fig. 1. Figure 4.