Atmos. Chem. Phys. Discuss., 10, C3974–C3977, 2010
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Optical-chemical relationships for carbonaceous aerosols observed at Jeju Island, Korea with a 3-laser photoacoustic spectrometer" *by* B. A. Flowers et al.

Anonymous Referee #1

Received and published: 14 June 2010

The authors present interesting new observations of the relationship between aerosol chemical composition and optical properties at Jeju Island, Korea during CAPMEX. The data is novel and potentially important. However, there are several weaknesses in the manuscript that need to be addressed before it will be ready for publication in ACP.

Major comments:

-In Section 3.3, the authors jump from the observed correlations between w405 and Åabs(405/532) with OC/SO4-2 and NO3-/SO4-2 to an assumption that organic nitrates are responsible for the observed absorption at low wavelengths. Later in the section they take this leap even further to pinpoint nitrated PAHs as the likely absorbers. No

evidence of organic nitrates or nitro-PAHs existing in the aerosol samples, let alone at concentrations significant enough to cause the observed absorption, is provided. Further, the authors state that “The changes in mass of OC and NO3- are relatively small across the transport episodes; what changes significantly is the relative SO4-2 percent composition. . .”, indicating that, in the absence of a more detailed discussion of the role of SO4-2 (or aerosol acidity?) in the formation of organic nitrates, there is little reason to presume that the concentration of organic nitrates will vary from episode to episode. The authors need choose their language more carefully to reflect the uncertainty in this line of reasoning. It is fine to suggest that organic nitrates may be involved, but it should be acknowledged that there are plenty of other possibilities that cannot be ruled out. A wealth of field measurements and laboratory data on the chemical identity and potential mechanisms of formation of “brown carbon” in atmospheric aerosols has come out since the 1998 modeling study cited by the authors, and these could provide additional insight. For example, NO3-/SO4-2 could be an indicator of aerosol pH. Multiple studies by the Noziere, Iraci, McNeill, DeHaan, and Nizkorodov groups have shown that aerosol salts can react with organics to form light-absorbing organic oligomers and nitrogen-substituted organics (not necessarily organic nitrates), and these reactions are generally pH-dependent.

- The authors infer aerosol size from the PASS-3 data for use in their data interpretation. The analysis presented in section 3.4 would be more convincing if the parameters used were constrained by experimental particle size measurements. Abstracts from the 2009 AGU Fall Meeting on CAPMEX (Venkata Ramana et al., 2009; Kim et al., 2009) stated that aerosol size distribution measurements were made during the campaign; why wasn’t this information used?

- This paper would be improved by a stronger connection of the findings with the literature on field observations of aerosol "brown carbon". The literature discussion started at the end of Section 3.4 should be expanded. One recent study that is of particular relevance and should be discussed is Hecobian et al. (2010).

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- Section 3.5: Did the authors test the possibility that the observed absorption could be attributed to organic aerosol with no soot core? Can the authors provide an estimate of the organic layer thickness that would be necessary to create an "enhancement factor" of 3-6 as observed here?

Detailed comments:

Abstract, line 13: "(an optical model of soot)" is unnecessary and a bit confusing, consider deleting.

Abstract, line 17-18: "Carbonaceous aerosol absorption can alter... substantially; underscoring ..." This sentence is overly general and awkward. Consider changing to something like "The results of this study underscore the need to understand and predict the effects of chemical composition on optical properties."

Page 9373 line 2: "CAPMEX" misspelled.

Page 9376 line 16: consider inserting "possibly associated with aerosol organic material" between "absorption" and "plays"

Page 9378, lines 1-3: it would be much less confusing for the reader if you move the sentence "The lines indicate how w(l) may behave ..." to earlier in the section.

Page 9378, lines 3-5: This sentence is unclear, my suggested revision: "The deviation of the measurements from the modeled curve at 405 nm is consistent with an increase in imaginary index of refraction with decreasing wavelength; this could be due to the presence of absorbing organic compounds."

Page 9738, line 10: no data is presented here for "organic nitrates" (see discussion above) so please remove this from the list.

Page 9378, line 19-20: "an empirical model with idealized assumptions" is a bit vague. Perhaps "an empirical model" is good enough.

Page 9378, line 25: period at the end of sentence missing

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Page 9379, line 5: period at the end of sentence missing

Page 9379, line 7: period at the end of sentence missing

Page 9379, line 19-21: The uncertainties reported seem too small given the number of estimations made here. What are the cited uncertainties based on?

Page 9379, line 28: “should” misspelled.

Page 9379, line 28: “chemistry/transport models” is overly informal shorthand, please change to more formal language

Page 9380, line 7-9: The sentence starting “These empirical. . . “ is awkward and grammatically shaky. To improve, I suggest deleting “that will help develop prognostic treatments for models.”

Page 9380, line 19: The style in which the w values have been listed here is awkward and disrupts the flow of this paragraph. This is just a matter of taste, but you may consider summarizing the findings in words only here, or finding another format for listing the values.

REFERENCES Hecobian et al., Atmos. Chem. Phys. Discuss., 10, 7601-7639, 2010

Kim et al., American Geophysical Union, Fall Meeting 2009, abstract #A13K-07, 2009.

Venkata Ramana et al., American Geophysical Union, Fall Meeting 2009, abstract #A13K-08, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 9369, 2010.