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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 #2

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Some interesting results of aerosol optical properties and chemical compositions are presented via using the ambient measurements from CAPMEX field campaign (August and September 2008). The authors investigated the relationships between the two important aspects of aerosols and showed some potentially important conclusions, highlighting the importance of the impacts of non-elemental carbon fractions (e.g. organic carbon) on aerosol light absorption. However, likely due to the complication of this issue and the limitation of the data, the interpretations and the conclusions have not been sufficiently supported. Several aspects need to be further addressed before it is ready for publication in ACP.

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Major comments:

[1] In section 3.3, authors interpret the negative relationship between w_{405} and OC/SO₄-2, and the positive relationship between $\beta_{sca}(405/781)$ or $\beta_{abs}(405/532)$ and OC/SO₄-2 in Fig. 2 as that increase in OC fraction enhancing short wavelength absorption plays a major role in the reduction of $w(405)$. This interpretation is not convincing since:

* In line 8-10 (p 9396), the authors state “the changes of OC relatively small across the transport episodes, what changes significantly is the relative SO₄-2 percent compositions. . .”. Also shown in Table 2, SO₄-2 changes from 12.3 $\mu\text{g}/\text{m}^3$ in episode #3 to 2.8 $\mu\text{g}/\text{m}^3$ in episode#8, where OC changes only from 1.1 $\mu\text{g}/\text{m}^3$ in episode#3 to 1.5 $\mu\text{g}/\text{m}^3$ in episode #8 with a very slightly increase taking account of uncertainties. It is obvious that the change in OC/SO₄-2 ratio (from 0.12 to 0.56) is mainly due to the decrease in SO₄-2 instead of increase in OC.

* According to equation (2) on p 9372, the change in w_{405} should depend on the changes in both β_{abs} and β_{sca} . As shown in Table 1, β_{abs} (405) in episode #3 is very close to that in episode #8 (no statistic difference, taking account of uncertainties), whereas β_{sca} (405) in episode #3 is a factor of 4 larger than that in episode #8, suggesting that β_{sca} (405) is the main factor influencing the $w(405)$ but β_{abs} (405). It is also realized that the change in SO₄-2 from episode #3 to episode #8 is \sim a factor of 4, coincidentally. As well known, SO₄-2 is a major component contributing to aerosol light scattering (β_{sca}). Therefore, the decrease in SO₄-2 from episode #3 to #8 is likely the main factor for the corresponding decrease in $w(405)$ instead of OC increase.

It is suggested to plot OC vs. β_{abs} with listing the correlation coefficient (R²) to support the conclusion (i.e. OC fraction enhancing absorption). Although OC might play a role in enhancing short wavelength absorption, it is not likely playing the major role in this case (episode #3 vs. episode #8).

[2] Regarding brown carbon: although brown carbon is important in affecting the radia-

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tive forcing in the atmosphere, identifying/characterizing brown carbon and qualifying its optical properties are as not easy as expected. The authors did not provide any review in the introduction regarding brown carbon and its possible formation mechanism (which may help deepen the discussion of the results). There is no definition of brown carbon nor the methods for its identification and quantification provided in the paper. In section 3.5, suddenly, the concept of brown carbon is occurred. In line 19, on p 9378, authors used MAC_{total} to refer the total mass aerosol absorption cross section, which is contributed by the two parts: elemental carbon (EC) and brown carbon (BRC). However, they calculated MAC_{total} by using $\beta_{abs}(mea)$ divided by the total carbon mass ($EC_{mass} + OC_{mass}$). It should be aware of that brown carbon, in terms of chemical refractory, is between OC and EC. It is reasonable to assume that some OC_{mass} are brown carbon, but definitely not all OC mass are brown carbon. The authors need to provide a clear definition of brown carbon for this study and show the steps regarding the calculation of MAC_{total} and MAC_{brc} , otherwise, the results in Table 3 are not convincing.

[3] In the abstract (lines 14-16), authors attribute organic carbon absorption accounting for up to 50% of the measured aerosol absorption at 405 nm for the high OC/SO₄ episode. Again, author mixed the brown carbon mass with total organic mass here. This should be addressed through linking [2] with [3].

Technical comments

- Missing lots of punctuation marks (e.g. period), particularly in the Introduction (most of them are just after these right brackets).
- Since chemical compositions (EC, OC, SO₄²⁻, NO₃⁻) are important components in this paper, Section 2 (“method”) need to include more information, even briefly, regarding filter sampling and analytic procedures for above species. The standard and blank used in those measurements should be also reported. The precision and accuracy (indicated from the results of standard measurements) for each species should be

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included.

- In line 27 on p 9374, should it be “northwest” or “northeast”? It seems that the northeast of Jeju could not be largely influenced by the air mass from southeastern China.

- The text in lines 17-25 (p 9376) may need more work to express precisely what the author try to say.

- It is hard to read the text between lines 13-27 (p 9377). It would be better to re-write it for improving readability.

- In Table 1 or in a separate table, the β_{abs} (532), β_{sca} (532), w (532), β_{abs} (781), β_{sca} (781), w (781) should be listed/reported since these results are used in Figure 2 and 3.

- In Table 3, the percent contribution of EC + coating to β_{abs} (532) in episode #7 may be a typo since the sum of 0.93 from EC+ coating and 0.13 from OC is larger than 100%.

- The title (Optical-chemical relationships for carbonaceous aerosols observed at Jeju Island, Korea with a 3-laser photoacoustic spectrometer) seems not well reflecting the content of the paper since the chemical compositions were not measured via the photoacoustic spectrometer. It may be better to replace “with a 3-laser photoacoustic spectrometer” with “during CAPMEX”.

- It is noticed that this paper is one of the contributions to a special issue on Measurement and Modeling of Aerosol Emissions from Biomass Burning. It seems that there is not much content on biomass burning in the paper. I am wondering how this fits to the scope of the issue.

- A recent ACP paper (Observations of OM/OC and specific attenuation coefficients (SAC) in ambient fine PM at a rural site in central Ontario, Canada by Chan, T et al, Atmos. Chem. Phys., 10, 2393–2411, 2010) is also about study of optical properties

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with chemical compositions via using ambient measurements. Is possible to compare the results from this study with the result from that paper?

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

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10, C4099–C4103, 2010

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