Response to Reviewer Comments for "Optical-chemical relationships for carbonaceous aerosols observed at Jeju Island, Korea with a 3-laser photoacoustic spectometer" by B. A. Flowers, et. al. Atmos. Chem. Phys. Discuss., 10 C3974-C3977, 2010.

Major comments from Anonymous Referee #1

— The possibility of secondary acid-catalyzed reactions producing light absorbing material is an important consideration we did not acknowledge in the submitted version of the manuscript. It is possible to estimate the acidity of the particles arriving at Jeju during CAPMEX by comparing the measured molar concentration of cationic species (NH_4^+ and Na^+) with the amount predicted to neutralize the molar concentration of anionic species present in the aerosol. We have performed this analysis and find the particles neutralized in the bulk. We have added a paragraph to the manuscript describing the particle acidity analysis.

— We have attained the size distribution measurements from our co-authors and have repeated the size dependent analysis of enhanced absorption at 405 nm. This has added significantly to the depth of the analysis and we are grateful to the reviewer for bringing this to our attention. By obtaining the particle size distributions throughout the campaign, we derived wavelength-dependent complex refractive indices that we use in addition with the chemical composition and optical property measurements to perform an optical closure study on the aerosol observed at Jeju during CAPMEX. The details of this analysis are the basis of the considerably re-written manuscript.

— In re-writing the manuscript, we have addressed the place of brown carbon in aerosol light absorption by improving our description of the definition of mass absorption cross sections and delineating the contributions of black and brown carbon to MACs for carbonaceous aerosol. Studies such as those reported by Hecobian et al. (2010) are based on extracting water-soluble organics from sampled aerosol. Our instrumentation measures BC and BrC directly, without using filter-based techniques and off-line analysis. We do note in the manuscript that the particles in CAPMEX are not acidic and have likely not undergone secondary chemical reactions to produce significant amounts of light absorbing material. Instead we suggest particle nitration as one means of increasing light absorbing material in the aerosol as an effect of long-range transport.

— The 3-laser photoacoustic absorption measurement provides for a measurement of soot (at 781 nm) and organic carbon (at 405 and 532 nm) within the aerosol. Because we observe absorption of light at 781 throughout the campaign as well as report measured masses for elemental carbon, we did not consider the absence of soot in our analysis.

Detailed comments from Anonymous Referee #1

— We deleted " (an optical model of soot)" from the text

— We made the recommended change to the text after agreeing with the reviewer's perspective on the sentence.
— We corrected the spelling of CAPMEX.
— We accepted the reviewer's suggestion and changed the sentence accordingly.
— As part of the re-writing of the manuscript, this section has been changed significantly and re-written. Hopefully the confusion has been alleviated.
— We accepted the reviewer's suggestion and changed the sentence accordingly.
— We removed "organic nitrates" from the list.
— We modified the sentence to remove "with idealized assumptions".
— This was an error of the typesetting program. We will check more thoroughly for these issues in the future.
— This was an error of the typesetting program. We will check more thoroughly for these issues in the future.
— See previous two comments.
— Our new version of the manuscript has explicitly described how the uncertainty estimates are established.
— We corrected the spelling of "should".
— The conclusion section has been re-written considerably. The results and their significance have been written and delineated much more clearly after revision.
— We deleted the suggested text.

Major comments from Anonymous Referee #2

[1] The episodic OC/sulfate ratios are listed as the prime metrics for composition in this study. These are for individual episodes where the source of the aerosol is NOT constant across the campaign, hence for the optical properties, each mass/composition must be considered individually. We state the change in OC/sulfate is based on changes in sulfate explicitly in the text and do NOT conclude or imply that changes in OC are the sole causes of changing ω_{405}.
We agree with the referee that single scatter albedo depends on BOTH β_{abs} and β_{sca}, which is why the OC/sulfate ratio, a composition metric describing both absorbing and scattering components, is an appropriate metric for connecting optical properties with chemical composition.

[2,3] We appreciate both reviewers comments on properly introducing brown carbon and mass absorption cross sections. We have re-written the introduction of the manuscript to further introduce brown carbon and its absorption in the context of this paper. We have lengthened our discussion of our definition and calculation procedure for MACs from BC and BrC. We do note in the references cited (Andreae et al. 2006) that no clear consensus exists defining black or brown carbon. We do address the authors comment that 'not all OC mass is BrC' by estimating the MAC for BC, then determining a so-called coating factor, derived at 781 nm, but applied at all 3 wavelengths, which accounts for absorbing material not considered OC. The re-written version of the text clearly defines MAC_{BrC} as "additional absorption than has been accounted for by coating of elemental carbon cores".

Detailed comments from Anonymous Reviewer #2

- We will examine typeset versions of the manuscript more closely in the future to eliminate this concern.
- We have updated the Methods section of the manuscript with a more detailed description of how the chemical composition measurements were made.
- Northeast is indeed the correct description. The back trajectories in this episode were circuitous and these are included in the supplementary information.
- This sentence has been clarified and simplified.
- The description of the Mie code has been amplified and made clearer in the new version of the manuscript.
- The optical properties at 532 and 781 nm are stated and amply discussed in the text. They do not appear in Figure 3 of the new version of the manuscript, and they never appeared in Figure 2 of the original or new version of the manuscript. The values shown in Figure 2 are listed in Table 1.
- This is indeed a typo, for which we are grateful to the reviewer for noticing. The correct value is 0.07 instead of 0.13 and the appropriate change has been made to the Table.
- The 3-laser photoacoustic spectrometer data is new and this is the first PASS-3 data reported from a field campaign.
- Episode 8 is a fire-impacted aerosol transport episode. This paper puts the optical properties of carbonaceous aerosol (from several complex sources, including fires) into context with their chemical composition. Especially when read along side Chakrabarty et. al.'s paper in the special issue, the optical properties reported here contribute to the overall understanding of biomass burning aerosols in the atmosphere. Due to significant re-writing of the manuscript, we have changed the title to a more inclusive statement.
- The paper by Chan et. al. suggested by the reviewer shows results from filter-based optical property measurements and limited discussion on wavelength dependence. We feel it is outside the scope of this paper to compare ambient and filter-based aerosol absorption measurements. We do note, however, that comparisons of this kind are appropriate for a separate analysis and manuscripts.