

Interactive comment on “In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of aerosol coatings” by M. Gyawali et al.

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Response to all Reviewers

The authors would like to thank the reviewers for their critiques and very constructive suggestions. These comments were very helpful and enabled us to improve the quality and clarity of the manuscript. Almost all of the five reviewers shared a common concern regarding the modeling discussions, measurements and analysis (particularly the separation of smoke dominated month and vehicular emission dominated month). We revised our manuscript based on them. We added more discussion and analysis particularly in those sections, and added some clarifications to other sections of the manuscript to make the modeling, measurements and conclusions more relevant and convincing as suggested by the reviewers, without departing from the main issues we raised in the original manuscript. Also, additional papers relevant to our research were published after we had submitted our manuscript. We now have augmented the presentation with reference to some of the results from these newer publications as well as the other relevant papers that were already in existence.

The following revisions were made to manuscript in response to the suggestions of the reviewers. The revisions are given between ►◄ symbols. We will refer to these added/changed items as **AC1**, **AC2**, etc, and use these notations to reply to the particular questions of the reviewers. The reply to the individual reviewer will be sent separately.

AC1: Title

Original Title: In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of aerosol coatings

Revised Title: ►In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non absorbing organic coatings on spectral light absorption ◄

AC2: 1 Introduction

The manuscript was modified on line 28 of page 14061 to add the following sentence that helps clarify the calibration of the instrument.

This sentence should follow the sentence, “Scattering measurements are carried... (Lewis et al., 2008; Mulholland and Bryner, 1994; Rahmah et al., 2006).”

► The instrument is calibrated by using a high concentration of absorbing gas or kerosene-flame soot or laboratory generated aerosol with extinction dominated by scattering (Arnott et al., 2000; Lewis et al., 2008). ◀

AC3: 1 Introduction

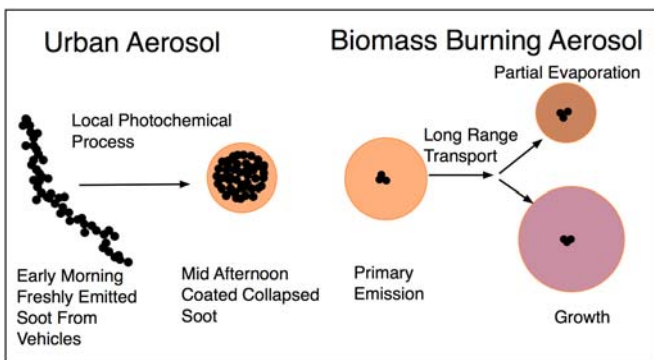
The manuscript was modified on line 9 of page 14062 to add the following sentence that helps clarify the ALAOC.

The sentence should follow the sentence, “In addition to truly light absorbing organic carbon (LAOC), the light absorption of BC can be enhanced by non-absorbing OC that will be referred to as apparently light absorbing carbon (ALAOC),”

► independent of whether the coating absorbs light or not. It is the total effect of the coating on the fractal soot core. Part of the affect the ALAOC may be to collapse the core, thereby changing its absorption. ◀

AC4: 2 Measurements and analysis

The Figure 1 in the revised manuscript was changed to ► Fig. 1 (a) ◀ and a new figure ► Fig. 1 (b) ◀ was added in this section to support the added discussion.



► Fig. 1 (b). Conceptual model of emission and aging of urban and biomass burning aerosol. ◀

AC5: 2 Measurements and analysis

The manuscript was modified on line 23 of page 14062 to add the following description that helps clarify the issue of smoke generation, trajectory measurements, and characteristics of BC particles from different sources.

This description should follow the sentence, “Fig.1 (a) shows a satellite image of... which was the smokiest day of the summer 2008 in Reno, NV.”

► The image shows that Reno is about 100 miles away from the center of the major wild fire sources of July 2008. This smoke source and wind trajectory were similar for much of month of July. The dual wavelength photoacoustic instrument was operated continuously from the starting of July to the end of August, 2008, where the measurements were carried out for 22 days in July and 27 days in August (interrupted for instrument calibration). The measurements were obtained at the Physics building of University of Nevada, Reno, USA. Besides having a couple of minor roads circling the University, two freeways are nearly one mile away from the University. The University is located in the center of the city.

Detailed examinations of BC particles show that they consist of agglomerations of small spherical spherules (Liu et al., 2008). The spherules bind together due to the electrostatic force (Bruce et al., 1991). The mixing state of BC particles changes in the aging process due to condensation, coagulation and photochemical oxidation (Oshima et al., 2009). In this process the coating with water soluble compounds like ammonium sulfate makes them hydrophilic.

Fig.1(b) shows the conceptual model of the aging of the urban aerosol and biomass burning aerosol (especially for primary emission of ponderosa pine-like burning aerosol) in which small BC cores are heavily coated with organics (Lewis et al., 2009). Open chain BC particles from biomass burning becomes closely packed spheres due to water uptake and condensation of inorganic and organic compounds (Hallett et al., 1989). Immediately after emission from the source the BC particles might have internal and external mixing states but with the aging process in the atmosphere the simple shell-core model (described later) is plausible for biomass burning aerosols (Martins et al., 1998) and mid day urban aerosol coated with photochemically generated particulate matter. ◀

AC6: 2.1 Aerosol extinction variation

The manuscript was modified on line 2 of page 14063 to add the following analysis and discussion that helps clarify the issue of bulk analysis of the measurements of biomass aerosol and vehicular emission aerosol for July and August respectively.

This analysis and discussion should be placed at the beginning of the section 2.1

► The extinction (sum of absorption and scattering) variation presented here was the daily average of half hour measurements for the period of July and August 2009 respectively. The photoacoustic measurements of absorption and scattering coefficients have 5% and 15% relative uncertainty respectively (Lewis et al., 2008). The time series of scattering and absorption (half hour average) measurements reached maximum values of 1230 Mm^{-1} and 106 Mm^{-1} at 405 nm and 476 Mm^{-1} and 30 Mm^{-1} at 870 nm on July 10, 2008 and of 154 Mm^{-1} and 18 Mm^{-1} at 405 nm and 52 Mm^{-1} and 7 Mm^{-1} at 870 nm on July 30, 2008 respectively. The scattering and absorption were decreasing between these days. Between July 5 to July 10 the scattering and absorption coefficients were higher for both wavelengths but less than the maximum values mentioned above. Similarly, the time series of scattering and absorption coefficients were maximum at 129 Mm^{-1} and 28 Mm^{-1} at 405 nm and 64 Mm^{-1} and 11 Mm^{-1} at 870 nm on August 05, 2008.

These higher values in early August might be due to some sporadic smoke in the local area, but of much lower magnitude than July. Slightly lower values of scattering and absorption were found from August 11 to 15. For the rest of days these values were significantly less. In Mexico City, due to the diurnal change in the primary aerosol sources and photochemically-generated secondary aerosol, the scattering was maximum several hours later in the day than absorption maximum (Paredes-Miranda et al., 2009). In our study scattering contributed most of the extinction at both 405 nm and 870 nm for both months. The evidence of vehicular emission dominance for the extinction in August was apparent in the aerosol extinction variation (Fig. (2)) with the maximum in the extinction around the morning rush hour (from 6 AM to 11 AM), and nearly symmetrical low values on either side of the maximum for both 405 nm and 870 nm in August. In contrast, morning rush hour made only a small contribution to extinction in July. The extinction starts to decrease from 8 AM and becomes minimum around 1 PM and starts to increase continuously. These variations of extinction at both 405 nm and 870 nm in July exhibited the delaying effect of smoke on the day time boundary layer development (Rissler et al., 2006). ◀

AC7: 2.2 Naive estimate of LAOC: ALAOC

The manuscript was modified on line 25 of page 14063 to add the following sentence that helps clarify the size of BC particle.

This sentence should follow the sentence, “August, which is consistent with the notion that ...coating effects.”

▶ In addition, the BC core size for wood smoke is much smaller than for vehicular emissions. ◀

AC8: 2.3 Single Scattering Albedo (SSA)

The manuscript was modified on line 13 of page 14064 to add the following reference.

The reference should follow the sentence, “This can be attributed to the large proportion of LAOC in wildfire aerosol ...preferentially absorbs radiation to the UV regions”

▶ (Andreae and Gelencsér, 2006) ◀

AC9: 2.4 Ångström Exponent of Absorption (AEA)

The manuscript was modified on line 14 of page 14065 to replace becomes minimum by ▶ is minimized ◀

AC10: 2.6 Simulations

The manuscript was modified on line 12 of page 14066 to replace the section 2.6 Simulations by ►2.6 Simulations and Discussion◄ and with the following discussion and analysis that helps clarify the issue of modeling, the nature of the aging of BC particles and coating of ponderosa pine aerosol. This discussion gives more details, and also takes advantage of new Figures 8 and 9 that are labeled to facilitate discussion. The following added materials should completely replace the existing ones in the original manuscript of section 2.6.

►The optical model of a highly absorbing BC core surrounded by a nonabsorbing shell is suggested for biomass burning aerosols (Martins et al., 1998) and for aged atmospheric soot (Bond and Bergstrom, 2006). However, this model may be unreliable for freshly emitted aerosols and long chain aggregates of BC particles near the source in urban locations. Shell-core model may overestimate aerosol light absorption by less than 15 % in comparison to the random location of soot agglomerates (Fuller et al., 1999). This overestimate is due to the focusing of electromagnetic energy at the BC core due to lensing (Redemann et al., 2001). Laboratory investigations have been conducted to confirm the atmospheric BC aerosol coating effect on absorption (Schnaiter et al., 2005) in which soot particles were coated with secondary organic matter. Schwarz et al. (2008) discussed the use of the shell-core approach for in situ measurements of light absorption in the tropical atmosphere. The coating on BC particles increases with altitude to the lower stratosphere (Schwarz et al., 2008).

Ponderosa pine and other pine species were the typical fuels for the Northern California wild fires in 2008. In primary emissions of smoke from burning ponderosa pine, a mass fraction of less than 1% black carbon and around 99% organic matter was observed, and the black carbon core diameter was typically 50 nm (Lewis et al., 2009). The fractal structure of biomass burning aerosols becomes compacted during atmospheric aging (Chakrabarty et al., 2007) and increasingly spherical due to a coating of secondary species like organic matter, and nitrate or sulfate species from gas to particle conversion (Martins et al., 1998).

In August 2008, local traffic related emissions were the dominated black carbon source. Aerosol absorption measurements in the early morning are dominated by freshly emitted soot particles having low fractal dimension whereas within three hours after sunrise, absorption is due to photochemically-aged soot that is typically more hygroscopic than the fresh soot (Moffet and Prather, 2009). The soot core size after coating is typically around 200 nm (Moffet and Prather, 2009).

The shell-core model for aerosol light absorption and scattering was used to explore the parameter space of AEA relevant to observations. The aim of the simulations presented here was to investigate the theoretically possible variations on AEA for uncoated and coated carbonaceous spheres. The core refractive index was taken to be wavelength independent. For uncoated BC spheres with a complex refractive index of (1.55, 0.8i), AEA for 405 and 870 nm varies from 1.0 to 1.1 and to -0.24, as function of the core diameter as shown in Fig. (7). AEA increases to about 1.11 for a core diameter

of about $0.07\ \mu\text{m}$ and then decreases continuously, reaching a minimum of about -0.24 around $0.72\ \mu\text{m}$ and increases towards its geometrical limit of zero for large spheres.

The effects of non absorbing coatings (refractive index 1.5, 0.0i), and absorbing coatings (refractive index 1.5, 0.012i at 405 nm and 1.5, 0.0i at 870 nm) on AEA are shown in Figs. (8) and (9) respectively. The regions denoted by ‘C’ in these figures represent the aerosol conditions in Reno, and likely other cities, in the afternoon when secondary aerosol mass has condensed on soot cores and compacted them by collapse to a more spherical shape. ‘F’ in these figures represents the core sizes and coatings typical of ponderosa pine fire aerosol. ‘F’ and ‘C’ are representative of the afternoon aerosol conditions in the months of July (smoky) and August (urban aerosol only), respectively. For an identical core surrounded by a spherical coating with refractive index of (1.5, 0.0i), AEA becomes as large as 1.6, even though the coating is non absorbing, for example, in the parameter space near point ‘F’ in Fig. (8). It is noteworthy that point ‘F’ in Fig. (8) coincides with typical BC core and coating diameters observed in ponderosa pine smoke (Lewis et al., 2009). To reiterate, the coating need not be absorbing to give an AEA between 405 nm and 870 nm significantly larger than unity. By contrast, point ‘C’ in Figs. (8) and (9), representative of the parameter space commonly observed for urban soot when coated and collapsed at mid day due to secondary aerosol formation on the soot (Schnaiter et al., 2003), corresponds to AEA significantly less than unity for both absorbing and non absorbing coatings. However, freshly emitted soot in the predawn hours before collapse to a more compact shape is likely to have an AEA greater than or equal to unity (Bergstrom et al., 2007) as observed in Fig. (5).

The relative minimum in the AEA for the typical urban conditions in the afternoon as shown in Fig. (5) is associated with point ‘C’ in either Figs. (8) or (9). In other words, relatively large carbon cores typical of collapsed primary emissions of soot from motor vehicles in cities when coated with either absorbing or non absorbing shells have AEA significantly less than unity for wavelengths 405 nm and 870 nm. By contrast, the smoky month of July, as shown in Fig. (5), has AEA as large as 2, suggesting that the model point ‘F’ in Fig. (9) for the core with an absorbing coating is the correct interpretation for the cause of these observed values of AEA. ◀

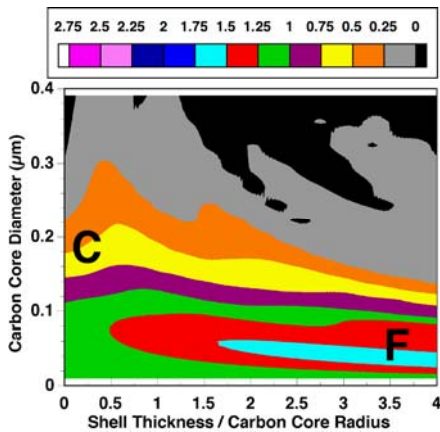


Figure (8)

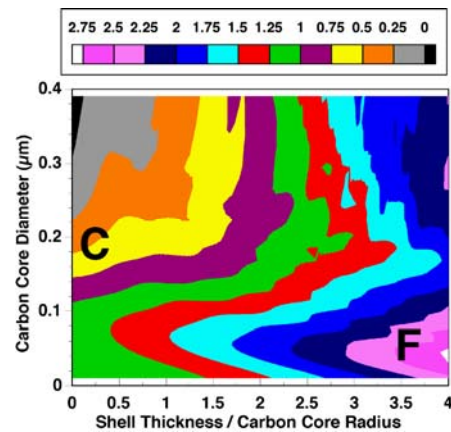


Figure (9)

Figure 8. Simulation of the Ångström exponent of absorption from values at 405 nm and 870 nm as a function of coating thickness and core diameter for a core with refractive index (1.55, 0.8 i) and a coating with refractive index (1.5, 0.0i). In this case, the coating does not absorb light. The regions denoted by 'C' represent aerosol conditions likely on cities in the afternoon when secondary aerosol has condensed on soot cores and compacted them to a more spherical shape. The regions denoted by 'F' represent the core size and coatings typical of ponderosa pine smoke aerosol.

Figure 9. Same as Figure (8), though for a core with refractive index (1.55, 0.8i) and a coating with a refractive index of (1.5, 0.012i) at 405 nm and (1.5, 0.0i) at 870 nm. This coating selectively absorbs light at 405 nm. The regions denoted by 'C' represent aerosol conditions for cities in the afternoon when secondary aerosol has condensed on soot cores and compacted them to a more spherical shape. The regions denoted by 'F' represent the core size and coatings typical of ponderosa pine smoke aerosol.

AC11: 3 Conclusions

The manuscript was modified on line 3 of page 14067 to add the following sentence.

This sentence should follow the sentence, “The simulations confirm that large AEA values are possible even when coatings do not absorb light”,

► especially when the BC core is much smaller than the wavelength of the laser used for aerosol optics measurements ◀

AC12: 3 Conclusions

The manuscript was modified on line 19 of page 14067 to add the following sentence.

This sentence should follow the sentence, “Particle absorption could be in the surface area ...for sufficiently large particles”.

► This work illustrates that aerosol optics can be quite different for vehicular-related and biomass burning-aerosol. ◀

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