

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

M. Gyawali
gyawalim@unr.nevada.edu

Reply to Anonymous Referee #2 (comments for Atmos. Chem. Phys. Discuss., 9, C3414–C3416, 2009 In situ aerosol optics in Reno, NV, USA...by Madhu Gyawali et al)

Please refer to the Author Comments for description of the added materials and for the notations AC1, AC2, etc that are used here in the reply to the particular reviewer questions.

Reviewer comments are given in bold type-face. Our replies are given in plain text.

The manuscript presents the results of optical measurements in the period of two months at a single site. The sampling period is divided into a ‘biomass burning’ and a ‘normal’ month.

Without additional measurement data, this classification seems to be overly simplistic. Having a high time resolution database of optical parameters, one would have expected a more detailed analysis of sampled air masses (e.g. by trajectories, combined with fire locations, intensities, type of biomass burned, etc.) that would have revealed the fine structure of variations of optical properties as a function of various parameters. For lack of detailed analyses, the majority of the conclusions of this ‘bulk’ analysis remains trivial and does not add much to our general understanding of absorbing aerosols.

Reply:

We agree and have revised the manuscript to add some additional description of the measurement. See **AC4**, **AC5**, and **AC6** for revised Measurement and analysis, and Aerosol extinction variation.

No trajectory or chemical analysis was needed to verify the source of the aerosol in July or August. In July, the source was clearly the wildfires in Northern CA based on the wind directions and the intensity of the smoke from this source (more information can be found at [http://en.wikipedia.org/wiki/Summer_2008_California_wildfires], as given in the manuscript). In addition, inhabitants of Reno were acutely aware of the heavy smoke burden as breathing conditions were at times unhealthy. In August, the measurements were similar to those obtained in previous years, and the prevalence of local urban sources dominated in that case. For the purposes of this paper, which concentrates on aerosol optical properties as a consequence of the chemical mixture, we do not need detailed chemical analysis to arrive at our main conclusions. Figure 1(a) shows clearly the vast smoke source from the fires in Northern CA, and their trajectory towards Reno. This figure was representative of the smoke source and wind directions for the month of July. The optical phase diagram in Figure 6 also shows the distinction between the unusual smoky month of July and the more normal month of August.

What is more ‘revolutionary’ in this manuscript is the statement that ‘the organic coating needs not to be intrinsically brown to observe effects commonly referred to as those caused by brown carbon light absorption’ (Page 14067 line 3-4): if proven, this could indeed change our view of brown carbon and the like. But going through the brief modeling section of the manuscript I am not at all convinced that these conclusions are atmospherically relevant and even theoretically well founded.

Reply:

The descriptions of the evidence of huge coating of some biomass burning aerosol have been described in the revised Simulations and Discussion section, see **AC10**. For more evidence see reply below.

It is well known that biomass burning emits BC particles that are very much different from soot particles emitted by internal combustion engines (see current literature for char EC versus soot EC), if we observe different AAE values for biomass burning aerosols than for urban aerosols, our first idea is not that soot particles are covered with a thick layer of semi-volatiles or water, but simply that the optical properties of biomass burning absorbing aerosols are very much different from those of fossil soot (Hoffer et al., ACP 2006). In a biomass plume of smoldering fires it is not soot that is covered with absorbed organics but more char particles or the like. If we make water solution of such biomass burning aerosols in which non-absorbing compounds do not interfere, we can observe similar wavelength-dependence (AAE) than in the airborne state

Reply:

Biomass burning is the major source of particulate organic matter, soot particles, including various salts (Lewis et al., 2009; Li et al., 2003; Martins et al., 1998; Posfai et al., 2003; Russell et al., 2002; Semeniuk et al., 2007). Aerosol from fossil fuel burning and that from biomass burning definitely have different wavelength dependencies and can be distinguished by the Ångström exponent of absorption (Kirchstetter et al., 2004); the former has AEA around one and the latter is 2. The vast difference in wavelength dependence is attributed to the organic carbon (OC) in biomass burning aerosol, if OC is removed from biomass burning aerosols then its AEA is reduced to 1.3 (closer to the BC from vehicular emission), see description on Kirchstetter et al. (2004). BC from flaming combustion (probably the only source of BC in biomass burning) is similar to internal combustion engine BC (see Chakrabarty 2006a, b). The soot particles (the third major carbonaceous particles) from biomass burning might have fairly ordered microstructures in comparison to the soot particles from fossil fuel combustion (Posfai et al., 2003).

I am also not convinced that in the smoke plume BC can be covered with vast amount of adsorbed organics: as the plume cools some adsorption may take place but as the plume mixes with ambient air and becomes diluted, the so-called dilution effect takes place and most of semi-volatiles are actually desorbed (see Grieshop et al., ACP 2009). *There are only the photochemical processes related ageing and secondary*

aerosol formation that can indeed produce some refractory coating that can be optically relevant: but these processes may not be directly linked to emission sources.

Nevertheless BC cores with thick non-absorbing shells still visible under an electron microscope are very rarely observed in individual particle studies. Since the boundary conditions of the model may not seem to be atmospherically relevant, I am skeptical about the conclusions of the second part of the manuscript, too.

Reply: (The italic part is separated from here, see reply below for this)

Some of the description of this issue was also included in the revised manuscript (see AC10). The following details are presented here for more clarification of the specific question.

Ponderosa pine and other pine species were the typical fuels for the Northern CA wildfires in July 2008, this can be inferred from the comparison of monthly averaged optical properties of aerosol for the months of July and August 2008, with that of the laboratory burned fuels (Fig. 6). Lewis et al. (2009) reported that 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. The visible organic coatings of atmospheric particle is a common phenomenon and the visible image of coating have been reported elsewhere; Russell et al. (2002) described the high-resolution X-ray image of carboxylic acids in organic coating, the evidence of particle coating with organic materials in biomass burning aerosols was described in TEM photography by Martins et al. (1998), the coating in young smoke smoldering and flaming fire particles by Posfai et al. (2003), images of sulfate, NaCl and other organics by Semeniuk et al. (2007), the description of the evidence of rapid coating by organic matter in Mexico City with TEM image by Adachi and Buseck, (2008). TEM images of amorphous organic coatings, potassium and ammonium salt particles in young smoke and aged biomass burning aerosols was given by Li et al. (2003). The huge amount of organic coatings in biomass aerosols especially for ponderosa pine burning (similar with our findings in California wild fire aerosols of July, 2009, and modeling discussion) was evident in the TEM image (Fig 9), and the proportions of core and other coating materials (Fig 5a) with the findings of Lewis et al. (2009).

There are only the photochemical processes related ageing and secondary aerosol formation that can indeed produce some refractory coating that can be optically relevant: but these processes may not be directly linked to emission sources.

We agree with the reviewer comment that photochemical process produces the organic coating, but it is not the only process for organic coating in the atmosphere, the soot particles also acquire non absorbing coating of sulfate and organic carbon due to condensation and cloud processing that lead to the absorption enhancement (see for detail M. O. Andreae and A. Gelencsér, 2006, <http://www.atmos-chem-phys.net/6/3131/2006/>). The nature of coating and absorption enhancement may also depend on the emission sources, as described in the previous question. So, these mechanisms can be linked to the emission sources depending upon the situations.

References for the specific reply

Adachi, K., and Buseck, P. R.: Internally mixed soot, sulfate, and organic matter in aerosol particles from Mexico City, *Atmos. Chem. Phys.*, **8**, 6469-6481, 2008, 2008.

Kirchstetter, T. W., Novakov, T., and Hobbs, P. V.: Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, *J. Geophys. Res.*, **109**, D21208, doi:10.1029/2004JD004999, 2004.

Chakrabarty, R. K., H. Moosmüller, W. P. Arnott, M. A. Garro, and J. W. Walker (2006). Structural and Fractal Properties of Particles Emitted by Spark Ignition Engines. *Environ. Sci. Technol.*, **40**, 6647-6654.

Chakrabarty, R. K., H. Moosmüller, M. A. Garro, W. P. Arnott, J. W. Walker, R. A. Susott, R. E. Babbitt, C. E. Wold, E. N. Lincoln, and W. M. Hao (2006). Particle Emissions from Laboratory Combustion of Wildland Fuels: Particle Morphology and Size. *J. Geophys. Res.*, **111**, doi:10.1029/2005JD006659.

Lewis, K. A., Arnott, W. P., Moosmüller, H., Chakrabarty, R. K., Carrico, C. M., Kreidenweis, S. M., Day, D. E., Malm, W. C., Laskin, A., Jimenez, J. L., Ulbrich, I. M., Huffman, J. A., Onasch, T. B., Trimborn, A., Liu, L., and Mishchenko, M. I.: Reduction in biomass burning aerosol light absorption upon humidification: roles of inorganically-induced hygroscopicity, particle collapse, and photoacoustic heat and mass transfer, *Atmos. Chem. Phys. Discuss.*, **9**, 15247-15294, 2009.

Li, J., Posfai, M., Hobbs, P. V., and Buseck, P. R.: Individual aerosol particles from biomass burning in southern Africa: 2, Compositions and aging of inorganic particles, *J. Geophys. Res.*, **108**, doi: 10.1029/2002JD002310, 2003, 2003.

Martins, J. V., Artaxo, P., Liousse, C., Reid, J. S., Hobbs, P. V., and Kaufman, Y. J.: Effects of black carbon content, particle size, and mixing on light absorption by aerosols from biomass burning in Brazil, *J. Geophys. Res.*, **103**(D4), 32,041-32,050, 1998.

Posfai, M., Simoni, R., Li, J., Hobbs, P. V., and Buseck, P. R.: Individual aerosol particles from biomass burning in southern Africa: 1. Compositions and size distributions of carbonaceous particles, *J. Geophys. Res.*, **108**, D13, doi:10.1029/2002JD002291, 2003.

Russell, Maria, S. F., and Myneni, S. B. C.: Mapping organic coatings on atmospheric particles, *Geophys. Res. Lett.*, **29** (16), doi:10.1029/2002GL014874, 2002.

See, S. W., Balasubramanian, R., and Wang, W.: A study of the physical, chemical, and optical properties of ambient aerosol particles in Southeast Asia during hazy and nonhazy days, *J. Geophys. Res.*, **111**, D10s08, doi :10.1029/2005JD006180, 2006, 2006.

Semeniuk, T. A., Wise, M. E., Martin, S. T., Russell, L. M., and Buseck, P. R.: Water uptake characteristics of individual atmospheric particles having coatings, *Atmos. Env.*, 41, doi 10.1016/j.atmosenv.2007.04.001, 6225-6235 2007.