Interactive comment on "Photoacoustic optical properties at UV, VIS, and near IR wavelengths for laboratory generated and winter time ambient urban aerosols" by M. Gyawali et al.

M. Gyawali
madhug@unr.edu

Reply to Anonymous Referee #1(Comments for Atmos. Chem. Phys. Discuss., 11, C8718–C8720, 2011
www.atmos-chem-phys-discuss.net/11/C8718/2011/)

We would like to thank Reviewer #1 for his/her comments on this paper.

Reviewer comments are given in bold typeface. Our replies are given in plain text.

Photoacoustic - measured aerosol absorption measurements at UV, visible and near IR wavelengths are presented for laboratory and ambient aerosols. There is certainly unique data presented, including the use of the new UV photoacoustic instrument. Although there is unique data, the manuscript falls short in a number of ways that lead me to believe major reconsideration is needed. For the most part, the text that exists is well written, however it is very long for the content. The new photoacoustic instrument is a great advance however the details of this instrument are presented in supplementary material. The claim by the authors is that these are the first measurements of PAS UV absorption. I would expect much greater detail in the description of the instrument, how the UV was calibrated and some form of data that shows the UV measurements can be trusted (although this would lengthen the manuscript).

Reply:
We agree with the reviewer and have moved the details of instrument and its calibration to the main paper. More comparisons and analysis of published aerosol optical properties have been added so that the measurements at UV and other wavelengths are shown to be reliable and trusted. We agree the situation is conflicting; we wish to detail the operation of the instrument and correlate data from various sections in the manuscript while simultaneously keeping the manuscript concise.

The laboratory data section does not show any validation of the instrument, rather simply shows that the PAS-measured AEA for soot is 0.8 and for incense smoke is □4. The authors state that these experiments serve as an evaluation of the instrument performance and accuracy. I do not see any mention of either.

Reply:
We agree with the reviewer and removed the following sentence in the revised manuscript: "Use of these aerosols under laboratory circumstances allows for the evaluation of the instrument's performance and accuracy". We compared measured AEA and AES of kerosene with published data (e.g., Sandradewi et al., 2008a, 2008b; Schnaiter et al., 2006; Ajtai, et al., 2010, 2011) in the revised manuscript . The presentation of incense smoke is to demonstrate the enhanced absorption towards the UV region by brown carbon (BrC).

The t-matrix section is disconnected from any other part of the manuscript. The main conclusion is that according to theory, the SSA changes without a change in aerosol mass or composition purely by differences in monomer packing. The authors do not provide any link into why this is important for this paper. I would recommend removing this section entirely. Although work went into the modeling, it serves to distract the reader from the other data.

Reply:
Agree and removed.

The ambient data is adequately presented, however it really feels as though it has been presented as 'first data' without an attempt to provide a reason to the reader as to why it is significant. The ambient aerosol data could likely form the core of a much shorter manuscript.

Reply:
In the revised manuscript we added more analysis and comparison of the ambient data to make it more significant.

The gas phase section also does not connect well with the rest of the manuscript. A lot of data is presented and suddenly this manuscript is describing PM mass instruments, ozone, NO, NO2 levels?
Reply:
The description of PM mass instrument is removed (only the references are given), the description and analysis of ozone data is also removed. NO and NO2 measurements are very important to identify the sources of black carbon and organic carbon and their concentration (See Sandradewi et al., 2008 a, b). We augmented the analysis to explain why these measurements are important to understanding the nature of aerosol during polluted and clean days.

There is speculation that organic nitrate aerosol may form but there is no evidence to the effect, and so this section finishes with seemingly irrelevant data and speculation.

Reply:
As presented in the manuscript, we had on average a substantial increase of PM2.5 nitrate on polluted days (1mg/m3 on clean days to 8 mg/m3 on polluted days). We believe it is relevant to present the data that explain the increase of PM mass concentration during the most severely polluted episodes in this area, with the statement "NH_4NO_3 was the primary cause of the lift in the $PM_{2.5}$ mass concentrations over the 35 μgm^{-3} threshold during polluted days". The valid question concerns the effect on aerosol optical properties, and how we compare this with other similar studies; if it were organic nitrate it should have enhanced absorption towards UV, which we did not observe in this study. Instead, it contributed to the enhanced scattering so the following sentence has been added to the revised manuscript: "NH4NO3 exhibits higher hygroscopic growth, increasing the amount of light scattering and direct radiative forcing by ambient aerosol population (Crosier et al., 2007). The consistently higher values of absorption coefficients at all wavelengths during polluted episodes can be attributed to dominance of NH4NO3".

Overall, I feel as though this paper is a collection of data that is poorly tied together and doesn't really tell the community a relevant story. That relevant story may be contained within the data, however this manuscript doesn't allow that to come out. The laboratory, t-matrix, ambient gas and ambient aerosol data all feel like separate tenuously connected pieces of work. The middle two, I think, should be eliminated.

Reply:
As mentioned before we removed the T-matrix section, description of PM instrument, and ozone description. In our opinion, the presented data and analysis in the revised manuscript tell the relevant story.

I would recommend the authors consider removing many of the poorly connected sections, and re-evaluate whether this is a 'proof of instrument' paper, a robust analysis of aerosol optics for a specific location or otherwise.

Reply:
We have strongly connected various sections in the revised manuscript: PM size distribution with the AES (diurnal; variation angstrom exponent of scattering), gaseous data to infer the source contribution of aerosol emission and finally to the AEA (angstrom exponent of absorption).
While we do wish to stress that we consider our instrument proven, we also emphasize that the data collection and analysis result many significant findings regarding aerosol optical properties.

In this reviewers opinion, a concise manuscript describing the measurements is possible with some careful editing and removal of excess sections /text. I cannot recommend publication until this concise story is presented.

Reply:
As mentioned above, we edited/ removed the excess sections and augmented the gases section to make it more connected with the aerosol optical properties. The outcome form these additions is also reflected in the conclusion section of the revised manuscript. We include UV-VIS-IR wavelength aerosol optical properties measurements including aerosol size, chemical speciation and gaseous data, and meteorology. We believe the revised draft serves as a concise manuscript without sacrificing any information needed to understand our instrument, its operation, laboratory measurements, and finally the description of severe temperature inversion with meteorology data. We ultimately connected these data to aerosol light absorption and scattering.

References:

Crosier, J., Allan, J. D., Coe, H., Bower, K. N., Formenti, P., and Williams, P. I.: Chemical composition of summertime aerosol in the Po Valley (Italy), northern Adriatic and Black Sea, Q. J. R. Meteorol. Soc. 133: (S1) 61-75, doi: 10.1002/qj.88, 2007.

Sandradewi, J., Prévôt, A. S. H., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E., and Baltensperger, U.: Using aerosol light absorption measurements for the quantitative de- termination of wood burning and traffic emission contributions to particulate matter, Environ. Sci. Technol., 42, 3316–3323, doi:10.1021/es702253m, 2008a.

Sandradewi, J., Prévôt, A. S. H., Weingartner, E., Schmidhauser, R., Gysel, M., and Baltensperger, U.: A study of wood burning and traffic aerosols in an Alpine valley using a multi-wavelength Aethalometer, Atmos. Environ., 42, 101–112, 2008b.

Schnaiter, M., Gimmler, M., Llamas, I., Linke, C., Jäger, C., and Mutschke, H.: Strong spectral dependence of light absorption by organic carbon particles formed by propane combustion, Atmos. Chem. Phys., 6, 2981–2990, 2006, http://www.atmos-chem-phys.net/6/2981/2006/.