Atmos. Chem. Phys. Discuss., 11, C6827–C6831, 2011
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Origins and composition of fine atmospheric carbonaceous aerosol in the Sierra Nevada Mountains, California" *by* D. R. Worton et al.

Anonymous Referee #1

Received and published: 24 July 2011

This manuscript describes results from gas and particle measurements made during the two-month long BEARPEX campaign in 2007. The measurements were made at a remote location in the Sierra Nevada Mountains, although it received transport from urban locations, as well. A suite of measurements are presented, including primary and secondary gas-phase species as well as AMS measurements and SOA molecular markers measured by the TAG instrument. Factor analysis is performed using the TAG data and three factors are identified, which together explain 77% of the variance in the total organic aerosol concentration. Overall, interesting diurnal and seasonal behaviors are observed at the site. The manuscript is well written, and well-organized, and is recommended for publication after the following comments are addressed:

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Overall Comments:

The identification of Factor 3 (Local light-driven emissions and oxidation products) seems highly uncertain. First, there are only two markers used to identify this Factor, compared to 10 each for Factors 1 and 2. Second, both of these compounds are assigned a 'medium' 'Uncertainty of compound identification'. As the authors state, most condensed-phase isoprene oxidation products are not detected by the TAG – how much uncertainty is there in the identification of this factor, and assigning its source as oxidation products of light-driven emissions? To what extent does this factor fully represent all of the 'Local light-driven emissions and oxidation products' in the aerosol?

Factor 2 is identified as Oxidation products of temperature-driven local biogenic emissions, yet this factor appears to be anti-correlated with temperature: this seems very counter-intuitive. Additionally, Figure 4 indicates that pinonaldehyde concentrations were higher during the cold period than during the warm period, supporting the anti-correlation with temperature. How are these derived from local temperature-driven emissions, then? One brief sentence is given to explain this behavior, with the explanations being either nighttime oxidation, or transport of aged biogenic emissions. Although similar monoterpene diurnal profiles were observed in Bouvier-Brown et al. (2009), significantly more discussion is needed. There are a number of studies that report higher nighttime monoterpene concentrations that appear to be due to a mechanism that is not temperature-driven (e.g., Simon et al. (1994); Hakola et al. (2000); Janson et al. (2001)). Could something similar be happening here?

It is surprising that levoglucosenone, rather than levoglucosan, is used as one of the tracers for biomass burning emissions? Can the authors discuss this point? In the Williams et al. (2006) paper, levoglucosan is identified as a compound that the TAG can quantify – was levoglucosan correlated with levoglucosenone (especially during the periods of highest biomass burning influence)? If it is hypothesized that levoglucosan underwent oxidation during transport (as several recent studies suggest), then providing levoglucosan/levoglucosenone ratios would be extremely interesting.

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The discussion and results of the organosulfates adds little to the manuscript. I.e., the quantified organosulfates contributed less than 1% of OA mass, yet substantial discussion (and 2 of the 8 figures) is dedicated to this topic (Section 3.7). Figure 7 has very little practical value – it could be removed from the manuscript, or at the very least should be moved to the supplemental. Another manuscript (Worton et al., (2011)) is promised on this topic: Figure 7 is out of place here, but may fit better in that paper.

Since the finding of a correlation between OA and CO is significant here (and is given appropriate discussion in the manuscript) move Supplemental figure S3 from the supplemental to the main paper.

Specific Comments:

1. Provide the actual measurement dates in the abstract

2. Abstract, line 16: Methyl chavicol is not well known to many in atmospheric chemistry: a brief clarifier here identifying its source(s) would be helpful

3. Abstract, line 17: "…during both identified meteorological periods." At this point, the periods have not been defined, and they are only vaguely defined later in the abstract.

4. Abstract, pg. 17073, ln. 28: "…evidence for the likely importance of aerosol sulfate in spite of neutralized aerosol." Perhaps the neutral aerosol is the reason for the low contribution of organosulfates to the total OA? (comment applies to Section 3.7 as well)

5. Pg. 26, ln. 26: the Robinson et al. (2007) and de Gouw et al. (2011) references should not really be put in the same group as the Goldstein and Gallbally (2007) reference, since the Robinson et al. (2007) and de Gouw et al. (2011) studies identify IVOCs and SVOCs collectively as SOA precursors.

6. Pg. 17097, ln. 20: provide inlet height

7. Pg. 17080, ln. 18: is sample volume not 0.81 m3?

8. Pg. 17085, ln. 21: "A small discrepancy (∼20%) might be expected from the differing

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size cuts. . .” citation needed

9. Pg. 17086, ln. 4: “. . .30% non-fossil” seems reasonable, but provide a citation

10. Pg. 17089, ln. 19-20: “. . .which may temper the statement that most of the OA is derived from biogenic sources.” Not necessarily: lifetimes of gases and particles are different. This may be a reflection of the regional background, and the relative daily production in relation to that regional aerosol.

11. Pg. 17095, ln. 9-11: specifically, which compounds could not be detected in injections of liquid standards?

12. Figure 4: the aspect ratio of all of the left hand panels are very poor: either improve the aspect ratio, or delete these panels from the figure.

Technical Corrections:

1. Pg. 17082, ln. 25: Herckes misspelled

References:

Bouvier-Brown, et al., In-situ ambient quantification of monoterpenes, sesquiterpenes, and related oxygenated compounds during BEARPEX 2007: implications for gas- and particle-phase chemistry, Atmospheric Chemistry and Physics, 2009

Hakola et al., The ambient concentrations of biogenic hydrocarbons at a northern European, boreal site, Atmospheric Environment, 2000.

Janson, et al., Biogenic emissions and gaseous precursors to forest aerosols, Tellus B, 2001.

Simon et al., The LANDES experiment – Monoterpenes emitted from the maritime pine, Journal of Geophysical Research-Atmospheres, 1994.

Williams et al., An In-Situ Instrument for Speciated Organic Composition of Atmospheric Aerosols: Thermal Desorption Aerosol GC/MS-FID (TAG), Aerosol Science

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and Technology, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 17071, 2011.