

Investigating Types and Sources of Organic Aerosol in Rocky Mountain National Park
Using Aerosol Mass Spectrometry
Referee #2 Comments

Author Response: Please see in-line responses denoted by “AR:”

Comments

The authors nicely discuss the chemically speciated AMS data measured at the Rocky Mountain National Park between the 2nd of July and the 31st of August 2010. The organic fraction is deconvolved by the means of the positive matrix factorization algorithm (PMF), presented and mainly discussed within the manuscript. The authors also speculate based on scientific criteria about the presence of organonitrates (ON) and about the role of biogenic SOA most probably present in SV-OOA and LV-OOA.

AR: Thank you for your review and useful feedback!

I am most concerned about the fact that the BBOA and the SV-OOA profiles are still mixed up, as already recognized and stressed by the authors. This is also evident from the fact that both profiles are quite similar, if one neglects m/z 29 in the BBOA mass spectrum. Moreover, the time series of these two factors do also co-vary to some extent. Along this line, it would be beneficial for the reader to have a table containing the correlation values for the time series among the factor time series.

AR: This information is available in Figure S1(b). Also, the supplement is now revised to contain more statistics on the recombinant PMF solutions.

As emphasized by the authors, BBOA and SV-OOA are not completely unmixed (similarity of the profiles, ts-covariation, O:C ratio). The authors realized the importance of the exploration of the solution space, by performing the fpeak analysis, exploring higher numbers of factors and testing the PMF solutions excluding strong BBOA events. Unfortunately, the fpeak analysis is too unspecific and failed to retrieve an unmixed solution. The attempt of exploring a higher number of factors bears a high potential and to my knowledge, it was already tested and published for the AMS-Montseny data (see Crippa et al., 2014 and reference therein). I would suggest to rather either use the cleaner BBOA profile retrieved with such a technique. For the reapportionment of the secondaries I recommend to either regroup accurately the other profiles into the respective SV-OOA and LV-OOA families, or to constrain the obtained clean BBOA profile in a 3-4 factor solution employing the ME-2 algorithm (see the recent study of Canonaco et al. 2013). In addition, I was wondering if some minor contribution from the Colorado Hwy 7 road might be expected too? If so, and assuming that the authors are willing to test the ME-2 solver, I would also suggest to constrain the local traffic profile in order to estimate the advected traffic contribution from the surroundings.

I recommend this article to be published in ACP after a cleaner BBOA profile is extracted and the discussion part is updated accordingly.

AR: Thanks for your feedback. New *BBOA, *SV-OOA, and *LV-OOA from recombination of a 6-factor PMF solution were presented in the supplement. We did perform

all of the above-suggested analyses (save using ME-2), and those results were included in the supplement, which now includes more detail, such that the two types of solutions (original and recombinant) are independently interpretable. Though we have also noticed that recombinant factors are gaining legitimacy in the literature (and, thank you for the additional citations), the recombinant factors yield the same (qualitative, in both cases) information about the types and temporal variability of aerosols affecting the site; taking into consideration the support from the other referee for expansion of recombinant-solution detail in the supplement and retention of the original solution in the main text, it seems not inappropriate to use the original solution in the main text.

As to contributions from Hwy 7, we saw no evidence of periods with HOA-like aerosol in, for instance, the van Krevelen diagram (Figure 6). Given that Hwy 7 sees very light traffic and is removed from the sampling site by ~200 m and a vegetative barrier, it is unsurprising that traffic signal is not evident, given PMF's inability to resolve factors contributing <5% of mass.