General Comments

This paper presents the results from ambient aerosol measurements carried out in the Po Valley. Chemical and microphysical properties of the aerosol were measured – the focus of this study is on the observation and characterization of brown carbon (BrC), which the authors attribute to SOA. This conclusion is mostly based upon a strong correlation between the AAE and the OA/BC ratio. Overall, this study is novel and the findings will contribute to the growing body of work characterizing atmospheric BrC. There are some major issues that will have to be addressed before the manuscript is suitable for ACP, but I have confidence that the authors can address these comments (detailed below).

Specific Comments

- The concept of measurement uncertainty has not been addressed at all in the manuscript. This needs to be added throughout – especially for the measurements that are central to the analyses: AAE and OA/BC ratio (or BC-to-OA ratio, as in Fig. 4).

- Similarly, the QA/QC procedures and standards need to be discussed and defended. E.g., all data with $\sigma_a < 1$ Mm^{-1} and $\sigma_s < 10$ Mm^{-1} were removed – why were these the limits imposed? How much data were discarded? In Section 3, what is meant by "The dataset consisted of 11211 records (5764 in fall, and 5447 in winter), including 2551 records (covering 40 days of measurements) with no missing value, and 1087 records (150 in fall, and 937 in winter) of cleaned data after data analysis."? Does this mean only ~10% of the measurements were ultimately included in this analysis? Section 3.2 – what constituted an acceptable merge of the SMPS and APS size distributions? What was considered unacceptable? How many measurements were eliminated using the smoothing procedure and visual inspection?

- Section 4.1 and Figures 2-4: discussion is needed to explain the physical meaning of the PC score and the factor loading numbers.

- Section 4.1 – what does the following sentence mean? "BC mass concentration was assumed to increase mostly with increasing concentration of larger BC particles…"

- Section 4.1 – what does the following sentence mean? "Higher fBC values coupled to lower BC mass concentration were, therefore, interpreted as indicators of ultrafine BC particles, and vice versa."

- Figure 1: it is not clear why the present results from the Po Valley are compared to results from Leipzig made 7-8 years ago?

- The authors have missed some other relevant work that also shows associations between ambient SOA and BrC – see for example X. Zhang et al. (2011; 2013).

- Section 5.2 and Figure 7: although the 'paradigm' discussed here may have been developed in a prior paper, it is not something I think most readers will be familiar with (this reviewer was not). Provide the necessary explanation and context to interpret the present results.

- Section 5.2, lines 28-34: perhaps this is in line with the above comment, but I was completely confused by this entire passage.

- In my opinion, Figure 8 does not much at all to the paper – I would recommend removing it.

- Section 6: the findings do not “prove” the formation of BrC in the atmosphere.

- Pg. 7, line 21-22: “The dependence on the nitrate mass fraction (fNO3 , Fig.3d) is not obvious, as high AAE values and droplet mode scores are observed for both fNO3 <0.05 and fNO3 '0.25.” This does not seem consistent with the discussion of nitrate’s importance in the abstract or in Section 6.

Technical Corrections

- Pg. 3, line 10: change to “Following the approach of Saleh et al. (2014),…”

References

Zhang, X., Lin, Y.-H., Surratt, J. D., Zotter, P., Prevot, A. S. H., Weber, R. J., Light‐absorbing soluble organic aerosol in Los Angeles and Atlanta: A contrast in secondary organic aerosol, *Geophys. Res. Lett.*, 38, L21810, 2011.

Zhang, X., Lin, Y.-H., Surratt, J. D., Weber, R. J., Sources, Composition and Absorption Angstrom Exponent of Light-absorbing Organic Components in Aerosol Extracts from the Los Angeles Basin, *Environ. Sci. Technol.*, 47(8), 2013.