This manuscript by Costabile et al. reports field measurements of aerosol microphysical, chemical, and optical properties in the Po Valley. Combining the measurements with statistical analysis, the authors conclude that brown carbon (light-absorbing organic aerosol (OA)) is mostly associated with aged OA. They also show that the spectral-dependence of the aerosol light-absorption – the Absorption Ångström Exponent (AAE) – increases with the increasing mass ratio of OA to black carbon (OA-to-BC ratio), and suggest that this confirms previous literature findings.

The measurements and the analysis are interesting and timely, however, I have major concerns (see major comments below) regarding the validity of part of the analysis and the associated conclusions. I believe that this paper can potentially be suitable for publication in ACP if the authors adequately address these issues.

Major comments:

1. <u>BrC is mainly composed of aged OA</u>

This is one of the main conclusions in this paper. The authors arrive at this conclusion based on the correlations they found between AAE and OA-to-BC ratio, and between AAE and f44. The premise that this conclusion is based on is that AAE is an indicator of how brown the aerosol is. This is not accurate. AAE is an indicator of the wavelength-dependence of absorption. What is more "brown," OA with MAC=2 m^2/g (at 532nm) and AAE=2, or OA with MAC=0.2m^2/g (at 532nm) and AAE=4? Probably the brownest OA reported in the literature is that observed by Alexander et al. (*Science 321,* 833-836 [2008]), and it had relatively small AAE.

So, with their current analysis, the authors can only make conclusions on the wavelength-dependence of OA absorption, and not on how brown the OA is. However, even this needs further analysis to be convincing. The authors base their analysis on the AAE of the whole aerosol (including both BC and OA). The AAE of the aerosol is dictated by the relative contribution of the components (and coating effects, which we can set aside for now). The increase of AAE with increasing OA-to-BC ratio does not necessarily mean that the added OA upon aging has a larger AAE, it can simply mean that the relative contribution of BC to AAE gets smaller, so the overall AAE increases. In other words, fresh and aged OA could have similar AAE, and the increase in AAE vs OA-to-BC ratio is simply due to the decreased contribution of BC.

2. <u>Validity of PSAP measurements and BC concentration calculations</u>

Figure 4 and figure 6 show that AAE is ~2 at BC-to-OA ratio of 20. This puts in question the validity of AAE retrieved from the measurements because at such large BC-to-OA ratio, the

particles should have AAE closer to 1. The authors should address this issue to determine whether there is something wrong with the calculations or the measurements that resulted in this factor of 2 overestimation of AAE. Also, BC concentrations are largely underestimated because the calculations (section 3.1) rely on the assumption that AAE of BC is equal to 1 (which is not the case, as retrieved from the instrument in this study). Consequently, the fractions of the different components should be recalculated.

Specific comments:

P1 L15: What's the difference between brown OA and brown carbon?

P2 L1: What do you mean by "moderately volatile?" Also, how do you support this claim?

P5 L12: "we fixed threshold…" please explain.

P6 L24: It is not clear why this assumption is needed or whether it is valid. You need more than "mass scales with volume" to justify this assumption.

P7 top paragraph: What are PC2 and PC4? What would be in the primary aerosol that has modes 20-100 nm and 10-40 nm? One would expect those to be BC particles.

Section 5.1: The authors mention that they will compare results with Shinozuka et al. (2009) Russell et al. (2010), Arola et al. (2011), Saleh et al. (2014), and Lu et al. (2015) but only show comparison with Russell et al. and Saleh et al.

Section 5.1: The AAE values in this study are much larger than Russell et al., especially at low f_{OA} values. I believe this is due to a bias in AAE retrieval (see major comment 2).

Section 5.1: Comparison with Saleh et al. is not apples-to-apples. Saleh et al. reports the wavelength-dependence of the imaginary part of the refractive index of the OA only, while this study reports AAE (wavelength-dependence of the absorption coefficient) of the total aerosol (including BC).

Section 5.1: The finding that AAE in this study is twice that in Saleh et al. and Lu et al. is most probably not accurate due to overestimation of AAE in this study (see major comment 2).

Terminology: the authors go back and forth between BrC, brown, and "brown." Please be consistent.

Technical corrections:

There are many grammatical/sentence structure errors (I just list a few below). The manuscript should be edited for language before it can publishable.

Title: delete “an”.

P1

L2: Angstrom should be Ångström (here and elsewhere).

L4: Delete “a” before “brown”.

L5: “Composed by” should be “composed of”.

L5: Change “enriched in” to “contain”.