Review of revised manuscript submitted by Costabile et al.: “Characteristics of "brown" aerosol in the urban Po Valley atmosphere”

The revised manuscript addressed some of the referee comments from the first review, but significant problems remain (including new problems that have emerged in this version). Thus, even after the revision, I would still rate the manuscript as needing major revisions. My major comments are:

1. The discussion about the source of the “brown” aerosol is completely convoluted. For example, in the abstract and conclusion, the authors state “it does not necessarily equate to brown carbon”. However, most of the manuscript presents support for the hypothesis that this material is indeed BrC.

2. The influence of the case study seems to drive a lot of the broad conclusions (for example, the trends in Figure 3). If this one day (out of 40 total included in the analysis) is removed, to the broad trends hold up? How would the values in Table 1 be different if the one day case study is removed?

3. I also have some problems with the authors’ interpretations of their data – especially the data presented in Table 1. Specifically, the authors suggest that PC1, PC2 and PC4 are “BC primary aerosol” (line 274). The support for this statement is that “all are correlated to BC and f_{BC}”, but I completely disagree with that assessment. PC2 and PC4 are not correlated at all with BC or f_{BC} – the highest R^2 value is 0.04 for the correlation between PC2 and f_{BC}. PC1 is not correlated with f_{BC}, and is only weakly correlated with BC (R^2 value is only 0.36). Further, line 300 claims “a robust statistical relation linking AAE, this "droplet" mode component (PC3), and OA-to-BC, together with f_{OA}, $d_{med(S)}$, f_{44} and f_{43}.” I do not think that Table 1 provides evidence for a <u>robust</u> statistical relationship, given that the highest R^2 value among all the relationships analyzed was < 0.5.

4. Building on comment #3 above, since PC3 represents larger particles compared with PC1, PC2, and PC4, it is not at all surprising that it is more strongly associated with aerosol optical properties, since it represents particles that are more optically active. This seems relevant in the data interpretation (put another way, of course PC4 has no correlation with aerosol optical properties – since these particles are so small).

5. Again, in the abstract and conclusion, it is stated that nitrate likely contributes to the aerosol absorption (lines 9 and 473). However, I see no evidence from the data to support these statements. In fact, the authors seem to acknowledge this (discussion on line 310).

6. In the abstract and conclusion, the authors claim that theirs is the first study to “consider these issues” – such a claim needs to be clarified, as many prior studies have looked into secondary BrC.

7. I made this same comment in the first review, but I really don’t understand why the prior results from Costabile et al. (2009) have been added to Figure 1? In the text (lines 289-295), the authors suggest that this study was the only other instance in the literature where a droplet

mode PC was identified from size distribution measurements. That may be because aerosol size distribution data are not routinely subjected to this PC analysis, but the observation of a droplet mode is NOT unique to Costabile et al. (2009). Overall, Figure 1 and this discussion are quite confusing.

8. I am still not sure how Figures 7 and 8 (and their associated discussion) add to the manuscript. If anything, these figures and discussion add confusion.

9. Finally, the quality of the writing needs to be improved throughout the manuscript. There are too many grammatical corrections for this referee to itemize.