Review of "Characterizing the evolution of physical properties and mixing state of black carbon particles: from near a major highway to the broader urban plume in Los Angeles" by Krasowsky et al.

The authors present two sets of results quantifying BC mixing state, abundance and size distribution – one from a cemetery adjacent to Interstate 405 in Los Angeles, and one from a suburban site about 130 km away. The presentation of methods, results, and discussion of related literature are precisely worded and of a high caliber. My main criticism with the manuscript is that although the story is mostly complete, there are large elements lacking which would make it possible to draw more meaningful conclusions. I recommend the authors address the issues described below before publication.

General Comments:

1. **Connectivity of *f* between the two datasets:** the near-road measurements are all very close to the highway and the Redland site is relatively much further from BC sources. There would seem to be quite a bit of story to be found between these two situations. The authors try to make the connection explicit by comparing *f* from the furthest near-road position to that at the Redlands site pointing out that, surprisingly, *f* is nearly the same between these two if one looks at the Redlands PCA relationship. But then, in the same paragraph, the strength of this result is seriously challenged by noting that statistical averaging is likely hiding a more complicated picture.

   I am having a hard time understanding whether comparing *f* between these two sites is valid or not valid. If the idea is that the near-road measurements are characteristic of L.A. near-road conditions and that Redlands is characteristic of the urban plume, then why focus on atypical days at the Redlands site? Why not instead remove the atypical days and focus on the days when consistent westerlies carried pollutants across from downtown? Specifically, I think the use of Fig. 10 demonstrates how potentially disconnected these separate analyses are, at least for the data that is showcased in the manuscript, like Fig. 11.

   Two possible considerations which might help bring these two analyses closer together in my mind would be 1) filter the Redlands data so that a signal can be investigated for particles that have transported across downtown L. A. and 2) convert the distances for the near-road data to "effective distances" as suggested by Reviewer 2. If some "effective distances" get much larger, then this may potentially close some of the conceptual gap between the two sites. The approach would also possibly reduce some of the noisy behavior evident in Fig. 1 and explained by the authors as due to variability in dispersion.

2. **Size distributions:** the authors do report but do not really interpret the change in peak diameter at the near-road site. They also do not discuss the likelihood that they may be missing a smaller peak below their detection limit in the 30 m data. Is it feasible that a peak in this range is growing to form the peaks detected at further distances?

   In general, I think the existing discussion of size distributions is not very helpful unless "effective growth rate" metrics or modal analyses are presented as well. For example, urban and regional scale models could use this data if the particle observations were used to derive distinct aerosol modes with representative parameters (i.e. dg and sigma). It would also help future researchers add this study to broad surveys of BC emissions and mixing state.

3. **Context:** I think a paragraph comparing *f* magnitudes with those reported by existing publications is missing. Are these values high, low or expected compared to what has already been measured? Is this study adding a new perspective to the field, or confirming previous observations? Also, the authors

have not explicitly answered the age-old “what makes this paper novel?” question, but I also think that question is rather ridiculous, so I would leave it up to their discretion – it couldn’t hurt rewriting a sentence in the introduction so it stands out.

Minor/technical comments:

1. Consider adding a map of the two sites in the methods section to orient the readers.
2. The inclusion of section 2.5 is appreciated, but it does break up the flow of the manuscript considerably (it’s a 1.5 page monster paragraph!). Moreover, it contains a bit of everything, including specific technique description, discussion of general background from the literature and discussion of literature results that are directly relevant to results presented later. Consider moving it to supporting info, or breaking it apart so some of it goes to the intro, some stays in the methods and is added to the existing section 2.6, and some goes to the discussion of results.
3. Page 9, line 21-22: How sensitive are your reported *f*’s to the value of the cutoff that you choose? Can you create a graphic for the supporting info that shows how average *f* varies as a function of the cutoff value for a particular set of the underlying data?
4. Section 3. The titles are a bit cumbersome. How about changing the title of Section 3.1 to “Near-road spatial trends” and then 3.1.1 to “rBC concentration and coated fraction”. 3.1.2 becomes “rBC size distribution” and 3.1.3 becomes “Coating thickness”
5. Page 11, Line 22: replace “would could” with “and this could”
6. Page 11, Lines 23-24: Delete sentence beginning with “This is somewhat analogous…”
7. Page 12, Line 2: So you know what the lower detection limit was for the Zhu et al. study? How would it change the interpretation, if at all?
8. Page 13, line 18-19: You’ve included the outlier in the main plot and mentioned it again here. I would recommend removing it from the analysis and making sure to mention you took it out.
9. Page 14, line 24-25: What are “processes other than photochemistry?” Are you referring to condensation of semivolatiles or other processes too? Can you mention them all?
10. Table 1: Consider putting the Zhu et al data first, followed by the near-road results and then the Redlands campaign. This way the results are ordered generally by BC magnitude and conceptually flow a bit more naturally.
11. Fig. 3: Error Bars showing variability? The excessive grid lines make the size distribution plots here and in Fig. 11 hard to read.
12. Fig. 7: This could be moved to supporting info and averages mentioned in the manuscript.
13. Fig. 9: Some explanation of the strange increase in BC mass at f = 0.3-0.4 is probably warranted.