

Interactive comment on “Spectro-microscopic measurements of carbonaceous aerosol aging in Central California” by R. C. Moffet et al.

Anonymous Referee #1

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This manuscript contrasts aerosol composition measurements at two sites along a Lagrangian trajectory during the CARES campaign in California. The authors combine observations from OC/EC, electron microscopy, and X-ray microscopy. For the last technique, they apply a reduced set of X-ray energies to determine total organic carbon content (relative to the inorganic fraction) for a large number of individual particles and contrast these results, along with descriptions of soot-like character, to particles previously analyzed from the MILAGRO campaign in Mexico City. This manuscript is recommended for publication in Atmospheric Chemistry and Physics after the following comments are addressed:

General comments:

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Some logistical details are difficult to locate in the manuscript. For instance:

- How many particles were analyzed by CCSEM/EDX vs STXM/NEXAFS? Figure S1 and S2 contains the derivative of this information but should be stated up front. For STXM/NEXAFS, how many images/spectra were obtained with great detail (p. 9192 line 26: what is “numerous”?) vs. the quicker method with reduced number of energy levels? Also, in each of the analyses sometimes it is unclear which spectra was used to derive the results or metrics.
- Were some TEM grids and silicon nitride windows sampled on concurrently or in sequentially within the same time window? Cannot tell if there is overlap in the vertical lines in Figure 3.

The relationship between the CCSEM/EDX and STXM/NEXAFS particle categories are unclear following the discussions in Sections 3.2 and 3.3.

Regarding the elucidation of mechanisms discussed in the abstract and introduction in the results section:

- Small particles with potassium cores possibly indicative of biomass burning:
 - Tivanski et al. 2007 (previous work by one of the co-authors) studied the spectral signature of the NEXAFS K-edge of biomass burning particles. Is there any resemblance of these particles with these tarballs?
Tivanski, A. V.; Hopkins, R. J.; Tyliszczak, T. & Gilles, M. K. Oxygenated interface on biomass burn tar balls determined by single particle scanning transmission X-ray microscopy, *Journal of Physical Chemistry A*, 2007, 111, 5448-5458
 - Were there biomass burning events reported during these periods at the CARES study?

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- Increase in total carbon content due to nucleation and/or condensation of secondary organic aerosol:
 - e.g., p. 9195 line 5: were there nucleation events reported (e.g., from SMPS measurements)?
 - Do the NEXAFS K-edge spectra look homogeneous (if possible to tell from the reduced set of energies), suggesting similar organic aerosol composition? This would strengthen the case for gas/particle partitioning.

Specific comments:

p. 9186 line 3: nitrogen and oxygen analyses are not mentioned in the rest of this manuscript.

p. 9193 line 6: what were the spatial resolutions actually used?

p. 9194 line 22: what was the criteria for determining soot and carbonate regions from the reduced set of energies? Were the excluded regions on a per-pixel basis (which goes back to the question of spatial resolution of last comment)?

p. 9196 line 1: averages of organic carbon contents should be followed by an ($n =$).

p. 9196 line 23: Figure 10b and its description is rather confusing. Why are there so few points? If each point represents a different particle, there is no statement regarding the randomness of sample selection for these few particles so the figure may not be conveying a true correlation. Since the authors report estimates of total organic carbon, presumably based on a difference between 278 and 320 eV, for many more particles (where some randomness is introduced) in Figure 8, why are these not used for this correlation?

p. 9197 line 6: Is this decrease in C=C accompanied an increase in O:C ratio as can be determined from carbon or oxygen NEXAFS spectra?

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Figure 8: should not the equivalent diameter include the contribution from inorganic carbon to the particle size?

Figure 9: how many particles and what range of sizes are included in each average?

Figure 11: are these from detailed spectra or averages of the rapidly acquired spectra?

Minor comments:

p. 9192 line 25: typo “function”

p. 9193 line 10: also originally by Maria et al. (2003) (previous work by one of the co-authors)

Maria, S. F.; Russell, L. M.; Gilles, M. K. & Myneni, S. C. B. Organic aerosol growth mechanisms and their climate-forcing implications, *Science*, 2004, 306, 1921-1924

p. 9193 lines 10-15: as detailed/shown in (Moffet et al. 2010a) should be as detailed/shown by Moffet et al. (2010a)

p. 9195 line 18: “due to” → “consistent with”

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9179, 2013.

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