RED=REVIEWER COMMENT
GREEN=AUTHOR RESPONSE

The manuscript titled as “Morphology and Mixing of BC Particles Collected in Central California During the CARES Field Study” is about “characterization of the (BC) particle mixing state and morphology using Scanning Transmission X-ray Microscopy (STXM) at the carbon K-edge”. In their work, characteristics of BC aerosol particles collected at two sampling sites were a main focus, which is worth to be published in ACP.

We thank the reviewer for their comments. Below we provide our detailed responses.

There are some specific comments which can help improve this manuscript.

1. The description of sampling sites and conditions needs to be consistent and clearer in the manuscript. That is, in the experimental section, the two sampling sites were described as “the first site was in the Sacramento urban area (T0 site) expected to have enhanced fresh emissions(?), and the second site was located 40 km east of T0 in the Sierra Nevada foothills (T1 site) expected to have enhanced aged aerosol(?).”In the abstract and in the conclusion, it is said as “at both urban and rural sites” and “at source (T0) and receptor (T1) sites in the California central valley”, respectively. Of course, these three different descriptions on the sampling sites may be related to each other, but without some concrete connections. It is better to make these descriptions consistent.

We believe that the prevalence of aged aerosol at the urban site is perhaps contrary to expectations, so we have highlighted this by changing the sentence in the abstract (Page: 1 Line: 21-23 of the revised manuscript) to read:

“Observations of compacted BC core morphologies and thick organic coatings at both urban and rural sites provide evidence of the aged nature of particles, highlighting the importance of highly aged particles at urban sites during periods of high photochemical activity.”

In the experimental section, it is said that “Samples utilized here were collected over two days (June 27 and June 28) during a period of high temperatures and increased aerosol loadings over T0.”And in the abstract and conclusion sections, it is said that “During a period of high photochemical activity and pollution buildup”. I think these two description are not consistent.

As the reviewer has suggested, we have made the connection between “pollution buildup” and “photochemical activity” more explicit. Specifically:

The sentence on Page: 3 Line: 6 of the revised manuscript was changed to read:

“Samples utilized here were collected over two days (June 27 and June 28) during a period of high temperatures and increased aerosol loadings over T0 due to high photochemical activity.”

In addition, to relate the findings for BC particles collected at the two sampling sites, information on backward trajectories, sampling times and durations at T0 and T1 sites, and wind speed and direction needs to be given to better provide some clear idea about samples collected at T0 and T1 sites.

As the referees suggested, we have added a more detailed discussion of meteorological conditions during this episode during the CARES field study. Meteorology was covered extensively in the Fast et al. reference and details of CO modeling were further presented in the Moffet 2013 reference. We have elaborated on these points in the experimental section (Page: 3 Line: 8):

"CO tracer modeling indicated that significant transport from the San Francisco Bay Area affected the Sacramento site in the morning while the boundary layer was low. Later on in the day a larger fraction of emission at the T0 site were from the Sacramento metropolitan area. Similar contributions from the San Francisco Bay Area were modeled at T1, however, emissions from Sacramento constituted the largest source of emissions during this time period (Moffet et al., 2013; Fast et al., 2014)."

2. In the abstract and Results section, thick "organic" coating is mentioned, and in the conclusion, it is said that "During this period, the overall particle size at the receptor site was significantly larger due to the condensation of organic and inorganic species". Indeed, I am curious about the modification of "inorganic" species during the possible aging process. If some argument about inorganic species aging is given in the Results section, it will be interesting.

Although aging of inorganic species (NaCl reacting with acids, oxidation SO_2 to SO_4 etc.) was not the focus of this manuscript, discussion of the aging of inorganic rich particles through the process of coagulation with BC can be found here:

Revised manuscript Page: 5 Line: 21: The most striking difference between BC particles from T0 and T1 is the high amount of inorganic dominant regions for T0 particles. T0 was impacted by sea spray and sulfates from petroleum refineries located in the San Francisco Bay Area (Laskin et al, 2012). The large inorganic dominant particles can be attributed to sea spray particles that have coagulated with BC emissions from the Bay Area.

3. The abstract and conclusion parts need to be rewritten to convey the findings and meaning of this work more consistently and clearly.

This relates to point #1, and we have modified the abstract and results to make the connection between pollution buildup and photochemical activity more clearly and consistently.

(4) List of Awkward and/or ambiguous sentences and/or sections

-p. 3, lines 22-23

We understand that the reviewer may have found this awkward, so we have reworded the sentence to read:

Page 3, Line 25: "To generate a carbon-based map, aerosol particles were imaged at 278 eV (the carbon pre-edge), 285.4eV (sp^2 C*=C), 288.6eV (C*OOH), and 320 eV (the carbon post-edge). Characteristic single energy images at these energies are shown in **Figure 1A-D**. Typically, at each energy, a 15 x15 μm^2 image was acquired with 0.035 µm pixel size and 1 ms dwell time. Occasionally, ~120 different constant energy images were utilized in this study to obtain a high resolution carbon spectrum. For consistency, the same set of four constant-energy images (278, 285.4, 288.6, and 320 eV) were used to characterize particles for this analysis."

-p. 4, lines 4-6 (and needs to say why)

We have stated the purpose in the beginning of the sentence:

“To define organic carbon rich (OC), inorganic non-carbonaceous rich (IN), and black carbon (BC) regions, thresholds for each of the three maps in Figure 1E-G were set using the following criteria:”

-p.4, lines 9-21: only the expert of this technique could understand this part.

We have attempted to make this section more accessible to the layperson – however, we feel that while only an expert can understand parts of this section, the information is necessary to provide a more complete documentation of the analysis. Much of this paragraph outlines quality control measures. To make this clear, we have added an introductory sentence:

Page 4 Line 21 of revised manuscript: “A brief description of quality control measures is summarized here.”

-p.6, lines 3-6: Difficult to understand.

We have added an introductory explanation of the concept of diversity – which we suspect is what the reviewer found difficult to understand:

Page 6 Line 15 of revised manuscript: “In O’Brien et al. (2015), entropy metrics (Riemer and West, 2013) were used to calculate a diversity that represents the effective number of species per particle or in the bulk population. In this case diversities were specified using the OC, BC, and IN components such that a particle or population can have a maximum diversity of 3.”

-p.6, line 20: droplet mode ???

We have added the approximate range of the droplet mode from 200 – 1000 nm.

-p.6, lines 31-31and p.7, lines 4-5: This description is mostly inconsistent with some descriptions given elsewhere in the manuscript.

We understand the reviewer believes that the presence of fresh emissions is contradictory to the statements in the abstract and the results and discussion. We have carefully worded the results in the abstract to state that there was a small population of fresh emission detected at T0. To emphasize the relative contributions of fresh and aged particles we have added the following discussion to Page 7 Line 4 of the revised manuscript:

“…comparison of the convexity distributions between T0 and T1 indicates a small statistically significant population of less compact particles at T0 consistent with fresh emissions. However, it should be emphasized that the majority of particles at both sites have compact shapes and are likely due to the prevalence of aged BC containing particles.”

-p.7, line 16: “in source regions”and “the source of these particles”???

We changed “source regions” to “urban areas with similar meteorological conditions” as that more accurately conveys the main idea of the sentence.

-p.8, lines 25-29: Hard to understand.

Again, this relates to the seemingly contradictory concept that we observe fresh particles at T0 while it is emphasized elsewhere that most of the BC particles are highly aged. To address this we have emphasized that the population of thinly coated particles is small as well as other minor clarifying edits:

Page 9, Line 22 of the revised manuscript: “The T0 site had a small population of thinly coated particles compared to the T1 site. This is consistent with slightly smaller overall particle sizes at T0 and the assumption that BC at the source site (T0) should have thinner coatings due to the presence of fresh BC emissions.”

(5) Typos:

- P.3, line 8 : relative humidity ->moisture

corrected

- P.3, line 13 : definition of rho is missing.

corrected

- P.4, line 24 : molecular markers ->chemical (or functional group)markers

corrected

- P.4, line 32

Did not find a typo

- P.5, line 29: maps OCBC and OCBCIN, particles ->maps, OCBC and OCBCIN particles

corrected

- P.5, line 30: INOC->IN

Added “particle types” to the end of the sentence to convey our original meaning.

- Many places in the manuscript: DBC:DTotal->DBC/DTotal

corrected

- P.10, Figure 1 caption : A-C ->A-D

corrected

- P.11, Figure legend: Soot inclusion ->BC inclusion

corrected

- P.14, Figure caption : DBC:DTotal->DBC/DTotal

corrrected