RED=REVIEWER COMMENT
GREEN=AUTHOR RESPONSE

This study shows mixing states of individual BC particles from relatively fresh and aged samples using STXM. The data is valuable for those who study aerosol mixing states and BC climate impacts. The technique used in this study is unique and includes important information of mixing states of BC, organic, and inorganic matters. The manuscript is well written, and I believe the manuscript contributes to the community.

We thank the reviewer for taking the time to comment on our manuscript. We have taken the reviewers comments into account in a revised version of our manuscript. Below we detail how each comment was addressed in the revised manuscript.

P: Page L: Line
Specific comments 1: P1L23 “the contribution of fresh BC emissions at the urban site was relatively small”. The sentence somehow contradicts to that in P6L17-19 “This result is expected considering that the T0 site is located in a source region for freshly emitted BC particles”, P6L31-32 “It is probable that most of the particles sampled during CARES are substantially aged and/or that the aging time (and subsequent collapse into compact shapes) is rapid” and P7L4-5 “Hence, the presence of more particles with lower convexities is consistent with the presence of fresh emissions at T0.”. I suggest mentioning that “aging was rapid” or similar wordings.

We have added the possibility of rapid aging to the abstract;
Page 1, Line 23: “Based on the observation of thick coatings and more convex BC inclusion morphology, either the aging was rapid or the contribution of fresh BC emissions at the urban site was relatively small.”

2: P3L27: Please briefly describe how inorganic dominant region was obtained from the carbon edge regions in addition to the reference (Moffet et al., 2010). What kind of inorganic was detected here?

To address this comment, we have added some detail to the revised manuscript:

Page 3-4 Lines 33-2 The inorganic map, representing non-carbonaceous inorganic dominant regions, is derived from the ratio of the pre edge (278 eV) to the post-edge (320 eV) ratio (ODpre/ODpost) and is shown in **Figure 1F**. Non-carbonaceous, inorganic inclusions of (NH4)2SO4 and NaCl were confirmed using energy dispersive X-ray spectroscopy for these CARES samples (Moffet et al., 2013).

We also refer the reviewer to our response to comment #4 below in which we include more details about thresholding. Page 4 Lines 15-18

3: P3L28: Delete “to” from “to to”.

Corrected.

4: P4L6: Please briefly describe how “35%” was used for the threshold. I believe the thresholds in STXM imaging are important to distinguish the materials.

We have addressed this request by adding more specific detail about how the thresholding was carried out. These changes also address reviewer concern #2. The revised wording is as follows:

Page 4 Lines 11-18 "Thresholds for each of the three maps in Figure 1E-G were set using the following criteria: 1) pixels at 288.6 eV with intensities three times below signal to noise ratio were set to zero 2) pixels having ODpre/ODpost<0.5 were set to zero as discussed in Moffet et al. 2010 and 3) the %sp2 was set to zero below a value of 35%. The empirical determination of the threshold value of 35% is discussed elsewhere (Moffet et al., 2011). Areas of each of the maps with fewer than 7 conjoined pixels were excluded. Thresholds for maps in Figure 1E-1G were applied to produce binary images. In Figure 1H, to highlight the BC mixing state these three binary images are overlaid in the following order: organic, inorganic, sp^2."

5: P5L6: Please define (or refer the caption in Fig. 3) "OCBC, OCBCIN, OC, IN, INOC" in the main text in addition to the caption Fig. 3.

We have added these definitions to Page 5 Line 8 of the revised manuscript:

Page 5 Line 8 "Based on these maps, the mixing state of the particles was stored in a database for the subsequent analysis of BC morphology. The label-based mixing state is defined from particles having organic carbon (OC), black carbon (BC), or inorganic (IN) regions defined by the binary maps shown in **Figure 1H**. For example, if a particle contains both organic and black carbon regions it is labeled OCBC and so on."

6: P6L26-27: "suggests that restructuring of the particles to more compact shapes upon transport is negligible" and Figure 5. I suggest discussing how useful to discuss the shape of BC using relatively low pixel size resolution (35 nm) comparing to BC monomer size (_40 nm). Although the particle in Fig. 1 has fractal shape, most particles in Fig 2 do not show BC branches or BC monomers. I wonder if the BC images in Fig 2 is due to BC restructuring or a lack of image resolution.

In order to address this comment, as well as to add the possibility that resolution affects our ability to detect restructuring, we have added the following text:

Page 7 Line 20 The resolution of the STXM instrument limits the ability to identify small (<100 nm) branched/fractal inclusions. However, even freshly emitted soot particles become more compact at smaller sizes (Park et al., 2004). Moreover, monomer size tends to be around 40 nm, which should be detectable by the STXM instrument, though operating at the very limit. The monomers of BC aggregates are typically connected in order to form a branched, fractal particle, resulting in the observation that only larger particles have high fractal dimension.

We have added the following reference to support this added discussion:

Park, K., Kittelson, D. B., and McMurry, P. H.: Structural properties of diesel exhaust particles measured by transmission electron microscopy (TEM): Relationships to particle mass and mobility, Aerosol Sci Tech, 38, 881-889, 10.1080/027868290505189, 2004.

7: P7L18: "3.2" should be "3.3"

Corrected

8: P8L11: “Specifically, the phase of the organic and inorganic material must be considered” and Figures 2 and 7. This comment is a suggestion. In Fig. 2, it looks most BC locate inside of organic matter and outside of inorganic particles. It may be interesting to see if such difference is statistically true using the similar plot of Fig. 7.

We felt that this was an important point that is related to the observation made on P5L14-15 of the original manuscript: “Frequently, BC inclusions were seen on the outside edge of an inorganic dominant region; this arrangement may have occurred upon efflorescence, when a salt excludes the aqueous phase (Liu et al., 2008).”

In order to address this hypothesis in a more quantitative fashion, we observed that the radial distribution of BC inclusions of OCBCIN particles has a significantly higher fraction of BC inclusions closer to the edge of the particle. Particles that contained large IN (inorganic) inclusions had an even higher proportion of BC inclusions closer to the edge of the host particle. We have added these distributions to Figure 7 along with the following discussion in the results section:

Page 8 Line 16 of the revised manuscript:

“Analysis of particles from two sampling sites showed minor differences in the locations of the BC inclusions within host particles, suggesting that the distribution of BC inclusions does not vary substantially between the urban (T0) and rural (T1) sampling sites. Slightly more BC inclusions were found closer to the edge of the host particle at T0. This is likely due to the higher frequency of inorganic inclusions at T0; the crystalline inorganic phase may push the BC inclusions away from the center upon efflorescence. Moreover, BC particles may more easily mix with the OC phase when the particle is in the dry state. The bottom panel of **Figure 7** demonstrates that particles containing inorganic rich phases (OCBCIN particles) have an enhanced number of particles with the BC inclusion near the edge of the host; this trend is enhanced when particles with large (500 nm) inorganic inclusions are considered. These results demonstrate that the majority of the BC inclusions were found in the center of their impacted host particle at both sites and that the presence of inorganic dominant inclusions acts to push the BC inclusion farther from the center of the host.”

A sentence was added to the conclusions of the revised manuscript (Page 9-10:Lines 32-1):

“Particles containing inorganic rich inclusions were more likely to have the BC inclusion pushed towards the edge of the host.”

The following was added to the abstract (Page 1,Line 27):

Most particles were observed to have the BC inclusion close to the center of the host particle. However, hosts containing inorganic rich inclusions had the BC inclusion located closer to the edge of the particle.

9: P10 Figure 1. 9.1: Please add a scale bar.

We have added a scale bar.

9.2: In Figs.1C, E and H, the organic matter in E is larger than that in Fig.1C (COOH distribution), especially for particle 1. Is this artificial effect or real distribution? The BC Map (G) also looks similar enhancement C2(e.g., images B vs G for particles 1 and 2). I think the choice of threshold relates to this issue.

For particle 1, it is likely that the organic matter appears to be larger in E due to the fact that the map is derived from more than one image and the contrast is different. Images A-D are single energy images with large differences in absorbance values due to differences in thickness. For example, comparing B vs. G is difficult/non-trivial because B is a single energy image whereas the “BC Map” (G) is derived by the ratio of image B divided by the total carbon (Image D – Image A). Thus, Image B is dependent on thickness, whereas map G is independent of thickness. There is reason to believe that they are different because they are representing two physically different measurements.

9.3: Images E, F, and H: When focusing on relatively large inorganic dominant particles (e.g., right upper particle or left bottom particles), I see some inorganic rich inclusions coated by organic. However, in the combined map (H), I do not see such features but see organic only in the rim. Please explain what happen here.

This is a threshold issue. Even though the rims are less enriched in inorganics, the OD_{pre}/OD_{post} is still greater than 0.5, indicating inorganic rich coatings.

10: P15L4: “see illustration above plot): ”Take out “)” or add “(“.

We have made the correction.