Response to comments

Anonymous Referee #1

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This paper discusses the activation of black carbon (BC) containing particles into cloud droplets at a high altitude location. The authors collected cloud-interstitial aerosol (INT), residual aerosol from dried cloud droplets (RES), and aerosol during cloud-free periods. The impact of particle composition and size were then evaluated in regards to the aerosol's ability to activate into a cloud droplet. The largest particles and those with the highest fractions of secondary components, such as sulfate, had the highest activation fractions. However, the relative impacts of these two conditions (size v. composition) on the ability of the particles to be scavenged by the cloud are difficult to distinguish, as the largest particles also contained the highest fractions of secondary components. In addition, two different notations are used to describe the fractions of BC and BC-containing particles in cloud droplets: activation of particles and cloud scavenging. Choosing one of these notations (activation or scavenging) and then evaluating for both number concentrations and BC mass fraction would help streamline the presentation of the results.

We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript.

We agree with the comment that the relative impacts of these two conditions (size vs. composition) on the ability of the particles to be scavenged by the cloud are difficult to distinguish. As indicated by the reviewer, larger particles also contained higher fractions of the secondary components. We showed the importance of mixing state through a comparison of the individual particle types of cloud RES with the cloud INT and cloud-free BC-containing particles. As shown in Fig. 5 in the revised manuscript, the number fraction of BC-OC-sul (~8%) was much lower in the cloud RES than those (~25%) in the cloud-free and cloud INT BC-containing particles.

We agree with the comment that choosing one of the notations (activation or scavenging) would help streamline the presentation of the results. We have replaced "activation" to "scavenging" in the revised manuscript, e.g., section 3.2.1 "Size-resolved scavenging of BC-containing particles".

Specific Comments:

Line 163: Of the 7 wavelengths measured, why was 880 nm chosen to use for values of EBC?

Thanks for the comment. At 880 nm wavelength, light absorption can be attributed to BC alone rather than the other aerosol particles due to their significantly less absorption at long wavelength (e.g., Sandradewi et al., 2008; Yang et al., 2009). Therefore, the absorption coefficient at the wavelength of 880 nm is typically chosen to use for the concentration of EBC. We have clarified it in Lines 41-43 of the revised Supplement.

Line 182: For the category of BC-sul1, should this line read "…and less sulfate" or is the sulfate concentration also high for this category? Also, how were the categories determined, i.e. what was the cut-point for categorizing a particle as "more intense sulfate" and "abundance of both sulfate and organics"? Was a specific mass-fraction used to divide the categories?

Thanks for the comment. There was not a specific mass-fraction used to divide the categories. An adaptive resonance theory-based neural network algorithm (ART-2a) (Song et al., 1999) was applied to cluster the individual particles, based on the presence and intensities of ion peaks. The generated particle clusters were further manually grouped. Therefore, the cut-point for categorizing a particle as "more intense sulfate" and "abundance of both sulfate and organics" is based on the intensities (or relative peak area, RPA) of sulfate and organics. To make it clear, we have shown the statistical analysis on the ion peak ratio of OC to BC and the average mass spectra for the BC types, which also shows the relative intensities of sulfate for BC-sul1 and BC-sul2. More intense sulfate (RPA = ~0.3) was found for BC-sul2 and BC-OC-sul, relative to that (RPA = ~0.15) for

BC-sul1 type. More abundance of OC was found for BC-OC-sul, the mean peak area ratio OC/BC of which is ~1, higher than those (< 0.3) for other BC types. Please refer to Fig. S2 in the Supplement.

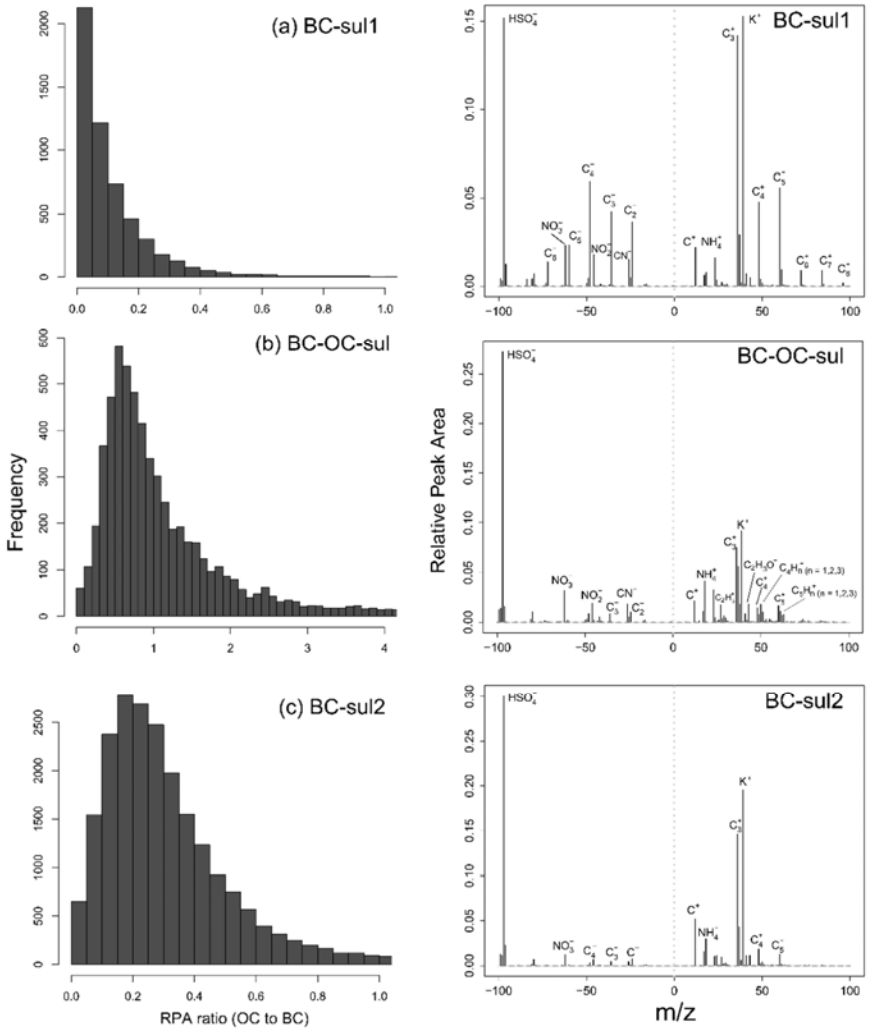

Figure S2. Statistic analysis on the RPA ratio of OC to BC (left), and the average mass spectra (right) for the BC types. Markers were selected as m/z 27, 43, 50, 51, 61, 63, -26 for OC, and carbon ion clusters ($C_n^{+/-}$, $n \leq 5$) for BC, the same as those in Fig. 3. More intense sulfate (RPA = ~0.3) was found for BC-sul2 and BC-OC-sul, relative to that (RPA = ~0.15) for BC-sul1 type. More abundance of OC was found for BC-OC-sul,

the mean peak area ratio OC/BC of which is ~1, higher than those (< 0.3) for other BC types.

Line 204: Is ~0.1% the percentage of BC-containing particles detected only during the 2 hr window or for the whole sampling period for RES?

The ~0.1% here refer to the fraction of cloud RES BC-containing particles during the 2-hour window, when an average temperature was ~ –7 °C. We have revised the sentence to "The cloud RES BC-containing particles only accounted for ~0.1% of all the detected ones in a 2-hour window when the average temperature was ~ –7 °C" to make it clear. Please refer to Lines 211-212 of the revised manuscript.

Line 259: Please clarify what is the meaning of "cannot be ruled out by" in this context and how this relates to the results presented.

Thanks for the comment. To make it clear, we have revised these sentences "An abundance of BC-coated materials was also observed at Mt. Soledad (Schroder et al., 2015). Unfortunately, their chemical compositions cannot be ruled out by a single particle soot photometer. Therefore, our analysis reflects the importance of the chemical mixing state on the cloud processing of BC." to "Although an abundance of BC-coated materials was also observed at Mt. Soledad by a single particle soot photometer (Schroder et al., 2015), the chemical compositions of the coated materials cannot be obtained to provide further information on the mixing state of BC. Our analysis further reflects the importance of the chemical mixing state on the cloud processing of BC."

Line 297: Please clarify how the particles at 700 nm decreased in size to 100 nm for the higher LWC. Is the decrease in size for the diameter of the activated droplets?

Thanks for the comment. It does not mean the decrease in size for the diameter of the activated droplets for the higher LWC. It is the decrease of the half activated diameter for particles to be activated. To avoid the ambiguity, the discussion has been revised to

"Relatively lower scavenging efficiency (0.05–0.45) in the present study was most likely attributed to less dense clouds (with a liquid water content or LWC < 0.1 g m^{-3}). Generally, the half activated diameter increases with decreasing LWC. Henning et al. (2002) stated that particles with d_{ve} = 700 nm were only half activated with LWC < 0.1 g m^{-3}, in contrast, particles with d_{ve} = ~100 nm can be half activated when the LWC > 0.15 g m^{-3}.". Please refer to Lines 319-325 of the revised manuscript.

Line 309: The paragraph starting at line 309 deals solely with the role of mixing state in activation of BC-containing particles. This paragraph would fit more logically in "Section 3.1 Mixing state of BC for cloud-free, residual, and interstitial particles" than in its current location, "Section 3.2.1 – Size-resolved activation of BC-containing particles."

We agree with the comment. The paragraph has been moved to section 3.1 in the revised manuscript, please refer to Line 267-279.

Line 318: The statement that organic-dominated particle types were "activated to a lesser extent" does not seem to be supported by Figure S9. For half of the diameters marked, the organic-dominant particles were nearly equal to or above the activated fraction of BC-containing particles. For the highest 3 diameters marked for the organic-dominant particles, the error bars encompass the range of the BC-containing particles.

Thanks for the comment. We have deleted the statement in the revised manuscript.

Line 322: Is this information (frequency of observation) included in one of the figures (possible figure 3)? If so, please include a reference here to the appropriate figure.

Thanks for the comment. This information is included in Fig. 5. We have clarified it, please refer to Lines 277-279 of the revised manuscript.

Line 370: Was LWC measured in this study? If not, why is the assumption made that the conditions are low-LWC?

LWC was not measured in this study. The LWC in this study was expected to be < 0.1 g m^{-3} according to the discussion in Line 319-325 of the revised manuscript. Generally, the half activated diameter increases with decreasing LWC. Henning et al. (2002) stated that particles with d_{ve} = 700 nm were only half activated with LWC < 0.1 g m^{-3}, in contrast, particles with d_{ve} = ~100 nm can be half activated when the LWC > 0.15 g m^{-3}.

Figure 1: Please add units for the vertical-axis categories. Also, are the PM2.5, EBC, and Num. of BC data for all categories (INT,RES, and cloud-free combined)?

Thanks for the comment. We have revised the Figure 1 accordingly. We have added "$PM_{2.5}$ during the cloud events corresponded to the cloud INT particles. EBC and number of BC-containing particles data were shown for all categories, including the cloud-free, cloud RES, and cloud INT particles. The cloud INT particles were only measured during Cloud III." in the caption of Fig. 1 to clarify the data.

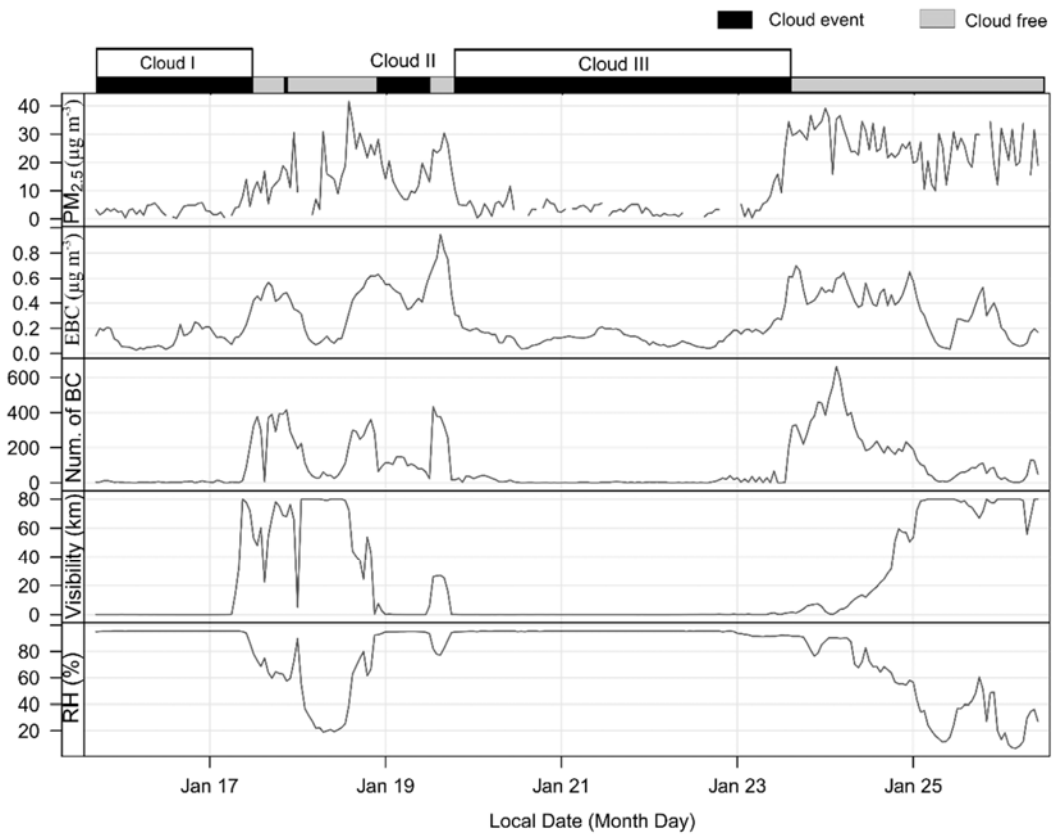

Fig. 1. Temporal profiles (with a 1 hour resolution) of $PM_{2.5}$, EBC mass concentrations, number of BC-containing particles by SPAMS, RH and visibility. Three cloud events are illustrated with black bars above the figure. $PM_{2.5}$ during the cloud events corresponded to the cloud INT particles. EBC and number of BC-containing particles data were shown for all categories, including the cloud-free, cloud RES, and cloud INT particles. The cloud INT particles were only measured during Cloud III.

Supplement Line 59: Please clarify what is meant by "they were taken into account" and how this relates to the calculation of the uncertainties that resulted in 10%.

Thanks for the comment. We have revised to these sentences to "The mean $Mf_{scav,EBC}$ was recalculated to be 30-36%, when the assumed largest underestimate (i.e., 30%) of the cloud RES particles and ~15% underestimate of the cloud INT BC were taken into account in R1. Compared to mean $Mf_{scav,EBC}$ = 33%, the overall uncertainties for the estimate of mean $Mf_{scav,EBC}$ is with 10%." to clarify the statement. Please refer to Lines 65-68 of the revised

Supplement.

Figure S8: Please clarify in the caption the line: "the other particles also contained OC particles (10%)." Does 10% refer to the percent of total particles containing any amount of OC or the percent of total particles that had OC as the dominant species?

Thanks for the comment. We have clarify that 10% refer to the percent of total particles had OC as the dominant species. Please refer to Line 128 of the revised Supplement.

Technical Corrections:
Lines 336-337: Please divide these lines into two sentences: "...areas (Huang et al., 2012). It is similar to those..."

Thanks for the suggestion. It has been revised as suggested.