Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-769-AC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements to determine mixing state of black carbon emitted from the 2017/2018 California wildfires and urban Los Angeles" *by* Joseph Ko et al.

Joseph Ko et al.

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Received and published: 1 May 2020

Author Response to RC2

We appreciate the thoughtful and detailed review from Referee 2. We have taken the comments made by Referee 2 into careful consideration and they have helped improve our manuscript.

The general format of this response is as follows:

- Reviewer comments are in bold and labeled as (N.1), where N is the number of the comment block.

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- Author response to comments are in regular, non-bolded text, and labeled as (N.2).
- Modifications in the manuscript are described in italics and labeled as (N.3).

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(1.1)

Major comments regarding source attribution and estimated plume age

(1.2)

We agree with the reviewer that the source attribution and plume age sections of the manuscript required some major revisions (e.g., section 3.7, formerly section 3.6). We now shift our focus towards comparing the mixing state during different known source impacts, rather than focusing on the plume age. As the reviewer notes, rBC from biomass burning (BB) is coated much more quickly than rBC from urban emissions, and BB rBC has also been observed to have thicker coating overall compared to its urban counterpart.

We would like to address the nuances associated with the specific concerns that the reviewer raises in the comment.

Regarding biomass burning source attribution:

First, we wanted to clarify that we are not definitively attributing ~10 nm coating thickness values to fresh BB rBC particles, and we changed the language in the revised text to make this clear. ~10 nm was the median coating thickness from a population of aerosols that had a larger spread of individual coating thickness values. The coating thickness values on the higher end of the distribution tail (and outliers) are likely attributable to the BB impacts. We clarified in the new text that the peaks in the 2nd campaign (e.g., L4) were likely dominated by urban emissions, but that we could not exclude the likely impact of BB emissions mixing into the broader urban plume. In

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Discussion paper

fact, we still believe that biomass burning did impact our measurements to some degree, even if it was a minor fraction of total sampled rBC. In particular, the Thomas Fire was one of the largest fires in California history, and it was still active during the 2nd campaign (20-22 December 2017). With the center of the Thomas Fire less than 150 km away, and strong atypical Santa Ana winds recorded before and during the time of measurements, it is hard to imagine BB having no impact on the regional rBC loading at the time. In addition to geographic proximity and meteorology, the air quality monitoring stations in Santa Barbara, Ventura, and Los Angeles all recorded elevated concentrations of PM2.5 right around this time period. Additionally, as part of the new supplemental analysis, the HYSPLIT dispersion model was run to simulate the plume dispersion of multiple active fires during the December 2017 campaign. The HYSPLIT dispersion model shows the plumes from the Thomas Fire and several other smaller Southern California fires directly impacting the point of measurement (Catalina Island). These results are included in the revised Supplement. We also added a new qualitative analysis in the Supplement using CALIPSO lidar transects in the Southern California region during the 20-22 December period. From the CALIPSO transects we observed aerosols that were attributed to BB sources present just off the coast of Southern California around this time. This data is also shown in a new section in the Supplement.

Second, since the paper was first submitted, we have obtained levoglucosan data from November 2018 (3rd campaign) that were collected by colleagues at USC who were conducting an independent air quality study in the LA Basin (Soleimanian et al. 2020). Although the reviewer's comment was particularly focused on the L4 period, we would like to point out that the conditions during the 2nd campaign (December 2017) and the first portion of the 3rd campaign (November 2018) were quite similar. Geographically, there were multiple fires throughout the Southern California region in both campaigns (see Figure 3). Both campaigns were also characterized by Santa Ana (i.e., northerly and easterly) winds. The weekly average concentration of levoglucosan between 7 to 14 November and 15 to 22 November was 187.5 ng m^{-3} and 83.89 ng m^{-3}, respectively. Note that the 3rd campaign took place between 12 and 18 November 2018. For

reference, levoglucosan concentrations during July 2018 (non-wildfire season) ranged between ~4 and 17 ng m^{-3}. The elevated concentration of levoglucosan inside the LA Basin during November 2018 removes any lingering doubt that BB aerosols were mixed into the broader regional air mass that was measured on Catalina Island. Given that similar fire and meteorological conditions were present during the 2nd campaign (December 2017), we have high confidence that BB also played at least a minor role in this campaign as well.

Regarding plume age comments:

For the LEO periods mentioned (L3, L8, L9, and L10), the aging timescale range of ~days to a week was meant to serve as a range of possibility rather than an exact aging timescale. We fully acknowledge the limits of HYSPLIT, especially for complex trajectory patterns. That is exactly why we present a very general range of timescales that was based on physical distance from major sources rather than relying on the exact timing of crossovers from the back-trajectories. The reviewer also mentions the loss of rBC coating with aging. This is entirely consistent with the CT_{BC}values measured during periods impacted by long range transport of biomass burning impacted air masses (e.g., L8 and L9). The median CT_{BC} values were within the range of ~60-70 nm during this time period of impact from the Camp Fire. Previous airborne studies have measured average coating thickness values of ~100 nm within hours of emission within the plume. Given that our values are significantly lower, the rBC measured in our study likely did experience coating loss at some point during transport. We added a short discussion on this topic of coating loss in the coating thickness discussion section and below in the section (1.3). Furthermore, we have added a new section that comprehensively compares our campaign measurements with past mixing state studies conducted with an SP2.

Regarding precipitation comments:

Although the data were not reported, precipitation and cloud cover were monitored

throughout the campaigns. There were no precipitation events in the region during any of the measurement days, and most of the days were clear to partly cloudy.

(1.3)

Major edits were made to section 3.5 (formerly section 3.4) and section 3.7 (formerly section 3.6). A new section 3.8 was added to comprehensively compare our results to past similar studies. Additional evidence (i.e. using CALIPSO lidar data, HYSPLIT dispersion model and levoglucosan measurements) and figures were also added to the Supplement to make our discussion on source attribution more robust. Specifically, please refer to Supplement section S2 and figures S11 through S20.

Revised main points regarding variability of coating thickness:

- *Timescales of less than 24 hours were too short to significantly coat rBC from urban emissions. This is in direct contrast to biomass burning rBC, which has been shown in previous works to acquire thick coatings within hours or even minutes, near the source of emission.*
- *Aged rBC from biomass burning sources were generally more thickly coated, although the time evolution of the mixing state could not be quantified directly over the duration of transport. Periods of "fresh" biomass burning impacts were characterized by slightly thinner CT_{BC} compared to aged biomass burning rBC particles (e.g., L3 vs. L8), but larger CT_{BC} compared to fresh urban rBC particle (e.g., L3 vs. L4). This agrees with previous studies that have also observed thicker coatings in fresh biomass burning rBC relative to fresh urban rBC. The overall larger CT_{BC} for aged biomass burning rBC relative to fresh biomass burning rBC indicates that there is significant coating formation that occurs between the timescale of ~1 day to ~1 week for biomass burning rBC, even after rapid coating formation that occurs soon after emission. An important caveat is that CT_{BC} of biomass burning rBC may not be monotonically increasing over time.*

Past studies have observed rapid coating of biomass burning rBC within the first few hours to more than 100 nm, but we observed a median CT_{BC} of 42 nm for L3, which suggests that CT_{BC} for biomass burning rBC might decrease at some point during atmospheric transport and then increase later at longer timescales (e.g., median CTBC of 68.6 nm for L9). We make no definitive claims about the rate of change of CT_{BC} for biomass burning rBC throughout atmospheric transport since we only observed the CT_{BC} from a single discrete point in space, but our measurements do suggest that CT_{BC} for Southern California biomass burning rBC were generally lower than CT_{BC} for Northern California biomass burning rBC.

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(2.1)

Major comments regarding number size distribution data

(2.2)

Although we generally acknowledge the concerns about fitting a log-normal distribution to a set of observations without a discernable peak, we also believe that the log-normal fits have value and should be reported (with associated uncertainty clearly described). First, there have been a number of past studies that have also included log-normal fits for their number size distributions, even in cases where the peak in the measured data was ambiguous. At the end of (2.2) is a comprehensive, but not exhaustive, list of studies using the SP2 that have included log-normal fits to rBC number size distributions. Full references are provided at the end of the document.

Second, the physical lower bound on BC core size makes log-normal fitting reasonable in the Aitken range, even if it is below the SP2 detection limit. Single BC nanospheres (i.e., individual spherules) have been observed to be ~20-30 nm in diameter by using TEM imaging techniques (Ellis et al., 2016; Wentzel et al., 2003). Although the de-

tection limit of the SP2 for rBC cores is ~70 nm, it seems reasonable to assume that the peak of the rBC number size distribution in this Aitken range would be between 50 and 80 nm (Kondo et al., 2011b), given that individual BC spherules are unlikely to be smaller than 20 nm. This would naturally imply that most (if not all) BC cores in the ambient air are larger than 20 nm, but smaller than the point at which we observe a sharp increase in the slope on the right-hand side of the number size distribution. This inflection point on the right-hand side is clearly observed from SP2 data, even when the peak is not completely discernable.

Third, even if there was an unmeasured bimodal peak beyond the detection limit of the SP2, the median of the extrapolated log-normal fit would not be a completely useless metric for comparison. As long as the log-normal fitting is consistent between all instances of distributions, it would serve to characterize the Aitken mode of the rBC core size distribution, even if there was another mode lurking in the ultrafine range. This would suggest that the existence of an unknown local maxima in the ultra-fine range is possible, but that it would not invalidate the inter-comparison of Aitken mode distributions for different time intervals.

Fourth and lastly, the appropriateness of the log-normal fit is not entirely contingent upon the explicit observation of a local maxima. It might be entirely inappropriate if we saw that all the observed data points deviated sporadically from the fit curve, but we observe the fit curve describing the observed number size distribution data points very well, with fairly small residuals. We see that the rate of change of the slope is well captured by the fit, which strongly suggests that a log-normal fit is likely representative of the actual distribution. Analogously, we find the LEO-fit for coating thickness quantification as a robust method for mixing state analysis, even though we only use the leading edge of what we expect to be a Gaussian signal. Indeed, the LEO-fit uses an even smaller fraction of the expected Gaussian scattering response compared to the log-normal fits for the number size distribution. Likewise, we are using the existing “edge” of size distribution to fit what we expect to be log-normal.

To address the reviewer's concern with this issue, we made a clear caveat in the text explaining the limitations of the extrapolation, in addition to the already existing disclaimer about the lower detection limit in the first paragraph of section 3.6 (formerly section 3.5). We made it clear and explicit that the peak based on log-normal fits are not definitive measured values, but rather modeled based on reasonable assumptions about the behavior of the distribution in the Aitken range.

The typo in Figure 8 regarding the wrong median value label has also been fixed.

List of publications that have used log-normal fits to the number size distribution data:

Cheng et al., 2018; Kondo et al., 2011a; Kondo, et al., 2011b; Krasowsky et al., 2018; Metcalf et al., 2012; Moteki et al., 2012; Raatikainen et al., 2017; Reddington et al., 2013; Sahu et al., 2012; Schwarz et al., 2008; Shiraiwa et al., 2008

(2.3)

An additional caveat has been added to section 3.5 (formerly section 3.5) in the manuscript in tracked changes to address the comments and concerns made by RC2.

"Figure 10 shows that log-normal fits adequately capture the measured size distributions, though we cannot rule out the possibility of another rBC mode outside the detection limits of the SP2. Although the peak of the observed points is not always discernible (e.g., number size distribution for L5 in Figure 10), it is reasonable to fit these points assuming that a log-normal distribution is a realistic representation of ambient rBC number size distributions in the Aitken mode. The rate of change of the observed points is also captured very well qualitatively by the log-normal fits, further indicating its appropriateness."

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(3.1)

Major comments regarding increasing rBC diameter with atmospheric aging

(3.2)

We agree with the reviewer that the effect of coagulation on the rBC core size is likely overplayed in the manuscript since the rBC number concentration is relatively low in the ambient air at the point of measurement, compared to the rBC number concentration very close to the source of combustion (e.g., in a tailpipe or in a BB flame). We would like to point out, however, that there is a noticeable shift in the rBC size distributions during time periods dominated by urban emissions (e.g., L4 and L5) relative to size distributions that were measured inside Los Angeles near a major highway by Krasowsky et al. (2018). This is a particularly useful comparison because the exact same SP2 was used with the same operating variables. Focusing on the number size distribution, we observe a larger count median diameter during the L4 and L5 periods compared to the count median diameters measured downwind of a highway in a polluted urban environment (Krasowsky et al., 2018). The size distribution of rBC can only be affected by, (i) the emission source type and/or (ii) coagulation of rBC-containing particles. Related to the reviewer's comment regarding source attribution, we believe that both of these factors likely played some role in the variability of rBC core sizes. We are quite confident that BB sources did contribute, at least in part, to time periods dominated by urban emissions. (see comment block 1 above for details). So, there is likely a source effect. It seems plausible that a mixture of BB impact and coagulation (at least near the source, within the polluted urban basin), contributed to this noticeable shift in the core size distribution.

The reviewer also notes that the cited studies were conducted under higher rBC concentrations than what we encountered in our study. However, while the studies mentioned did have higher campaign-averaged concentrations, the peak concentrations were within the same magnitude, especially for the Shiraiwa et al. study (2008), which took place in the East Asia outflow. The peak magnitudes reported in Shiraiwa et al. reached ~ 1 μg m^{-3}, which is within a factor of two relative to the larger peaks measured in our study (~0.6 μg m^{-3}). Shiraiwa et al. (2008) briefly mention that coagulation could

be a potential mechanism that explains why aged particles from China and Korea were larger than particles associated local urban emissions from Japan. While we agree that coagulation at measured concentrations would be slow and possibly negligible, we believe that coagulation could have played a minor role during atmospheric transport from the LA basin to Catalina Island. We make no attempt at quantifying the rate at which coagulation occurs for LA basin dominated air masses, but we qualitatively acknowledge that coagulation likely contributed to the growth of particles, as per the logic above, especially within the first few hours of aging.

(3.3)

The focus of the paragraph mentioned by the reviewer has been shifted towards an emphasis on source-related impacts rather than impacts from atmospheric processing (i.e., coagulation). A short mention of coagulation still remains, but it serves as a qualitative acknowledgement of its likely minor effect on rBC size distributions. See section 3.6 (formerly section 3.5) for tracked changes.

Relevant excerpts from new text in section 3.6:

"A survey of past studies that have reported rBC mass median diameter (MMD) and count median diameter (CMD) shows that the source of emissions has a strong influence on rBC core diameter (Cheng et al., 2018). The MMD [CMD] for biomass burning influenced rBC, which has been reported to range from ~130 nm to 210 nm [100 to 140 nm], is generally much larger than the MMD for urban emissions influenced rBC, which has been reported to range from ~100 nm to 178 nm [38 to 80 nm] (Shiraiwa et al., 2007; Schwarz et al., 2008; McMeeking et al. 2010; Kondo et al., 2011a; Sahu et al. 2012; Metcalf et al., 2012; Cappa et al., 2012; Laborde et al., 2013; Liu et al., 2014; Taylor et al., 2014; Krasowsky et al., 2018). The MMD [CMD] for aged air masses in remote regions were reported to range from ~180 nm to 225 nm [90 nm to 120 nm] (Shiraiwa et al., 2008; Liu et al, 2010; McMeeking et al., 2010; Schwarz et al., 2010).

Figure 11 shows the rBC MMD and CMD based on the log-normal fits for each LEO

period in this study. Based on the source identification discussed in section 3.1 and section S2 in the Supplement, the MMD and CMD values in this study are generally consistent with the ranges reported in past studies. For LEO periods when measurements were strongly influenced by biomass burning emissions (L3, L8, L9, L10), MMD ranged from 149 nm to 171 nm, which is within the range of ~130 nm to 210 nm compiled from past studies. Similarly, when measurements were strongly influenced by urban emissions (L2, L4, L7), the MMD dropped, ranging from 112 nm to 129 nm. This falls within the range of ~100 nm to 178 nm previously reported for measurements of urban emissions from past studies."

"In addition to varying source type, coagulation is the only physical mechanism that increases rBC core size (Bond et al., 2013). Shiraiwa et al. (2008) observed an increase in rBC core diameters in aged plumes compared to more fresh urban plumes, suggesting that coagulation can alter the rBC size distribution during atmospheric transport (i.e., aging). Although the emissions source type appears to be the dominant influence on rBC core sizes in this study, there is evidence to suggest that coagulation did occur during transport from the Los Angeles basin to Catalina Island (~70 km away) in this study. For example, we observed an MMD [CMD] of 112 nm [53 nm] during L4, when we know that urban emissions were dominant, but this is noticeably larger than values of 93 nm [42 nm] reported in Krasowsky et al. (2018) for measurements conducted 114 meters downwind of a major highway. Furthermore, Laborde et al. (2013) observed an MMD of ~100 nm when impacted by fresh traffic emissions in Paris. Even though it was determined that L4 was predominantly urban emissions influenced, we cannot rule out the possibility of local wildfires influencing the size distribution as well. While the rBC size distribution from L4 suggests that coagulation plays at least a minor role, both factors (source type and coagulation) likely influence rBC size distributions to varying degrees in areas with heterogenous source profiles and relatively elevated rBC concentrations (e.g., polluted urban areas)."

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(4.1)

Page 1, line 18, 20. The passive voice exemplified by the use of the word "suspect". Are the author's hedging their bets? Suggest using a different - less passive - word.

(4.2)

The wording has been changed.

(4.3)

New text:

"In contrast, during periods when measured rBC was dominated by emissions from the Southern California region, both fBCand CT_{BC} were significantly lower, with a mean fBC of ~0.03 and median CT_{BC} ranging from ~0 to 10 nm."

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(5.1)

Page 1, lines 23-25. The author's write "we conclude that an aging timescale on the order of ~hours is not long enough for rBC to become thickly coated under the range of sources sampled and atmospheric conditions during this campaign." This is misleading as several papers that have studied biomass burning (and those currently under review and data currently being analyzed) have (and are) showing that rBC particle become thickly coated very quickly. While this might be true for urban plumes, it certainly is not for BB (biomass burning) plumes. Please clarify.

(5.2)

We agree with the reviewer and we have changed the main conclusions of our paper to reflect this. Further response to this specific issue has been discussed in more detail

above in comment block 1.

(5.3)

Any text related to the generalization of thin coatings for particles aged less than 24 hours has either been removed or modified.

This was also discussed in greater detail in comment block 1 and applicable changes have been made in sections 3.5 and 3.7 (formerly sections 3.4 and 3.6).

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(6.1)

Page 2, lines 43-44. The author's write "BC is emitted mostly as an "external" mixture, physically separated from other aerosol species." This is a bit misleading. It is very dependent upon when the plume is sampled. With respect to biomass burning, research has shown that rBC becomes coated within the first few minutes following generation due to the chemical richness of the smoke plumes.

Please reword to reflect this.

(6.2)

We acknowledge that BC can become coated very quickly and that this statement could potentially be misleading. The original intent was to give a conceptual overview of externally versus internally mixed BC. The description has been altered to remove any ambiguities regarding emission point and timescale since emission.

(6.3)

The text in the introduction (section 1) has been altered to describe the two general types of mixing state without potentially misleading readers into believing that all BC is uncoated in the near-field plume.

"A hypothetical BC particle that is completely, physically separate from other non-BC aerosol species is considered externally mixed. On the other hand, BC is considered internally mixed if it is physically combined with another non-BC aerosol species (Bond et al., 2006; Schwarz et al., 2008). As freshly emitted BC particles are transported in the atmosphere, they can obtain inorganic and organic coatings from either gaseous pollutants that condense onto the BC, oxidation reactions on the BC surface, or the coalescence of other aerosol species onto the BC, making them more internally mixed (He et al., 2015). In short, externally mixed BC is referred to as "uncoated BC" and internally mixed BC is referred to as "coated BC." In general, the mixing state of BC describes the degree to which BC is internally mixed, with uncoated (i.e., externally mixed) BC particles on one end of the mixing state spectrum (Bond et al., 2013). The BC mixing state near the point of emission as well as the evolution of the mixing state can vary widely, depending on the source of emissions and atmospheric context."

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(7.1)

Page 3, line 74 and 75. The authors need to be very disciplined in their use of "mixing state", as one can be describing the aerosol mixing state (e.g., external vs internal) or the particle mixing state (e.g., coated or uncoated rBC). Yes, the authors sort of point this out on page 2 (lines 48-50) but then start interchanging "internal mixing state" with mixing state. For example, on the opening sentence of the cited paragraph, are the authors referring to the internal mixing state or the aerosol mixing state? Later in this paragraph, the authors reference internal mixing state of rBC (line 80). Please ensure consistency.

(7.2) We acknowledge this potential for confusion and changed the language throughout the manuscript to ensure consistency. For the sake of simplicity and consistency, we initially define externally mixed BC as "uncoated BC" and internally mixed BC as "coated BC." Furthermore, we use the general term "mixing state," to refer to the extent

to which BC is coated, either at an individual particle level or aggregated (i.e., sample population-wide) level.

(7.3)

We edited the text to ensure consistency between any language describing the mixing state. This topic was also discussed in comment block 6 above.

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(8.1)

Page 3, lines 74 - 75. Here are two additional references to the use of microscopy with quantifying rBC mixing state that the authors are encouraged to consider: Adachi, K., Chung, S. H., and Buseck, P. R.: (2010) Shapes of soot aerosol particles and= implications for their effects on climate, J. Geophys. Res. Atmos., 115. Adachi, K., Moteki, N., Kondo, Y., and Igarashi, Y.: (2016) Mixing states of light-absorbing particles measured using a transmission electron microscope and a single-particle soot photometer in Tokyo, Japan, JGR.,121, 9153–9164.

(8.2)

Thank you for the references and suggestion. They have been added to the manuscript.

(8.3)

These references have been added to the introduction of the manuscript where microscopy is briefly mentioned.

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(9.1)

Page 3, lines 80 - 83. Authors are encouraged to review (include) the work by Sedlacek et al., who investigated the utility of the SP2 lagtime methodology [In-

vestigation of Refractory Black Carbon-Containing Particle Morphologies Using the Single-Particle Soot Photometer (SP2) (2015) Aero. Sci. Tech., 49:872]

(9.2)

Thank you for the suggestion. We have incorporated this reference into our study and expanded on our analysis by including discussion about negative la-times and rBC morphology in the discussion section. See also (11.1) below, which is related to this comment.

(9.3)

See section 3.4 on negative lag-times and rBC morphology for newly inserted analysis and discussion.

Excerpt from new text:

"In this study, we observed negative lag-times, although at a relatively low rate, with flag,neg calculated to be much less than 0.1 throughout most of the measurement periods. We defined flag,neg to be identical to the "fraction of near surface rBC particles" metric used by Sedlacek et al. (2012), using a lag-time threshold of -1.25 μs to account for uncertainties associated with the lag-time determination. The campaign-wide flag,neg was 0.017 for the first campaign (September 2017), 0.018 for the second campaign (December 2017), and 0.026 for the third campaign (November 2018). Comparatively, Dahlkötter et al. (2014) observed flag,neg of ~0.046 during an airborne field campaign measuring an aged biomass burning plume in Germany, and a much higher disintegration rate of ~0.4 to 0.5, based on a method that examines the tail end of the time-dependent scattering cross-section (Laborde et al., 2012). Sedlacek et al. (2012) reported flag,neg of more than 0.6 for ground-based measurements of a biomass burning plume in Long Island, New York, originating from Lake Winnipeg, Canada; and the scattering-cross section method was not used to calculate an additional disintegration rate."

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(10.1)

Page 3, line 83. The authors are encouraged to review (include) the work by Moteki and Kondo who have also contributed significantly to improving the quantification of the rBC mixing state [Method to measure time-dependent scattering cross sections of particles evaporating in a laser beam (2008) J. Aer. Sci. 39:348].

(10.2)

Thank you for the suggestion. This study was not initially included in the manuscript because the method described in Moteki and Kondo (2008) was not used for our mixing state analysis. Nevertheless, we have added the reference in the initial description of the LEO method because of its relevance to the Gao et al. (2007) method, which we used in our study.

(10.3)

The reference has been added to section 2.7 in the manuscript.

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(11.1)

Page 9, lines 226 - 228. The authors might consider reviewing (including) the work by Sedlacek et al., who looked at the SP2 lagtime for a biomass burn plume. [Determination of and Evidence for Non-core-shell structure of particles containing black carbon using the single particle soot photometer (SP2). (2012) GRL. 39]

(11.2)

Thank you for the suggested work. We have added an additional short section about the morphology of rBC in the results and discussion section of the manuscript, and we use the same near-surface fraction analysis that Sedlacek et al. (2012) employed in

their study. The reference has been added as well.

(11.3)

See section 3.4 on negative lag-times and rBC morphology for newly inserted analysis. Also see comment block 9 above for related discussion.

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(12.1)

Page 10, Line 266. As highlighted earlier, please refrain from relying on a passive voice (e.g, "suspect".)

(12.2)

Passive voice removed.

(12.3)

The word "suspect" has been removed from referenced text.

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(13.1)

Page 12: The authors show the back trajectories for each day of the campaign. Why not put this figure in the supplemental and, instead, show those trajectories for the specific periods under discussion. This would make it easier to evaluate the HYSPLIT datasets.

(13.2)

Thank you for the suggestion. Although we see the value in the suggestion, we prefer to leave Figure 3 in its current state and add a*separate* HYSPLIT figure either in Section 3.7 or in the Supplement. Our reason for showing all the trajectories in Figure 3 is to show the campaign-wide perspective on the source locations of the particles. We also

thought it would be useful for visually comparing between the different campaigns, and not just for 10 to 15-minute LEO time periods, which give limited snapshots instead of showing a broader campaign-wide "fingerprint" of trajectories.

(13.3)

Additional figure with only LEO period back-trajectories has been added to the Supplement. This can also be added to Section 3.7 if it is determined to be more appropriate there.

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(14.1)

Page 14 line 307. The authors reference Figure S9, but I think they mean S8?

(14.2)

Thank you for catching this typo.

(14.3)

Changed from Figure S9 to Figure S8.

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(15.1)

Page 15, lines 337 - 344. The authors are encouraged to review paper by Subramanian et al., [(2010) Black carbon over Mexico: the effect of atmospheric transport on mixing state, mass absorption cross-section, and BC/CO ratios ACP 10] where attention is drawn specifically to figures 3, 12 and 13.

(15.2)

Thank you for the paper suggestion. The figures you suggested were carefully reviewed and they were helpful in putting our results in context of past studies like Sub-

ramanian et al. (2010). Brief comparisons are made to the results presented in Subramanian et al. (2010) to our results. Reference to the article has also been added to the manuscript.

(15.3)

See minor additions in Section 3.3 and Section 3.7.

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(16.1)

Page 22, lines 507 - 508. As highlighted above, this reviewer has concerns regarding the estimated plume ages.

(16.2)

Appropriate changes have been made to the main conclusions from this paper, as described in more detail in Comment 1.1 above. Most importantly, all blanket statements regarding an aging timescale of more than one day required for thick coating have been altered or removed.

(16.3)

See revised manuscript for tracked changes.

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(17.1)

Supplemental: page 1. As noted earlier, there is no table 3 in the main manuscript.

(17.2)

The table was accidently omitted. Apologies for any confusion.

(17.3)

Table 3 has been merged with Table 2. The old Table 3 is now part of Table 2.

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(18.1)

Supplemental: page 1, line 7. Suggest that the authors review Lund et al., [(2018) Short Black Carbon lifetime inferred from a global set of aircraft observations, npj Climate and Atmospheric Science 1, 31 doi:10.1038/s41612-018-0040-x]

(18.2)

Thank you for the suggested article. The mean BC lifetime of ~4 days over the Pacific as suggested by Lund et al. (2018) further supports our estimated range of source-to-receptor timescales. We would like to clarify here that our loosely restrained timescales are only meant to give readers an idea of the range of possibilities regarding how long measured particles were transported in the atmosphere. Since the estimated value of ~4 days was meant to represent the mean, individual particles measured during our campaigns could certainly have been aged longer (i.e. ~week).

(18.3)

Citation added to text.

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(19.1)

Supplemental: page 7. This is a stylist comment. Would suggest using a different color to denote the sample location on Catalina Island. The currently used green color is hard to discern with the yellow star.

(19.2)

Style change made as suggested.

(19.3)

Green circle removed from the figure as it was unnecessary.

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