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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.

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Author Response to RC1

We appreciate the thoughtful and detailed review from Referee 1. We have taken the comments made by Referee 1 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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Discussion paper

- 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)

Unless I have misunderstood something, the core conclusions of this paper regarding coatings with ageing timescales seem to be based on the assumption that both urban and biomass burning BC are emitted with thin coatings. However, there is much evidence to the contrary, as most SP2 measurements of biomass burning at or near source would indicate that they have thick coatings at the point of emission. Furthermore, the thickness of this coating can vary significantly fire to fire (see https://www.atmos-chem-phys.net/14/10061/2014/ https://www.nature.com/articles/ngeo2901, https://www.atmos-chem-physdiscuss. net/acp-2019-157/). It therefore doesn't seem correct to infer conclusions regarding the effect of ageing timescales on coating thicknesses when comparing aerosols from different sources. The authors should review their findings taking this into consideration.

(1.2)

We agree that that it was presumptive and largely erroneous to make blanket statements about coating thickness without properly taking the emission source(s) into account. As the reviewer noted, we agree that the existing literature shows overwhelming evidence that biomass burning rBC particles are quickly coated after being emitted, and that they are, on average, significantly more coated than urban rBC particles of comparable atmospheric age. We made major revisions to section 3.7 (formerly section 3.6) and shifted the focus away from the aging timescale. The focus is now on the differences in mixing state during different identifiable source impacts (e.g., biomass

burning versus urban). We left some discussion in the manuscript about the effects of aging on mixing state, but we made sure to keep comparisons consistent between the same source or mix of sources.

(1.3)

See section 3.7 (formerly section 3.6) for major revisions shown in tracked changes. Please also see the Supplement for additional information on source attribution and also new additional analysis including measurements of levoglucosan (i.e., a good tracer for biomass burning) and lidar data from CALIPSO.

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(2.1)

The conclusions section is long but mainly seems to recap the earlier observations rather than focus on the key scientific advancements being offered by this work. In order to properly judge this aspect of the paper and therefore its suitability for publication, this should be restructured.

(2.2)

We agree that the conclusion should be restructured and focused on the most salient "key scientific advancements" rather than merely "recapping earlier observations." That being said, we also believe that recapping key observations and details in the conclusion section may be valuable to readers who might be quickly reading through the paper, trying to glean the most important take-away points from the abstract and/or conclusion.

Regarding significant changes to the conclusion, we have added clarification to the key scientific advancements. These key points have been slightly modified in light of comment (1.1) and additional comments from Referee 2. In addition, some extraneous details have been stripped from the conclusion as suggested. The main conclusion points of the manuscript are summarized below.

(2.3)

Conclusion Point 1: The rBC size distribution was strongly affected by the emission source type. rBC particles measured during periods of biomass burning impacts had larger rBC core diameters (MMD ~ 150 to 170 nm) relative to rBC particles measured during time periods dominated by urban emissions (MMD ~ 110 to 130 nm). rBC particles from well-aged, continental air masses were characterized by an MMD ~ 150 nm, which likely reflects a mix of large-scale transported BC from unidentified biomass burning and urban emissions.

Conclusion Point 2: During urban emissions influenced time periods, rBC particles were found to be either uncoated or very thinly coated, with median CT_{BC} less than ~15 nm and average fBC less than ~0.15.

Conclusion Point 3: During biomass burning influenced time periods, rBC particles had thicker coatings overall, with median CT_{BC} ranging from ~40 to 70 nm and fBC ranging from ~0.23 to 0.47.

Conclusion Point 4: rBC particles from continental air masses coming from across the Pacific Ocean were found to have moderately thick coatings, with an average CT_{BC} of ~30 nm and fBC of ~0.27.

Conclusion Point 5: 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.

Conclusion Point 6: 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 CT_{BC} 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 CTBCf 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.

Conclusion Point 7: The high variability of the rBC measurements on Catalina Island during three different campaigns demonstrates how meteorology, emissions source type, and atmospheric aging can drastically affect the physical properties and mixing state of the broader BC population within the same region.

See the updated manuscript (with track changes) for comprehensive view of changes made in the Conclusion section.

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(3.1)

Measurements of coating thickness can become biased if the particles are sufficiently small that the signal-to-noise ratios of the instrument's scattering channels aren't sufficient to successfully retrieve a coating thickness or a delay time. Was the rate of failed retrievals monitored? How was this reflected in the data?

(3.2)

To minimize the signal-to-noise ratio for the LEO analysis, we only considered LEO-fits for particles with rBC core diameters between 200 and 250 nm (as mentioned in Section 2.7). For the particular SP2 unit used in our study, Krasowsky et al. found that a lower threshold of 200 nm was sufficient to reduce the scattering signal-to-noise ratio (Krasowsky et al., 2018). Previous work by Taylor et al. (2014) defined a lower bound of 135 nm for LEO fitting, which corresponds to a 50% fraction of rBC with detectable split-detector notch position. We chose an even more conservative lower bound of 200 nm in this study to further minimize the scattering signal noise. The 250 nm upper bound is also conservative, considering that previous studies have reported LEO-fit coating thicknesses for particles with rBC core diameters up to 290 nm (Dahlkötter et al., 2014). To check for potential biases due to the saturation of the scattering signal at larger rBC diameters, a subset of the SP2 data (from 7 September 2017) was assessed to see what proportion of the low-gain scattering channel data were saturated. None of the particles (in this subset of data) with rBC core diameters under 250 nm were found to have saturated scattering signals. The fraction of rBC-containing particles that were selected for LEO-fitting (with respect to all rBC particles measured) was not explicitly reported in the manuscript, but the total number of particles analyzed in each LEO period was listed in Table 2. A number of previous studies have also reported only the size range of LEO-fit rBC particles, without explicitly stating the fraction of particles that were LEO-fit versus not LEO-fit. We think that including the size range of the LEO-fit particles and the total number of particles analyzed in the manuscript is sufficient to show that we adequately constrained noise in the scattering signal and also analyzed enough particles to produce robust coating thickness statistics for the L1 to L10 analysis.

For the lag-time (delay time) analysis, a lower threshold of 170 nm was implemented for rBC core diameters. The reasoning for the lower limit is the same as explained above. The only difference is that we relaxed the lower threshold a bit (compared to 200 nm)

because the accuracy of the scattering signal is not as crucial to the binary categorization of rBC-containing particles as “thickly-coated” versus “thinly-coated.” Previous studies have conducted the delay time analysis with similar size ranges, or even lower thresholds (Krasowsky et al., 2018; Shiraiwa et al., 2007; McMeeking et al., 2011; Moteki and Kondo, 2007; and more).

(3.3)

A sentence will be added to Section 2.6 to clarify that the lower threshold for rBC core diameter was set to 170 nm for the lag-time (delay time) method.

”Only particles with an rBC core diameter greater than 170 nm were included in the calculation of fBC to account for the scattering detection limit of the instrument.”

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(4.1)

Setting ’calm’ winds as zero on direction on figure 5 makes no sense as this also corresponds to north. The periods should probably be blanked out instead.

(4.2)

Calm winds have been removed from the wind direction plot as suggested.

(4.3)

See updated Figure 5.

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(5.1)

The points plotted on figure S9 should be individually identified according to event.

(5.2)

Figure S9 (now Figure 12) has been modified to show the scatter between coating thickness and rBC core diameter for all measurements. 1-minute mean values for both coating thickness and rBC core diameter are used for the scatter plot. A statistically significant positive correlation was found and is shown on the figure (r = 0.5397 and τ = 0.3766, with p-value << 0.05).

(5.3)

See updated Figure S9 (now Figure 12 in main manuscript).

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