Response to reviewer

We thank the reviewer for constructive suggestions, which have helped improve the manuscript. This document outlines the review comments in *plain italics*, followed by the author's response in bold, and the tracked changes in the main texts are in blue.

Reviewer's comments:

The authors have substantially improved the paper. I have a few very minor comments related to the responses for the authors to consider.

1. Original Comment: Page 3, Line 21 - What is it about sources of BC in a clean environment that reduce its relative aging? Are BC emissions unaccompanied by fewer other emitted components?

Authors Response: Yes. Higher concentrations of condensable vapors (some of which are co-emitted species) contribute to faster aging of BC (Wang et al., 2014; Peng et al., 2016) in polluted environments, as compared to cleaner environments.

New Comment: When you refer to sources of BC in a clean environment, are you saying that there are no co-pollutants emitted with BC? What might be some sources of BC for which co-pollutant emissions are of less or little relative consequence?

Sorry for the confusion created here. 'Relatively cleaner environment' refers to the regions where fewer amounts of condensable vapors prevail, thereby contributing to less-faster aging of BC than the polluted regions. As the reviewer rightly pointed out, BC sources alone may not play much role in determining its relative chemical-ageing. This portion was revised in the first revison of the manuscript and is given below.

" The alteration to BC mixing state depends on various factors, which include the BC size distribution, nature of sources, the concentration of condensable materials that BC encounters during its atmospheric lifetime, and processes such as photochemical ageing (Liu et al., 2013; Ueda et al., 2016; Miyakawa et al., 2017; Wang et al., 2018). Consequently, the nature and extent of coating on BC vary in space and time, and as such, BC in a polluted environment chemically-ages faster than in a relatively clean environment (e.g., Peng et al., 2016; Liu et al., 2010, 2019; Cappa et al., 2019)."

2. In response to a comment regarding lines 30-32 of the original manuscript, the authors say "When air masses from such complex source regions are transported to remote regions devoid of any BC sources, the mixing state of BC becomes complicated. This is due to (a) restructuring of the BC aggregates during the transport due to different processes (Kutz and Schmidt-Ott, 1992; Weingartner et al., 1995; Slowik et al., 2007b; Pagels et al., 2009), and (ii) varying nature and amounts of coating material arising due to the

different atmospheric lifetimes and microphysical processes involving different species (McFiggans et al., 2015)."

New Comment: Rather than say the mixing state become more complicated, which I think is arguable, I suggest saying the mixing state may change. Also, make the bullets consistent.

Complied with. We have incorporated these suggestions in the revised manuscript.

3. Original Comment: Page 4, Line 28 - Do you know that particles smaller than 10 um were efficiently sampled or are you just assuming they were?

Authors Response — "Prior to the experiment, we have characterized the sampling inlet system and examined the sampling losses, both theoretically and experimentally. For this purpose, the particle number size distribution measurements were used. We found that the difference between the number concentrations with and without inlet system was < 5% for sizes up to 1000 nm, 5-20% between 1000-6000 nm, and > 30 % for sizes > 6000 nm."

New Comment: Good, but do you mention these results anywhere in the manuscript or supplement? We have not mentioned these details in the manuscript. The inlet design is based on Global Atmospheric Watch (GAW) guidelines/recommendations for aerosol sampling (WMO/GAW, 2016).

4. New Comment: You use "aged" in many places, but it is an ambiguous term. 'Aged' is a temporal term, yet here its use in aerosol science most often pertains to the particle chemical composition rather than time. The Arctic aerosol can be one example of an aged aerosol both temporally and chemically. On the other hand, a biomass burning or biogenic aerosol may be well aged in a chemical sense (although not necessarily completely) on a much shorter time scale. It seems unreasonable to call them both 'aged' without any qualification. I suggest using "chemically-aged".

Complied with. We have explicitly stated 'Chemically-aged" in the appropriate text throughout the revised manuscript.

5. New Comment concerning comment on Page 11, Line 33 of original manuscript: Rather than "The insoluble BC particles...", I suggest "Less-soluble BC-containing particles may be interstitial within a non-precipitating cloud."

Complied with.