Response to the reviewer 2 comments for "Mixing states of Amazon-basin aerosol particles transported over long distances using transmission electron microscopy" by Adachi et al.

Referee comments are shown in black, normal font.
Authors replies are shown in red bold font.
Revised texts are shown in red, bold and italic font.

Anonymous Referee #2
Received and published: 4 July 2020
The study works on individual aerosol particles collected in Amazon basin where is important for carbon cycle as the green ocean in the world. The authors collected the aerosol particles during the Green Ocean Amazon compaign at T3 site. They used TEM to analyzed different kinds of particles during the LRT particles. They carefully divided particle types based on their compositions and mixing states. Also, they accounted for their number fractions and compared them during LRT and non-LRT periods in the Amazons. Certainly, the study is critical important to understand properties of background aerosols and LRT particles in the Amazons. Finally, they found these particles are internally mixed particles, in particular, sulfate and sea salts as important coatings. Therefore, I think that the knowledge is filled into the gaps in the Amazon.
Before the paper accepted, I would like have some minor comments here.

Authors reply: We appreciate the reviewer's comments to improve our manuscript. We considered all reviewer's comments and revised our manuscript based on the suggestions.

2-1. (1) I might suggest that the authors need to add clear TEM images as the Figure 1 including different types of particles, such as K-bearing, Sulfate, soot, and mineral particles. Then you can give the Figure showing their number fractions. The potential readers might be know better clear the particle types.

We added representative TEM images of each particle type in the revised Fig. 1.

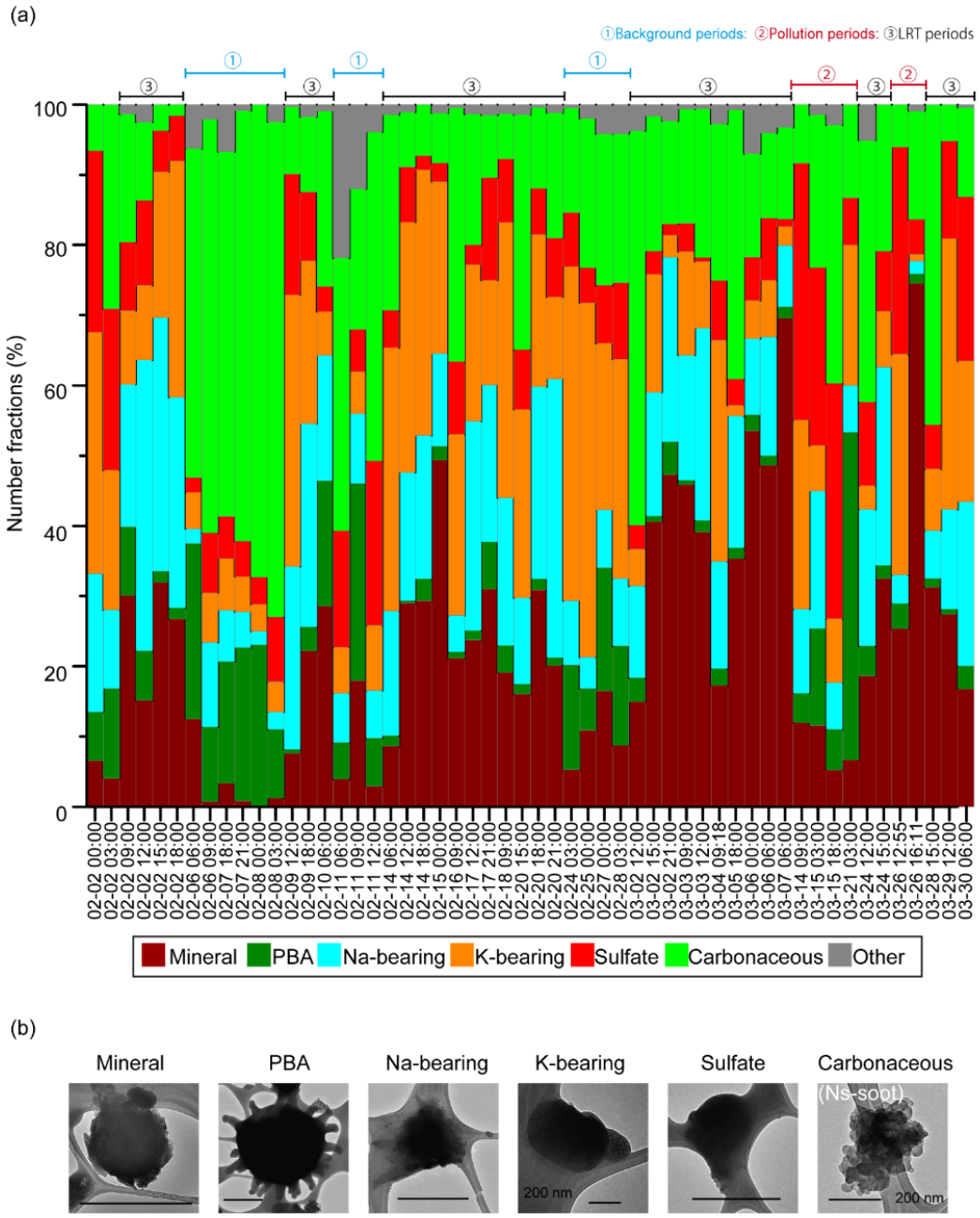

Figure 1. Number fractions of each aerosol particle type for all analysed samples (a) and the representative TEM images of each particle type (b). Scale bars for Mineral, PBA, Na-bearing, and sulfate are 1 μm, and those for K-bearing and carbonaceous are 200 nm. Compositions and low-magnification images of these particles are shown in Fig. S2 (mineral), Fig. S5 (PBA), Fig. 11 (Na-bearing), Fig. S7 (K-bearing), Fig S8 (sulfate), and Fig. S7 (carbonaceous). Samples were divided into LRT and other periods (bars in the panel (a)). Other periods were further divided into pollution periods (14, 15, and 26 March, 2014) and background periods

(all other periods except the pollution periods and 2 February, 2014) based on the classification by de Sá et al. (2018). Samples collected on 2 February were outside of the measurement period by de Sá et al.

2-2. (2) It is better to add more discussion. I noticed that they didn't deploy more discussion in discussion part. I might suggest that you need to consider the result from LRT and non-LRT. What are they different? And how could they have large impacts on ecology or climate? Also, the authors can add discussions on mixing structure of different aerosol components. These could be have more literature and scientific discussion should be enhanced.

We combined the results and discussion sections in the revised manuscript (please also see our reply to 1-2). Now we have only "results and discussion" section and have more discussion than the original based on the reviewers' suggestions.

Examples of additional discussions.
The size-dependent number fractions of samples from other period are roughly consistent with the results during the AMAZE-08 campaign (Pöschl et al., 2010).
Nitrate can also react with NaCl and form sodium nitrate. However, we did not observe it in this study because of lower concentrations of nitrate than sulfate during the campaign (de Sá et al., 2018) and insensitivity of nitrate detection in the current TEM analysis.
Wang et al. (2016a) showed that small particles are transported from the free troposphere into the boundary layer by vertical transports during precipitation events, and such particles can be nuclei of the SOA particles in the background condition.
As the LRT particles have more processed mixed particles than other background particles, we interpret that the mixings mainly occurred during the LRT through coagulation, condensation, or both.

Some minor comments:
2-3. L58: coated by acidic gas condensation

Revised as follows.
Aerosol particles coagulate and can become coated by volatile organic compounds or acid gas condensation.

2-4. L165 during the IOP1 (Figure 2)

It was revised as follows.

The model simulation shows that the horizontal distributions of mineral dust concentration at the surface level during the six LRT periods and the vertical distributions on 7 March, when the largest LRT periods occurred during IOP1 (Fig. 4).

2-5. L200, Did you observe the NaNO3? NaCl can easly react with HNO3 and H2SO4 forming NaNO3 and Na2SO4.

We do not have evidence of NaNO3 in our measurements, although we observed sodium sulfate (original Fig. 7, now Fig 11) and ammonium sulfate (Fig. S8). The possible reasons are as follows. First, nitrate concentration was low, i.e., ~20% of that of sulfate; 0.03-0.12 (nitrate) and 0.15-0.57 (sulfate) μgm-3 (de Sa et al., 2018). Second, nitrate can be volatile and be lost during sampling, storage, and analysis. Third, TEM analysis is insensitive to detect nitrate when it occurs with ammonium sulfate because both include N. Fourth, we measured relatively small sea-salt particles, whereas nitrate prefers to stay with large particles. We added the following explanation in the revised text.

Nitrate can also react with NaCl and form sodium nitrate. However, we did not observe it in this study because of lower concentrations of nitrate than sulfate during the campaign (de Sá et al., 2018) and insensitivity of nitrate detection in the current TEM analysis.

de Sá, S. S., et al.: Urban influence on the concentration and composition of submicron particulate matter in central Amazonia, Atmospheric Chemistry and Physics, 18, 12185-12206, 10.5194/acp-18-12185-2018, 2018.

2-6. L223, you observed the 600 nm and >2 um. Recently, Li et al., (2020) found two modes, suggesting different types of biological particles. I am wondering whether the authors observe observed more types with different morphologies. Here the PBA name can be changed to PBAP. PBAP can be more acceptable in many studies such(Després et al., 2012); (Pöhlker et al., 2012); (Li et al., 2020).

We did not find specific bioaerosol particles in the small size. We think fragmented fungal spores (China et al., 2016) contributed to these particles. Although both PBA and PBAP are used for the acronym, we prefer keeping PBA in our manuscript because we termed aerosol species as xx + particle (e.g., sea-salt + particle and mineral + particle). Please find that Pöhlker et al. (2012) uses "primary biological aerosols (PBA)" in their Fig. 4.

These particles include fungal spores, pollen, bacteria, algae, protozoa, and fragments of

plants and organisms (Després et al., 2012; Li et al., 2020) and are known sources of K, Na, P, Cl, and Fe in the Amazon basin (Artaxo et al., 1993; Krejci et al., 2005; Wu et al., 2019). PBA particles >2 μm show unique shapes (Fig. 9), whereas small PBA particles can include fragmented fungal spores (China et al., 2016).

2-7. L227-229,The diurnal variations can commonly occur in the forests in the worlds. The emission mechanisms could be very complicated depending on winds, RH, and Temperature on plants and soil (Li et al., 2020)(Elbert et al., 2007).

Agree. We revised the sentence as followed.
The number fractions of PBA particles during the night can be enhanced by high relative humidity (RH), which increases active PBA particle emissions from plants, wind speed, and temperatures (Li et al., 2020; Elbert et al., 2007; Graham et al., 2003; Huffman et al., 2012; Whitehead et al., 2016).

2-8. L251, Again, what about nitrate.

We added the following sentence here.
Nitrate can also react with NaCl and form sodium nitrate. However, we did not observe it in this study because of lower concentrations of nitrate than sulfate during the campaign (de Sá et al., 2018) and insensitivity of nitrate detection in the current TEM analysis.

2-9. L270, Here the authors found 10% sulfate. Maybe need to add one TEM image to show their morphology in Figure 1?

We added a sulfate image in Fig. 1 as well as in Fig. S8.

2-10. L289, Maybe add one TEM image showing the SOA and POA? As the authors found many sulfate particles. The SOA is internally mixed with sulfate as its coating? For example, Li et al. (2016) shows most of SOA coat on secondary inorganic particles in the forest and remote air.

SOA can be a coating over sulfate or others. We added a sentence in Fig. S8 caption. The representative images are shown in Fig. S3, S8, and S9. SOA was also marked in revised Fig. 11.
Carbon occurs around the sulfate, showing that the sulfates are coated by secondary organic

matters and have ns-soot inclusions.

2-11. L294, why did the authors used biofuel not biomass here?

During the wet season, there is almost no biomass burning. Instead, local residents burn biofuel for cooking or brick kilns, which contributes to the source of ns-soot.
Ns-soot particles that are internally mixed with K-bearing particles can originate from LRT (Saturno et al., 2018) or local biofuel burning for cooking or brick kilns (Martin et al., 2016).

2-12. L308 other aerosol components
2-13. L309 form various internal mixtures. As Li et al., raise the concept models of mixing structure. The sulfate can be core with SOA coating. They also can be coating on mineral, POA or others.

The sentence was revised as follows. Please also see our replies to 2-10.
Sulfate particles are commonly coagulated or condensed with other aerosol components to form various internal mixtures.