

Answers to the review of anonymous Referee #1

We thank Referee #1 for reviewing our manuscript and giving useful suggestions. Below, comments from the referee are given in blue while our answers are given in black. In addition, the new text is marked blue in the revised version of the manuscript.

Review of Pei et al.

This work presents the morphological transformation of soot during condensation of sulfuric acid and limonene SOA. Besides the present work proposes a framework (method) to quantify the parameters of this morphological transformation, i.e. amount of material used for filling voids or diameter growth and fractions of internal/open voids. Overall, this manuscript is well organized and written, the results are clearly presented, and the scientific novelty is significant for the society. However, the MS still needs minor revision and some technical modification. After that, I believe this paper can be published on ACP.

Comments:

1. Abstract: some quantitative result should be added in the abstract rather than general description.

Response: We agree with the referee that some quantitative results should be added.

Action: One sentence “For the largest coating experiments, 6%, 10%, 24% and 58% of condensed material went to filling process, while 94%, 90%, 76% and 42% of condensed material went to growth process for 75 nm, 100 nm, 150 nm and 200 nm soot particles, respectively.” has been added. (See page 1, line 28-31 in the revised manuscript).

2. The expression “framework” is strange.

Response: This is new word in context to morphological transformation of soot aggregate. The methodology of quantifying the filling of voids and growth of mobility diameter with respect to ideal line is referred as “framework”. The framework here is method to quantify. “Framework” seems the most appropriate word in this context.

Action: No change.

3. Line 11: “soot. This work constitutes the first study that quantitatively tracks in-situ microphysical changes in soot morphology”. I don’t think it’s true.

Response: This is a work that had quantified the in-situ morphological transformation of soot aggregate i.e. filling of the voids and growth of particle, nevertheless we agree with the referee that the morphological transformation of soot aggregate has been studied in the previous studies.

Action: We have removed word “first” and modified the sentences wherever appropriate. (See page 2, line 6 and page 18, line 26 in the revised manuscript).

4. Page 1, Line 18 and in the whole manuscript. It is better to use “soot aggregate”, not “soot agglomerate” to keep consistent with most of the literature.

Response: We agree with the referee.

Action: We have changed all “soot agglomerate” to “soot aggregate” in the whole manuscript.

5. Page 1, Line 32: Change “Growth” to “growth”.

Response: We agree with the referee.

Action: Done. (See page 1, line 24 in the revised manuscript).

6. Page 2, Line 1-2: change ‘ ’ to “ ”

Response: We agree with the referee.

Action: Done. (See page 1, line 26 in the revised manuscript).

7. Page 5, Line 27-30: this paragraph describes the steps of the experiments, however, the experiment of soot coated with only SOA is ignored, it should be stated clearly.

Response: We agree with the referee the description of experiment of SOA coated soot should be added.

Action: The whole sentence is changed to “The experimental matrix included four major procedural steps: 1) characterization of the pure soot core; 2) modification of the soot core surface with sulfuric acid and characterization of the modulated soot; 3) modification of soot core surface with SOA and characterization of the modulated soot; 4) modulation of the sulfuric acid modified soot with limonene ozonolysis products and subsequent characterization.” (See page 5, line 24-28 in the revised manuscript).

8. Page 7, Line15-16: Kuwata et al (2012) did not report this 1.26 number but provide a method to calculate the density, this sentence should be checked.

Response: We agree with the referee that the statement is not clear.

Action: the sentence “This value is similar to the organic aerosol density ($1.26 \pm 0.04 \text{ g cm}^{-3}$) estimated from O:C and H:C determined via SP-AMS measurements performed by Kuwata et al. (2012), and the value ($1.3 \pm 0.2 \text{ g cm}^{-3}$) reported by Saathoff et al., (2009). ” has been changed to “In this study,

aerosol mass spectrometer (AMS) was also used in parallel with DMA-APM system. The O:C and H:C determined via AMS measurements were used to estimate the organic aerosol density ($1.26 \pm 0.04 \text{ g cm}^{-3}$) with the method given by Kuwata et al. (2012). The material density of limonene ozonolysis products (1.2 g cm^{-3}) used in this study is similar to the AMS results and the value ($1.3 \pm 0.2 \text{ g cm}^{-3}$) reported by Saathoff et al., (2009).” (See page 7, line 14-18 in the revised manuscript).

9. Page 7, Line 18: change the reference format to Saathoff et al., (2009)

Response: We agree with the referee.

Action: Done. (See page 7, line 17-18 in the revised manuscript).

10. Page 11, Line 6: should be sections 3.3 and 3.4

Response: We agree with the referee.

Action: Done. (See page 11, line 15 in the revised manuscript).

11. Table 3. I suggest that the authors should compare their work with other studies, e.g. Khalizov et al., EST 2013 etc.

Response: We thank for the referee’s suggestion. However, Khalizov et al. (2013) does not present their results of the diameter growth factor (Gfd) as function of coating thickness (Δr_{me}), ($-\Delta r_{me}$ for them cannot be estimated to compare with us) and the maximum coating thickness in this literature is only 6 nm. These two factors make **it difficult to** compare the results of two studies.

Action: No change.

12. Figure 1. The results of SP-AMS and CCN counter were not reported in this paper, should be removed from the figure.

Response: We agree with the referee that SP-AMS and CCN counter are not necessary in this paper.

Action: Done.

13. Figure 6 (a-d): in the abstract, the author state that “In fact, most of the fresh soot particles considered in this study were largely spherical (dynamic shape factor: ~ 1.1)”, however, in this figure, the dynamic shape factors of fresh soot with internal voids are around 1.5-1.9, please check the data consistency. And the black dots in the figure should be changed to blue color as other points without internal voids, or in the legend change “fresh soot” to “fresh soot without internal voids” to make it more clear.

Response: We agree with the referee that the values are not consistent. We have checked the data carefully and we found that in the figures the values are wrong due to a mistake in calculation. The values should be in the range of 1.03-1.08.

Action: Figure 6 has been updated with correct values in the revised manuscript. In addition, the values in Table 1 have also been corrected. The legend for the black dots in Figure 6 has also been changed to “fresh soot without internal voids” according to the referee’s suggestion.