

Answers to the review of anonymous Referee #2

We thank Referee #2 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.

The authors present data on measurements on the mass equivalent and mobility size of fresh and coated soot particles, which is then interpreted using a framework that can explain the sequential transformations observed. The theoretical treatment is interesting and useful, and the conclusions are for the most part consistent with the data analysis. The subject is very suitable to ACP, and therefore I would suggest publication of the paper. There are some corrections and clarification that need to be made before publication, and I have also a few suggestions to improve readability.

1. Abstract (and conclusion): In the abstract, it is stated that the dynamic shape factor of fresh soot was in most cases ca. 1.1; this is also stated in the conclusions. This is clearly wrong, as none of the values reported e.g. in Table 1 are even close to 1.1. Please correct.

Response: We agree with the referee, the dynamic shape factor in the abstract and conclusions are not same as values in Table 1 and Figure 6. However, after checked the data and calculation carefully, we found that the values in Table 1 and Figure 6 are wrong due to a mistake in calculation. The statement and conclusion on the morphology of fresh soot particles in this study remain valid.

Action: The sentence in the abstract has been changed to “In fact, the dynamic shape factor adjusted for internal voids was close to 1 for the fresh soot particles considered in this study, indicating the particles were largely spherical.” (See page 2, line 2–4 in the revised manuscript). The sentence in the conclusion has been changed to “In fact, the dynamic shape factor adjusted for internal voids (χ_i) was close to 1 for the fresh soot particles considered in this study, indicating the particles were largely spherical.” (See page 18, line 20–21

in the revised manuscript). The values in Table 1 are corrected and Figure 6 is also updated accordingly.

2. Abstract (p2, 111): I'm not sure that it is correct to say that this is the first study to track microphysical changes in situ, as e.g. observations of changes in soot effective density have been made for a long time.

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).

3. p 4, 134: "(ii) volume equivalent inclusive of internal voids": just to clarify; are the internal voids assumed to be part of the particle volume, but external voids are not? If yes, this could be clarified in the explanation of the framework to make following it easier.

Response: We agree with the referee. The internal voids are assumed to be part of the particle volume.

Action: The sentence “(ii) determining the volume equivalent diameter inclusive of internal voids.” has been changed to “(ii) determining the volume equivalent diameter inclusive of unfilled voids.” (See page 4, line 28 in the revised manuscript).

4. p7, eq. 8; when calculating the mass equivalent density of a particle, which density is used? Does this translate also to the mass equivalent coating thickness? This could be useful to indicate, because a person using the

framework will not know which density (effective, sulfuric acid, SOA, etc...) to use.

Response: We agree with the referee, the explanation of the density should be clear. The material density is used to calculate the mass equivalent coating thickness.

Action: In response to address the issue raised by the referee, one sentence “For fresh soot ρ_m is the material density of the soot, whereas for coated particle ρ_m is the average material density over all the components of the particle, which can be calculated from Eq. (4)–(7).” has been added after Eq. (8). (See page 8, line 1-2 in the revised manuscript).

5. p9, 19; I did not fully understand what the difference between the nominal and actual mobility diameters are. They are selected with the same instrumentation and if nothing is done to the aerosol in between, they should be the same? Please clarify.

Response: The nominal mobility diameter is the setting value of the first DMA, whereas the actually mobility diameter is the value measured with the second DMA. These are the digital values by two identical sets of instruments within the instrumental noise/error. So in reality both mean the same.

6. p9, 117: I’m a little bothered by the use of ‘preferentially’ in the paper. If I understand the text correctly, the open voids are filled first (shown by horizontal lines in figs (4), and then the particles start to grow. Are the internal voids filled at all? To my understanding, the internal voids are assumed to be left open (in the framework at least). This could be stated more clearly.

Response: We agree with the referee that open voids are filled first, but the filling of internal voids and particle growth can happen sequentially.

Action: The word “preferentially” appears 3 times in the original version of the manuscript, and the word “preferential” appears once in the original version of the manuscript.

On page 9, line 17 in the original version, the sentence “(i) in the case of SOA, the open voids in the condensed material are preferentially filled prior to the onset of growth” has been changed to “(i) in the case of SOA, the open voids are filled prior to the onset of growth”. (See page 9, line 21 in the revised manuscript).

On page 12, line 29 in the original version, the word “preferentially” has been removed. (See page 13, line 6 in the revised manuscript).

On page 12, line 30 in the original version, the word “preferential” has been removed in the revised version. (See page 13, line 7 in the revised manuscript).

On page 14, line 15 in the original version, the sentence “..., the filling of internal voids occurs preferentially to growth by high surface tension species, ...” has been changed to “..., the filling of internal voids occurs prior to growth by high surface tension species, ...”. (See page 14, line 27 in the revised manuscript).

7. p11, l 27: "The black, red, and green colors in each pie chart represent the mass fraction of black carbon, sulfuric acid, and organics, respectively". How were these mass fractions obtained? Also, this information should be in the caption.

Response: The mass fractions of black carbon, sulfuric acid and organics are calculated from APM measurements. The method to calculate these mass fractions is described in the section 2.2 Data analysis part, and given by Eq. (4)–(6).

Action: In the caption of Figure 4, one sentence “The black, red, and green colors in each pie chart represent the mass fraction of black carbon, sulfuric acid, and organics calculated from Eq. (4)–(6), respectively” has been added.

8. page 12, l25: Move the part starting with 'We assume...' and ending with 'in this work' to the start of the explanation of the framework, as it will clarify the explanation better than here.

Response: We think that it reads quite well here as well.

Action: No change.

9. page 12, line 33: The phrase step-wise filling is often mentioned. I understood that there are basically two steps: void filling, and subsequent growth. Are there more? The collapse of the structure is also mentioned at some point, but this is not shown in Fig. 2. I would suggest that the actual steps are explicitly marked and named in at least one of Figs (4), preferentially all. Also, they should be explained in more detail in the captions.

Response: there are only two basic steps: void filling and particle growth illustrated in the framework. However, in reality, void filling may lead to collapse of the structure (decrease in mobility diameter). We agree with the referee that actual steps should be marked and named in Figure 4.

Action: To give an example, actual steps including voiding filling, particle growth and collapse are marked by arrows and named in Figure (4d). The explanation of the purple lines "Purple lines parallel to the ideal sphere growth line (dashed black) represent growth of the particle diameter; purple lines parallel to the x -axis represent filling of voids; purple lines with negative slope indicates a combination of void filling and collapse of the soot particle." has been added in the caption.

10. Page 15, line 20-22: . . . "This may introduce some shift in the mapping of the mass and mobility size, leading to overestimation of Δr_{me} ." I don't really understand how the continuous growth causes a shift in the measured mobility

or mass; please clarify this. Also, is there a reason why the soot differs so much in the internal/open void properties between the present and literature studies in Table 3?

Response: In smog chamber experiments, SOA composition is changing continuously because it is constantly evolving from reactions and aging of SOA. In fact, SOA condensed on soot is not the same in any two measurements. So we think that the continuous growth of soot in smog chamber may be different for two consecutive points since it took about 20-30 min to acquire two measurement points. In other words, during the measurement the particles in the environmental chamber were evolving and there are several factors that can cause the differences between this study and literature studies:

1) The coating devices and time scales were different: in this study the coating device was laminar flow reactor and the residence time was 4.8 min while in the literature studies the coating device was a collapsible environmental chamber and the time for each their experiment was several hours.

2) The soot was different: in this study the soot was generated from a premixed-diffusion flame and denuded with a thermos-denuder at 400 °C, while in the literature studies the soot was generated from a Santoro-type laminar diffusion burner without thermo-denuder.

3) The condensed materials were different: in this study the coating materials were two types: sulfuric acid and limonene ozonolysis SOA, while in the literature studies the condensed materials were only SOA: toluene OH oxidation products in Qiu et al. (2012), and m-xylene OH oxidation products in Guo et al. (2016).

11. Conclusions, p. 17, l21: '...this is the first study that considers the effect of coatings with two chemical components'. Is there any conclusions drawn on the effect of the different components, and which properties cause these differences? I could not find these, and as this is not the main purpose of the paper, maybe this sentence could be changed.

Response: We agree with the referee that the statement on the effect of coatings with two chemical components is not clear.

Action: The sentence “To the best of our knowledge, this is the first study that considers the effect of coatings with two chemical components (i.e., sulfuric acid and SOA) on soot morphology.” has been removed. (See page 18, line 9 in the revised manuscript).