**Response to Reviewer 3 (Reviewer comments in black text):**

The authors describe interesting and important findings regarding phase separation in mixed inorganic/organic systems which apply to atmospheric aerosols. The paper combines interesting methods and gives detailed insights into the composition of the forming phases as well as comprehensive discussions of morphology and thermodynamics of the systems. The manuscript is well written and interesting to read. Structure, presentation of the results is excellent. The paper can be published as it is in ACP.

We thank reviewer 3 for helpful comments and suggestions. The detailed responses are given below with their respective locations in the revised manuscript.

I suggest that the authors may consider some minor remarks given below:
General: The notation "C5-C7" seems somewhat unlucky to me. I first understood that you mix gluatric, adipic and pimelic acid. It finally became clear, but I suggest to mention earlier in the paper that you investigated mixtures of isomeric dicarboxylic acids with five, six, and seven C-atoms, respectively. This should be already mentioned in the abstract p.29142, line 5f, something like
...we investigated complex mixtures of structure isomers of atmospherically relevant dicarboxylic acids containing with *5, 6, and 7 carbon* atoms (C5, C6 and C7) having oxygen-to-carbon...
and at the end of the introduction, p. 29146, line 16f, e.g.
For our model systems, we use *mixtures of isomeric* dicarboxylic acids with five, six, or seven carbon atoms (C5, C6, and C7).
We have modified the notation (p. 29142 and 29146).

Paragraph starting on page 29150, line 14: The authors could refer to Table 1 here; it is then easier to keep track of the systems.
We have changed the text accordingly.

Page 29151, line 10 - 22: Here the authors give information, which should have been given already before in the introduction section:
Line 10-13, beginning with *Instead*
and in the experimental section
Line 16-18 and line 20-22 beginning with *In a second*

We have extended the information in the introduction accordingly.

Page 29156, line 1: Can this capillary effect also be seen by comparing a2 and a5 in Fig. 3. It looks like as if a significant CH band grew in the crystallized AS-rich phase.
Yes, this is a good point. We mention this now in the revised manuscript.

Page 29156, line 25: The fact that you might miss something because of low contrast is important information. Important information should not be stated in brackets. It seems to be important as it is discussed later again. You could refer here to the discussions later in the manuscript.
We have removed the brackets.

Page 29159, line 13 and Fig. 6.: The discussion of Fig. 6a is difficult to follow. A reason maybe that a data point mf = 0.3 is not in the Figure, another that you move from high to low mf values. Can you please extent the explanation of Fig6a with one or two sentences.
We have improved the discussion of Fig. 6.

The caption of Fig. 6 should be improved, the legend is refering to *two liquid phases* while you show only spectra from one of these. Why don't you skip the legend in the figure and note the symbols in the caption?
The caption has been improved.

Page 29160, line 16ff and Fig. 7: The Fig 7a, 7c are neither used nor explained in the text. Either skip them or use/desribe/refer to them.
We have changed the order of (a) and (b) in Fig. 7 and mention now (b) and (c) in the text.

Page 29161, line 9ff I find the statement that the model agrees with the trend of decreasing mixing gap with decreasing C somewhat overoptimistic. The model finds that the mixing gap for C6 is smaller than for C7, but fails in case of C5. Actually, acidity is in many instances increasing with decreasing C length. Could neglect of dissociation be the cause why your model fails in the C5 case? It would be a similar argument as used on page 29163, line 19.
We think that this trend is clearly given because the modeled miscibility gap decreases in compositional extent from C7 to C4 dicarboxylic acids and is completely absent for a C3/AS/$H_2O$ system. Small Gibbs energy differences may decide whether a phase separation is present or not, therefore a perfect agreement

cannot be expected for a group-contribution approach. Moreover, the neglect of organic acid dissociation might be an additional reason for observed differences between the modeled and measured miscibility gap.

Page 29168, line 23 Are *literature studies* the same as *studies (reported) in the literature*?
We have corrected to “Based on this and other recent studies,”.