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Editor of Atmospheric Chemistry and Physics

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Author(s): L. Miñambres et al.
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Dear Prof. Petäjä,

Please, find herewith enclosed the final version of the referenced manuscript and the point-by-point replies to the referee's comments, which have addressed in full. These can be found starting next page.

Yours sincerely

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General comments:

The manuscript has improved during the peer-review process. However, there is still some improvement that is needed in terms of scientific quality that needs to be addressed before the manuscript can be accepted. The scientific conclusions from the data are quite strongly presented. The data is not contradicting these conclusions, but a more detailed analysis on the quality of the data should be presented or the statements should be revised / softened.

ANSWER: The main ideas in the "*Conclusions*" section have been softened when appropriate, so that they are compatible within our experimental errors. Alternative explanations have not been excluded. The rewritten sentences appear in the lines 640-654 of the revised version.

Error analysis: 4-9 degrees are assumed to be the error in temperatures (6.5 degrees in the other reply), but none are stated for relative humidity. In particularly in the deliquescence and efflorescence analysis, RH needs to be well quantified to produce comparable data with other methods and studies. The agreement of the data with theoretical value for NaCl DRH gives confidence on the method, but this needs to be mentioned in the text together with associated errors and their effects on the results.

ANSWER: Details about the RH measuring technique together with its error estimate have now been included in the second paragraph of the "*Materials and Methods"* section (lines 211-219). Accordingly, RH values in the text have been changed to the appropriate number of significant figures when needed. Additional explanations regarding this question are given in the answers to the "Technical and minor comments" questions.

Section 3.4.3 discusses various processes affecting deliquescence and efflorescence. Please underline more clearly, how your results add to this knowledge. Please quantify the effects, when possible.

ANSWER: The various processes that affect hygroscopicity now are discussed more clearly, in connection with our results. Experimental evidence has been given when possible, and in some cases the explanations are only tentative. This modified discussion can be found in lines 488-517 lines.

Section 3.5: The beginning could form a short sub-section in the methods section as it helps the reader to understand, how the spectra changes in different RHs.

ANSWER: In the corrected version the beginning of Section 3.5 has been moved to Section 2 (lines 199-206). The text in both sections has been conveniently adapted to conform to the changes.

There seems to be some mistakes with Figure numbering, which needs to be fixed. I list other relevant minor as well as technical comments that should be implemented below.

ANSWER: In fact figure numbering was incorrect in some cases. Particularly Figures 5 and 6 were both named "Figure 5". In the revised version all figures are properly named and recalled in the text.

Technical and minor comments:

NOTE: all style and language corrections have been fully carried out, and are not further commented. Only points requiring explanation are included here.

1. **Line 171**: What is the relevance of these sources (seed oil, cooking, emission from plants) for marine aerosols? What are sources in marine environment?

ANSWER: The relevance of the acids sources for marine aerosols consists of the presence of these acids in atmosphere near marine environment. Once marine aerosols are formed, the surfactant species generated in coastal areas can nucleate heterogeneously onto marine aerosols. This is now stressed in lines 174-175 of the revised manuscript.

2. **Line 207**: How well is RH controlled? How stable the system is? How is RH measured?

ANSWER: Relative Humidity is measured with a digital thermohygrometer (Vaisala Humicap HMT 337). The error of its measurments is 1% from 0 to 90% of relative humidity and 1.7% for the rest interval. The thermohygrometer sensor is placed at the exit of the flow cell, as seen in Figure 1.

3. **Line 310**: The last sentence in the paragraph needs a reference or a stronger support from the data.

ANSWER: This suggestion is based on infrared spectra of fatty acids presenting small amounts of water, that show a broad band near 3000 cm^{-1}. Of course there may be alternative reasons for the presence of the broad band of which we are unaware, so we have modified the sentence accordingly (lines 330-331). We also give a reference for this work in the revised version (line 331).

4. **Line 358**: Is the same number of particles verified with the CPC?

ANSWER: The Condensation Particle Counter measures the particle number in a flow. On the other hand, to obtain SEM images we deposited the particles in the aerosol flow onto a glass slide placed at the exit of the aerosol cell. The exposure time (several minutes) was adjusted by trial and error to obtain an enough number of particles in the

SEM image. As a consequence, the particle number density obtained by both methods cannot be compared.

5. **Line 360:** How do you verify thicker coating for the smaller particles?

ANSWER: When analyzing SEM images of the smaller particles at sufficient magnification, particles can be seen completely immersed in a surfactant drop. For bigger particles this effect was not observed. This is now explained in the revised version (lines 381-383).

6. **Line 389:** This paragraph belongs to the beginning of the next section, 3.4.2.

ANSWER: This paragraph describes the infrared spectra of particles in efflorescence mode and follows the description of deliquescence spectra in 3.4.1 section. So we think that the paragraph is properly placed. Next section (3.4.2) deals with deliquescence and efflorescence curves that have been obtained from infrared spectra.

7. **Line 408**: define “very similar”.

ANSWER: This point has been explained more clearly in lines 431-433 of the revised manuscript. Briefly, the shape of the deliquescence curves of NaCl and NaCl/HA is the same. In addition, the water uptake of both types of particles is the same. The main difference between them is the number of points measured to build both curves.

8. **Line 416**: Quantify the term “slight”.

ANSWER: The term has been quantified from the deliquescence curves in the reference. Now it reads "*These results are in agreement with previous reports in which a DRH of 70% was observed for NaCl particles covered with OA and LA acids (Hansson et al 1998).*" (Lines 438-440).

9. **Line 418**: New paragraph for the efflorescence discussion. Please provide references to other works that have studied efflorescence of NaCl. Are your results in agreement with them?

ANSWER: The new paragraph is now placed in lines 442-448.

The following references have been included:

- Weis, D.D.; Ewing, G.E. J., 1999. Water content and morphology of sodium chloride aerosol particles. Geophys. Res. 104, 21275-21285.

- Czizo, D.J.; Abbatt, J.P.D. 2000. Infrared observations of the response of NaCl, MgCl2, NH4HSO4, and NH4NO3 aerosols to changes in relative humidity from 298 to 238 K J. Phys. Chem. A 104, 2038-2047.

Now our value is compared with the value given in the references (lines 443-444).

10. **Line 427**: What is your error estimate for the stated RH?

ANSWER: As the RH error estimate is 1% for this value (as explained now in Section 2), the figure has been rounded to read 50% (line 451-452). RH values have been corrected in this respect throughout the text and in the figures.

11. **Line 432:** Grow faster? Based on your replies, I understand that your results are in equilibrium.

ANSWER: The expression is inadequate. The sentence has been rewritten and it appears in lines 456-457.

12. **Line 459**: … admits water condensation at any RH. (What is the lowest RH? Does this hold for 0 % RH?

ANSWER: Liquid water is not detected in infrared spectra collected at RH=0%. The lowest RH measured for which liquid water is detected is 6%. (changed in lines 483-484).

13. **Line 469**: Is there a better reference for the polarizabilities than a web-page?

ANSWER: Now a proper reference has been given for the polarizabilities (line 496).

14. **Line 489:** in some cases? The results indicate a large effect, please quantify this.

ANSWER: Now the variations in FWHM are explained more clearly. The rewritten text appears in lines 521-526.

15. **Line 495**: … a quite small change in DRH … Please quantify quite small.

ANSWER: The sentence has been modified to address results more quantitatively. Now it appears in lines 532-534.

16. **Line 496: …** are considerable indicating strong … Remove parenthesis and please quantify considerable.

ANSWER: The sentence has been modified to address results more quantitatively. Now it appears in lines 534-536.

17. **Line 509:** Please clarify the connection to this work.

ANSWER: The known salting out effect of NaCl will help organic molecules to move to particle surface and thus facilitate evaporation. On the other hand, the salting out effect for HA has been reported to be small, so this effect may not influence the HA behavior largely. The text has been modified to address this point (lines 545-550).

18. **Line 511:** Can it be seen in your data?

ANSWER: The effect can be seen in Figure 6. Now the text gives this information (lines 551-556).

19. **Line 520**: … must be: so there cannot be any other explanation?

ANSWER: The sentence has been changed, as we offer only one possible explanation (lines 560-562).

20. **Line 528:** Does there have to be a physical hole? Why cannot water just diffuse through the surfactant (mass transfer effect). Please add references to other similar work to support your statements.

ANSWER: The explanation was more pictorial than physical. The sentence has been rewritten (lines 568-569).

21. **Line 567:** Could there be some evaporation in SEM? How rapidly is gold added to the sample?

ANSWER: This question has been addressed in the revised version (lines 601-603): "Although in principle it is possible that part of the surfactant evaporates before particles are covered with gold, LA has a low vapor pressure (2.2×10^{-5} mbar, see Table 2), so we do not expect acid evaporation to alter the sample appreciably."

22. **Line 584:** … thickness is much larger than … What is the estimated monolayer thickness? Please specify.

ANSWER: This information has been included in lines 619-622.

23. I suggest moving the surfactant thickness related conclusions (from lines 601-605) towards the end of this section

ANSWER: These conclusions have been moved and are located in lines 668-672.

24. **Line 611**: Growth factor?

ANSWER: In the revised text this sentence does no longer appear.

References:

Please correct capitalization, journal names, consistent journal acronyms, typos etc. in many of the references.

ANSWER: The references have been corrected.

Figure captions

ANSWER: The suggested changes have been carried out.

In particular, in Figures 5 and 6 the significant figures of the RH values have been corrected.

In Figure 8, the option of a two dimensional plot was our initial idea, but the points for the different experiments overlapped largely, so finally we opted for the 3D version.