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Interactive comment on “Hygroscopicity of aerosol and its organic component at a coastal location” by J. D. Yakobi-Hancock et al.

Anonymous Referee #1

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It is of importance to study aerosol chemical composition and aerosol cloud interactions in marine environments, so new studies in this field are very welcome. In the study by Yakobi-Hancock et al. a number of measurements are carried out in a coastal environment in order to characterize aerosol particle properties. The focus is on the results of the CCN activity of the ambient aerosol particles and the role of the organic fraction of the aerosol particles with respect to the observed CCN activity. Other interesting data series are also presented but often the interpretation is not at a satisfying level. Particle size distributions were measured but for unknown reasons they are not presented or described. Often basic information about measurement techniques is missing, the error analysis is questionable, the article is not well structured, and a lot of speculation that does not seem to be based on the available data or results in the liter-

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ature is included. Furthermore, conclusions are based on a method to infer the organic fraction of the particulate matter by combining SMPS data and impactor data in certain size ranges. The method and the assumptions it is based on are highly questionable, and the method has not been proven by the authors to be reliable.

It is thus recommended that the article undergoes major revisions before publication can be considered.

General/Major comments

1. It is of importance to assess to what extent the aerosol can be considered marine or influenced by anthropogenic or continental emissions during different time periods. Otherwise it is questionable what the presented results can be used for. It should be possible from the available data to come up with a better classification of the aerosol to replace the included speculation which is useless in this context.

2. Size distributions were measured with an SMPS, but the size distributions are not presented nor is anything stated about particle number concentrations. Only the integrated particle volume is presented. The size distribution and number concentrations are very relevant for studies related to ambient CCN and the reported CCN activities – so they should be included in the paper.

3. A lot of basic information about the experimental set up, the measurements and the methods is lacking. From the limited information presented it is not possible to reproduce a similar study in the same way. This issue is particularly pronounced with respect to the measurement conditions for the MOUDI since the results are likely to be very sensitive to changes in RH. Furthermore, nothing is stated about how the data analysis was carried out with respect to the ACSM. The specific comments related to these issues should be addressed when brought up below.

4. The authors have decided to base several of their conclusions on results obtained on the basis of the assumption that there must be closure between the PM sampled

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with a MOUDI and size distributions measured with an SMPS. It is highly questionable whether it makes sense to expect such closure between the aerosol volume measured with an SMPS and the MOUDI under the given circumstances. Even under controlled lab conditions – with a well characterized aerosol - that is not an easy task – and it would require thorough calibrations that do not seem to have been carried out in the present study. In the present study the two instruments do not even share the same inlet. Furthermore, the results are most likely very sensitive to the diameter conversion – which in turn depends on a lot of assumptions. Some of the choices made in the calculations are not justified and errors and uncertainties are not estimated or even mentioned in this relation. The authors want to leave the impression that their results using this method are quite robust. In order to justify that a proper sensitivity analysis should be carried out.

5. The obtained results with respect to the organic fraction and κ_{org} are presented without any critical sense – both in light of the comment above (4) related to the method – and the suspiciously high κ_{org} presented. A κ_{org} of 0.5 is highly unlikely – but that does not seem to be clear from the discussion/conclusion. It would be appreciated if the authors could justify those findings instead of referencing a paper where the highest κ reported for a single organic compound is about 0.4. Furthermore, a significant fraction of the ambient κ values are reported to be 0.3 or lower – how can that be explained if the κ_{org} is in the range 0.3-0.5?

Specific comments

Structure: Order of figures: it would be easier for the reader if references were made to the figures so that the order of the figures correspond to when they are discussed in the text.

Title: ‘Hygroscopicity’ traditionally refers to measurements of hygroscopic growth. Replacing ‘Hygroscopicity’ by ‘CCN activity’ would be more appropriate. ‘Organic component’ could be replaced by ‘aerosol chemical composition’.

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Abstract: The results and statements related to the combination of MOUDI and SMPS data should undergo a more thorough sensitivity analysis before the findings are included in the abstract.

P 12526, L. 5-11: The main findings of the studies Albrecht (1989) and Twomey (1977b) are that an increase in CCN is likely to influence cloud properties and thus climate. That is not clear from the way these studies are cited.

P 12527, L. 9: “breaking waves” do generally not generate all the primary sea spray aerosol particles.

P 12528, L. 17-20: should be rephrased.

P 12529: what was the altitude of the inlet above the sea surface? Are the sampled air masses influenced by the surf zone – depending on wind direction? Is the tide pronounced? Are there any local aerosol sources nearby which could affect the measurements?

P 12529, L 11: ‘directly calculate the kappa’ - is usually not possible from CCN spectra alone. Which assumptions were made e.g. with respect to temperature and thus surface tension?

P 12529, L. 12-14: How ‘broad’ were the selected particle distributions, and what were the typical number concentrations?

P 12529, L. 14: ‘monodisperse’: How were multiply charged particles avoided?

P 12529 L. 12+23: Which RH was the aerosol dried to?

P 12529, L. 17: (0.07-1.2) -> (0.07-1.2 %)

P 12529, L. 17: It is recommended that details about how the supersaturation of the CCNC was calibrated should be included (Rose et al., 2008)

P 12529, L. 23-25: Was an impactor used in front of the SMPS?

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P 12530, L. 2: What does 'wet' refer to here?

P 12530, L. 11-12: if mobility diameters are reported here – then a comment about where to find the information of how they are derived should be given.

P 12530: It is misleading to report this collection efficiency – as pointed out by Ng et al.(2011) it depends on the aerosol being sampled. It should be stated which assumptions the ACSM data analysis is based on – and why.

P 12529-12530: Were the instruments kept in air conditioned surroundings at constant temperature or did the temperature fluctuate?

P 12531, L. 3: 'three types of airmasses' is this statement reasonable here? Can the type of an airmass alone be based on the backtrajectory? Showing just three backtrajectories is not very informative. It is recommended that more are included in the figure. The airmasses seem to be classified according to the backtrajectories, but no information is provided about the classification scheme.

P 12531, L. 14-17: why this discussion about 'marine' when the ratio clearly depends highly on many parameters that cannot be quantified here – as pointed out at the end of the paragraph?

P 12531, L. 21-12532, L. 7: The question here is: can these measurements in combination with wind direction and other time series be used to classify which airmasses that are highly influenced, moderately influenced or not influenced by anthropogenic emissions? This would be useful for the interpretation of the aerosol characterization.

P 12532, L. 2-7: where are the mentioned harbor, local industry and forests located relative to the measurement station? Is there any signature in the data series from these sources?

P 12532, L. 8-11: Is sea salt or sea salt sulfate detected by the ACSM? According to Ng et al.(2011) only non-refractory species are detected (at least in a quantifiable way). Please clarify how nss-sulfate is obtained or estimated.

P 12532, L. 13: Why is only total volume from the SMPS data shown? What were the total number concentrations? Which particle size modes dominated? It would be useful for the interpretation of the results to show number size distributions.

P 12532, L. 16-19+Fig.3: Is the time in Fig. 3 (and all other figures with time series) given in local time?

P 12533, L. 6: “where the solution is assumed to be water” – at which temperature?

P 12533, L. 8: ‘particle size (10%)’ -> does the 10% represent the standard deviation of the size selected aerosol? Or is it related to possible systematic errors?

P 12533, L. 9: It is not reasonable to calculate the error in Kappa based on the error in diameter and supersaturation alone. The error in kappa depends on the error in the critical super saturation, which also depends on the quality and resolution of the activated fraction vs SS curves. Such a figure could be shown in Supplementary material. Nothing is stated about how multiply charged particles are accounted for.

P 12533, L. 19: ‘two time periods’ -> it is not clear which two time periods are being referred to...is it 18-21 Aug and then the time periods before and after combined? Please clarify.

P 12533, L. 23-24: Again it is not clear what ‘first period’ and ‘second period’ refers to.

P 12533, L. 24-12534, L. 6: It is not clear how kappa values higher than 0.7 can be explained by biological activity/photochemistry. The discussion relating cloud coverage to CCN activity is speculative – support the discussion on findings in the data/literature or leave it out.

P 12534, L. 7-13+Fig.7: Be careful here –PM1 composition is being directly related to measurements carried out around 50 nm and 100 nm.

P 12534, L. 9: ‘Increasing sulfate’ – in Fig. 7 only a ratio is included – which does reflect the change in sulfate alone.

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P 12534, L. 9-13: 'dependency of the CCN activity' – of course the CCN activity depends on the chemical composition – please clarify what is meant here.

P 12535, L. 8-13+Fig.8: if the authors wish to include such a figure in the paper – they such provide more information in this section about the method and assumptions/errors involved. It cannot be justified to present fig. 8 as it is without a more thorough analysis.

P 12535, L. 21-24: This very general statement is not true – and it is not what is shown in the cited studies. O'Dowd et al.(2004) clearly show that the marine aerosol composition is highly dominated by sea salt – only during periods with high biological activity the particulate matter below roughly 500 nm is dominated by sulfate and organic species.

P 12536, L.1-9: If the aerosol is dominated by anthropogenic or continental sources – then a correlation between wind speed and κ_{ambient} cannot be expected. Also if the organics are primary marine organics and supposedly high in concentration mixed with sea salt – then it is not clear that a correlation should be observed between wind speed and κ_{ambient} . It seems more reasonable to investigate a relationship between particle number concentrations and wind speed to the power of ~ 3 (with winds approaching from the ocean and no expected influence from anthropogenic emissions) – if a signature of primary marine aerosol is to be investigated.

P 12536, L.12-13: PM1 usually does not cover the 'total aerosol loading'.

P 12536, L. 16-18: This is NOT a proper way to estimate the error on κ_{org} ! Either meaningful error propagation calculations should be carried out – or in this case a reasonable sensitivity study is probably more adequate.

P 12537, L. 20-23: It is claimed that it is likely to find a mixture of ambient organic compounds with a κ around 0.5 with a reference to Petters and Kreidenweis (2007). The highest reported κ for organic species reported in that study is about 0.4 – so it is not fair to base that statement on that reference. The authors should do a better

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job in justifying a kappa_org of 0.5 – if they really want to claim that is a result of their study. A substantial fraction of the kappa_ambient datapoints is below 0.3 – how is that explained if kappa_org is above 0.3?

P 12538, L. 4-8: rephrase

P 12538, L. 16-19: It is not clear what ‘in the past’ refers to here. It was e.g. suggested by Charlson et al.(1987) that secondary aerosol could play an important role as CCN in marine environments.

Fig.1: It would be useful to show the variance in backtrajectories and indicate the criteria for classification into North, West and South as shown e.g. in Fig S1.

Fig.3: It is hard to tell what the volume concentration is from the upper panel. Do the spikes reflect noise that has not been removed – or is it real? It would be very useful to show the number concentration and number size distributions also/instead. It may be useful to indicate in the lower panel when the wind direction was classified as ‘west’.

Fig.4: It seems like there is a trend in the SO₂ concentration – is that an instrument artifact?

Fig.5: It is hard to read this figure. Please use a different scale for the different species – or use a logscale.

Fig. 6: Caption: “dry mobility diameters of 50 nm (red circles) and 100 nm (blue squares)”. In the figure also red squares and blue circles are presented! With 1.5 to 2 h per scan there should ideally be 12 to 16 datapoints per 24 h. Are datapoints missing due to problems interpreting the CCN spectra or only due to instruments failing?

Fig.7: It is questionable whether this figure contains useful information – see comments above.

Fig.8: It is highly questionable whether this figure contains meaningful results – with respect to the organic fraction.

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Supplement:

Page 1: 1) More information about the measured RH please. The last sentence of this 1) paragraph is very problematic: I: it is simply not true that this sensitivity test validates the 'aforementioned assumption' – particularly not when it is being claimed elsewhere that about 80% of the mass is highly hygroscopic organics. II: a doubling of water present - which change in RH does that correspond to? III: If the size range of the MOUDI corresponds to a d_m , which is on average just 10 nm smaller than included in the calculations – then this will most likely have a huge influence on the calculated organic fraction presented in Fig. 8 – and thus the inferred κ_{org} .

Page 2: A lot of highly uncertain assumptions are made – without much/any justification and the authors want to give the impression that the results are very robust. Please provide proper references for the choices made and report/estimate errors. Please perform a proper sensitivity test based on reasonable error estimates: e.g. how does a 10-20 nm lower d_m (or whatever is reasonable) affect the results?

References

Charlson, R. J., Lovelock, J. E., Andreae, M. O. and Warren, S. G. (1987). "Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate". *Nature* 326 (6114): 655–661.

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, *Atmos. Chem. Phys.*, 8, 1153-1179, 2008.

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