

Interactive comment on “Aerosol hygroscopicity and its link to chemical composition in coastal atmosphere of Mace Head: marine and continental air masses” by Wei Xu et al.

Anonymous Referee #2

Received and published: 22 January 2020

Xu et al. report stationary measurements from Mace Head during the winter of 2009. They used HTDMA technique to measure the aerosol hygroscopicity and then compared measurements with hygroscopicity calculations using the aerosol chemical composition (using an HR-ToF-AMS) for size-selected particles. Overall, they found good closure between measured and calculated hygroscopicity assuming complete internal mixture for all components. The manuscript is well written, and the conclusions are justified. I recommend publication after the authors address the following comments.

General comments:

The authors compare measured and calculated hygroscopicity for size-selected parti-

C1

cles. However, the chemical composition measured with an AMS for particles smaller than 50 nm cannot be trusted due to significant inlet losses. I suggest that either the authors add adequate justification that the AMS they are using can accurately quantify the mass composition for sub-50nm particles or entirely remove the comparison between calculated and measured hygroscopicity for the sub-50 nm particles.

Specific comments:

Line 100: The authors use the composition-dependent CE (CDCE) to correct AMS concentrations. However, the CDCE method does not take into account sea-salt and organic particles which are ubiquitous in the marine environment. Therefore, I expect that the authors provide a rationale for using this CE correction method. The authors should estimate the CE using a mass closure approach (using DMA volumes and densities) to get another estimate of CE. If the two agree, then this should provide the rationale needed. If not, further discussion is needed.

Line 120: If both sea-salt and sulfuric acid have GF than are larger than 1.85, why call it sea-salt mode? This is confusing. Maybe use highly hygroscopic.

Line 131: Please provide a reference for the choice of density for the organic compounds that is relevant to the marine environment.

Line 160: I do not follow the rationale of only including measurements when BC concentrations were below 15 ng/m³ and then claim that these represent pristinely clean conditions. Surely if continental air masses spent several days above the oceans (while being diluted with cleaner air from the free troposphere) one would expect low BC levels (<15 ng/m³) however, the origin of the particles would still be transported pollution from the continents. Another potential source of pollution could be diluted ship exhaust. I suggest the authors include in the SI a scatter plot showing the non-refractory organic and sulfate concentrations measured by the AMS versus MAAP BC concentrations. If the two are not correlated than this would at least eliminate the influence of combustion aerosols.

C2

Lines 202-203: Can the authors expand as to why does the boundary layer height should affect an aerosol intensive property like hygroscopicity?

Lines 207-208: I am not sure I follow the argument that the authors are trying to make. The authors are claiming that because of the relatively low MSA concentrations measured during continental periods therefore there is low impact from marine sources. However, MSA concentrations during marine periods were clearly lower than those measured during continental periods (Table S2). Also, include details about how MSA concentrations were measured.

Lines 221-223: How do the authors confidently attribute the highly hygroscopic particles observed at 35 nm to sea-salt and not to sulfuric acid? Without providing evidence that these are indeed sea-salt particles, I suggest that the statement be removed.

Line 230: A recent article by Quinn et al. (2019) (<https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019JD031740>) reported a persistent organic and non-volatile (at 230oC) ultrafine particle mode that was likely entrained from the free troposphere, from measurements in the North Atlantic.

Lines 240-243: I am not sure what is the point that the authors are trying to make by pointing out the different size dependence of the GF during marine and continental periods. Expand or remove.

Line 246: "HTDMA" and not "HTMDA".

Line 224-259: Poorly written and confusing. Please re-write.

Line 263: I am not sure what Fig 7a and 7b refer to. Figure 7 is a six-panel figure and the left and right columns refer to continental and marine periods respectively. Adjust.

Line 264: Adjust the text to: "R2 values were 0.47 and 0.18 for 75nm particles during continental and marine events".

Line 269: perhaps "dynamic range" is better suited than "variability" in this context.

C3

Line 282: I do not agree with the statement that the AMS is an excellent instrument to measure sea-salt. Did the authors collect filters for IC analysis to retrieve sodium and chloride concentrations and then compare these measurements with AMS measurements of sea-salt for this particular study? Or are the authors simply relying on an old calibration from a 2012 study? Other studies in the literature have failed to get good closure between AMS salt measurements and those from IC filters. Can the authors also provide scatter plots of AMS salt concentrations versus wind speed? How do these compare?

Figures S3 and S4 in the SI are mislabeled. Please adjust.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-839>, 2019.

C4