*Reviewer 2*

*This manuscript presents the CCN closure study over the Northeast Atlantic Ocean. By assuming different mixing states and size-resolved κ values, the measured and estimated NCCN were compared. I do think the results are quite interesting and important for the model work, as the author points out in the conclusion. However, I strongly recommend the author be more careful about the data evaluation, analysis, and interpretation. The following major comments must be satisfactorily addressed before consideration for publication.*

**Response:** We thank the reviewer for the constructive comments.

*Major comments:*

*1. In Section 2.2.1, SMPS measured particle number size distribution only covers the particle size up to 500 nm. Once you used the integrated particle number concentration from SMPS-measured size distribution, particle number concentration in the size range larger than 500 nm (N>500nm) are not considered. N>500nm are CCN at supersaturations >0.20%. From Fig. 2, even the particle larger than 500 nm is not measured, it is clear that N>500nm cannot be neglected. This is also related to the interpretation of slopes in Fig. 4 & 5. One solution is to fit the larger Accumulation mode. Based on such a method, the N>500nm can be estimated.*

**Response:** We acknowledge that the particles larger than 500 nm certainly contributed to CCN. However, their number concentrations were typically very low in the remote atmosphere and normally within the measurement uncertainty of the total number. As shown in the Figure R1 below, the largest particle number concentration with sizes over 500 nm was observed in clean sector with the median N500 of 3.7 particles per $cm^3$. We can then compare with the number of particles above the characteristic Hoppel minimum size - 90 nm in the clean sector - which is arising from the activated particles in the clouds over Mace Head and obtain 145 particles per $cm^3$. Therefore, the fraction of N500 was < 3% of all activated particles, which was within the measurement uncertainty of CCN counter. The errors by ignoring particles over 500 nm were even smaller in mixed and polluted sectors.

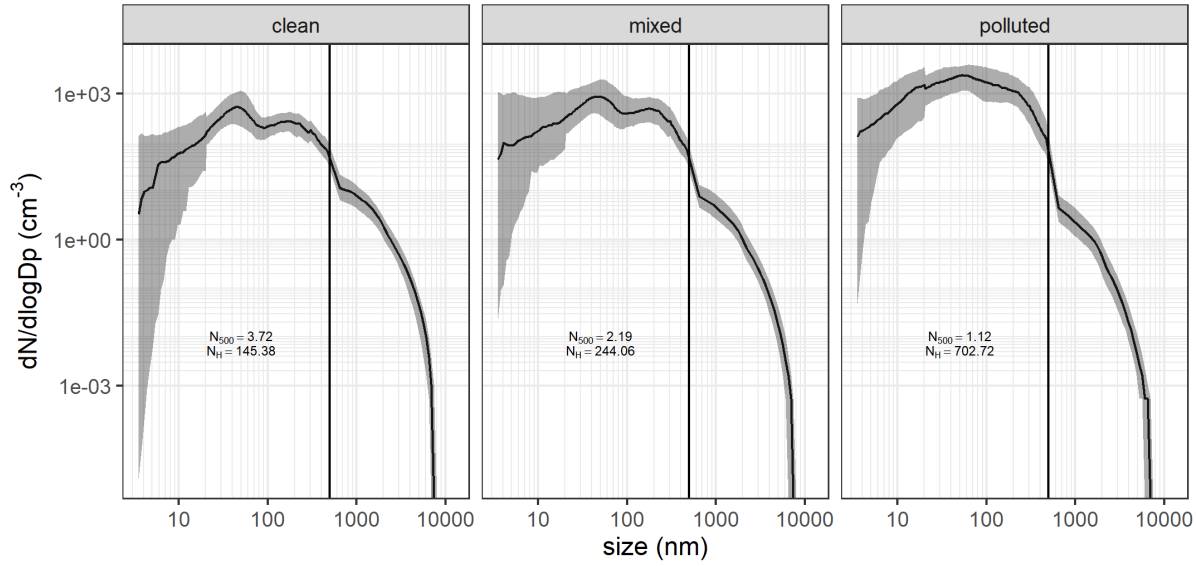

Figure R1. The aerosol number size distribution combining scanning mobility particle sizer (SMPS, 0.003 to 0.5 um) and aerosol dynamics sizer (APS, 0.5 to 10 um) for each sector. The lines represent the median values, the shaded area represent 25$^{th}$ to 75$^{th}$ quantile. The vertical lines represent SMPS sampling upper size limit of 0.5 um. Also shown are the number of particles above the Hoppel minimum ($N_H$) and over 500nm ($N_{500}$).

We have added a sentence for clarification in the revised manuscript:

"To note that the upper limit of 500 nm was used when integrating $N_{CCN}$, because the SMPS measured particles up to 500 nm. Particles larger than 500 nm certainly contributed to the $N_{CCN}$ because of their large size. While ignoring those large particles would cause a slight systematic underestimation of $N_{CCN}$, such underestimation would be negligible as their contribution to the total activated particle number (< 3%) was within the measurement uncertainty of the CCN counter."

*2. In Section 2.3, the observation was categorized into Clean-H, Clean-L, Polluted-H, Polluted-L and mixture in between. Why did you use the wind direction, rather than be backward trajectory? It is mentioned the pollution is long-range transport anthropogenic pollution from Europe. Therefore, backward trajectories should work better than the wind direction. Besides, after classification, it is better to show the wind rose plot; boxplot or frequency distribution of BC, N30 and/or meteorology data during Clean-H, Clean-L, Polluted-H, Polluted-L, Mix-H and Mix-L in the supplement. Reviewers and readers will have a better understanding of the classifications. It also helps your interpretation afterward. The "H" and "L" are classified by the biological activity seasons; therefore, it is needed to show the difference of biological activity during "H" and "L". For example, you can show the Chlorophyll-a and DMS concentration over different seasons. There are free and easy access data from NASA. Why WS>3m s-1 is one of the criteria of the clean sector? When BC<15 ng m-3, WD from 190 to 300 and WS<3 m s-1, will it be classified as which sector?*

**Response:** The clean sector criteria have been established over many years of rigorous investigation at Mace Head and we would like to continue to keep those criteria for consistency (O'Dowd et al., 2014). It is always beneficial to use BC, WD and WS operationally in data analysis as the trajectories are not practical to be retrieved at minute resolution and using BC and WD screens out all transient pollution events (like ship plumes or recirculated air masses due to sea breezes). We added frequency plots of backward trajectories, wind rose plots, and boxplot of black carbon and meteorological parameters and sea surface chlorophyll concentration plot in the Supplementary Material of the revised manuscript as suggested:

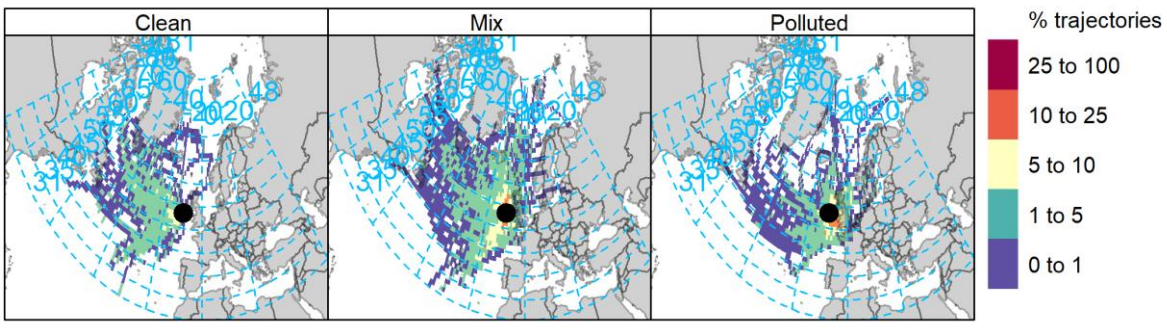

**Figure S2. The frequency distribution of 96 hours HYSPLIT backward trajectories ending at 100 meter height at Mace Head for each principal sector with colour codes representing frequency of the trajectories passing through the specific location.**

"As shown in Figure S2, most of the clean sector air mass advected over the North Atlantic without land contact for at least three days. The polluted sector showed a large anthropogenic impact with the highest frequency pixels located over land, but also revealed re-circulated air masses because of geographical location of MHD."

We also added wind rose plot of each sector, boxplot of BC concentration, number of particle larger than 30 nm (N30) and meteorological parameters as shown in the revised Figure S3.

"The wind rose, BC distribution and meteorological parameters of each category is given in Fig S3."

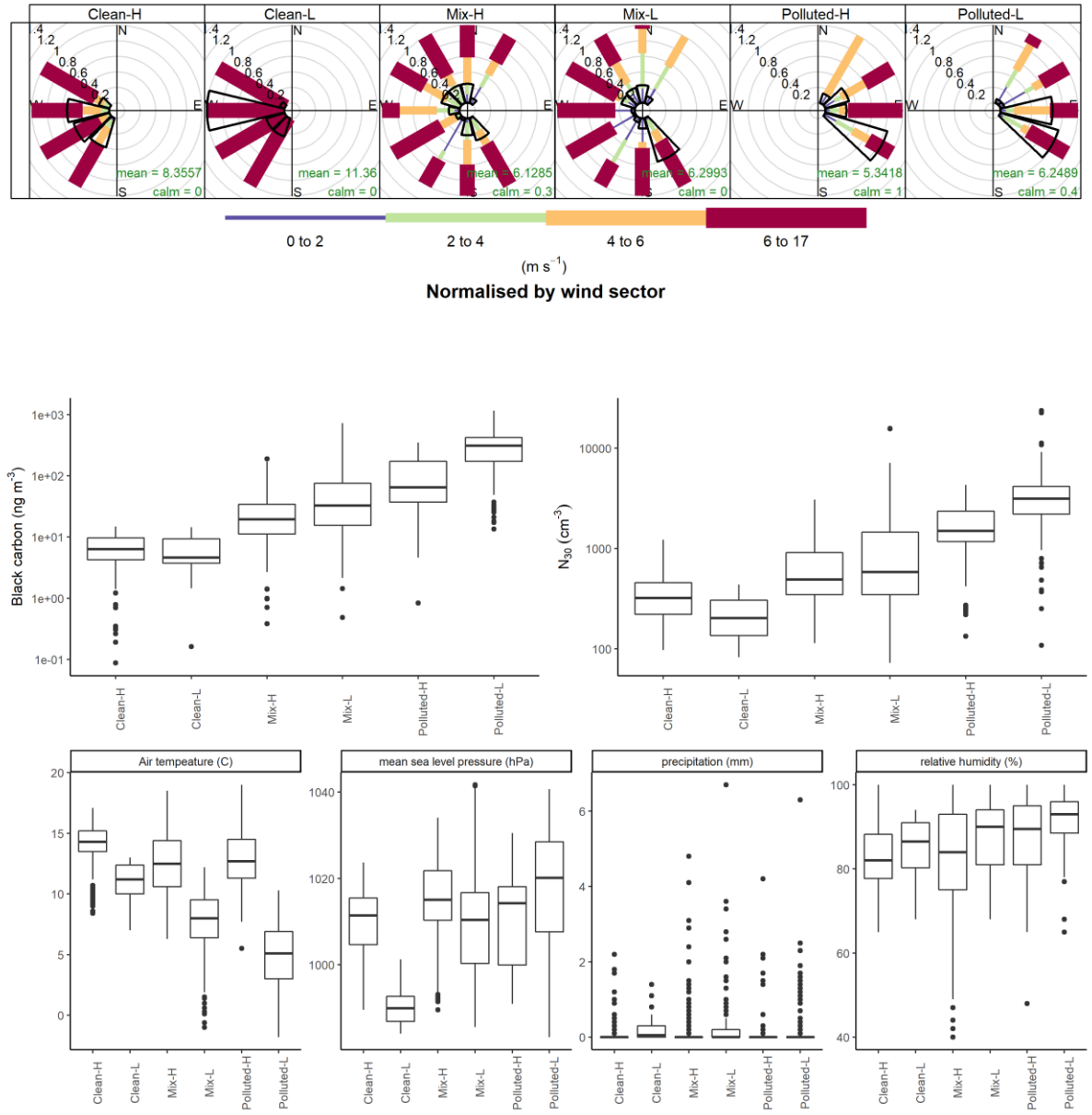


**Figure S3. The normalised wind rose plot (top) for each air mass with colour codes representing the frequency of occurrence; the boxplots represent black carbon, number of particles larger than 30 nm (N30) (middle plots), air temperature, mean sea level pressure, precipitation, and relative humidity in each air mass (bottom plots). The horizontal lines represent median value, boxes represent 25$^{th}$ to 75$^{th}$ quantile, whiskers represent 1.5 inter-quarter range and markers represent outliers.**

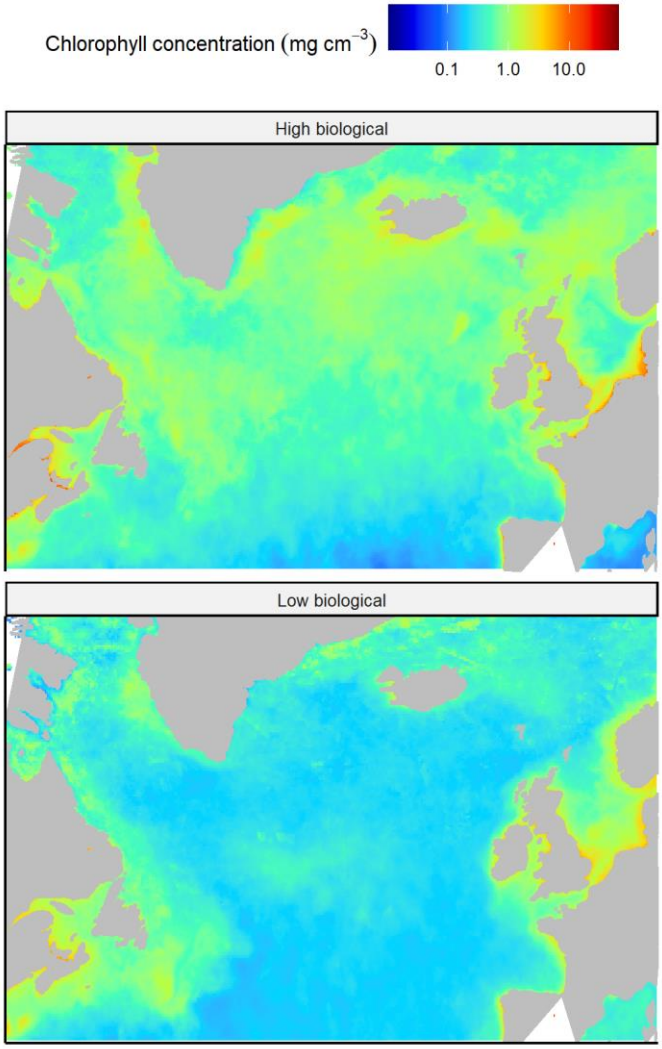


**Figure S1. Sea surface Chlorophyll concentration during high biological activity period (top) and low biological activity period (bottom) for the year 2009 to 2010 in the North Atlantic Ocean. The monthly sea surface chlorophyll-a concentration data were downloaded from the EU Copernicus Marine Environment Monitoring Service (http://marine.copernicus.eu/) based on a multisensory approach.**

Line 175: "The clean, polluted and mixed sectors were further divided by the level of oceanic biological activity further noted as "H" or "L" for high and low biological activity seasons, respectively. The duration of high biological activity periods classified according to prominent phytoplankton blooming periods (O'Dowd et al., 2004; Yoon et al., 2007). The sea surface chlorophyll concentration varied significantly in H and L season for the North Atlantic (Fig. S1).

*Why WS>3m s-1 is one of the criteria of the clean sector?*

**Response:** The wind speed > 3m/s was used (1) to remove stagnant air masses with corresponding sea breezes

that could potentially become polluted and (2) to minimize costal effects.

*When BC<15 ng m-3, WD from 190 to 300 and WS<3 m s-1, will it be classified as which sector?*

**Response:** will be classified into Mixed sector.

Table 1 was modified for clarification.

*3. Some of the criteria and numbers are very arbitrary, lack evidence to support.*
*For example, in Lines 194-201: First, as far as I know, the CPC model 3010 TSI detection limit is 10 nm particles. Second, SMPS upper limit is 500 nm, whereas CPC depends on your inlet cut size. To make it clear, as CPC covered a broad size, N10/Ncpc should be smaller than 1. In a normal distribution, $\mu \pm \sigma$ covers 68% and $\mu \pm 2\sigma$ covers 95%. I could not under why 84.15th quantile. Why "extra 10% of uncertainty was allowed due to typical uncertainty of particle counting"? Here you mean the measurement uncertainty of SMPS?*

**Response:** We thank the reviewer for pointing this out. We actually used the ratio of Ncpc/N10 rather than N10/Ncpc and the text was now corrected. The number of particles in the range from 10 nm to 500 nm was in fact measured by two SMPS systems: one from 3nm to 20nm (nano SMPS) and the other from 20 nm to 500nm. The method section has been updated to reflect that. The 84.15$^{th}$ quantile represents a normal distribution where 84.15% of the data is smaller than $\mu + \sigma$. Therefore, we limited the 84.15$^{th}$ quantile to 1.1 to take into account CPC "over-counting" during coastal nucleation events.

We have modified the text in the revised manuscript for clarification:

"A nano-SMPS was used to measure aerosol number size distribution from 3.5 to 20 nm."

"The SMPS data were cross-checked by an independent condensation particle counter measurement (CPC, CPC3010, TSI). The ratio between $N_{cpc}$ (the total number measured by CPC) and $N_{10}$ (the total number of particle larger than 10 nm measured by SMPS) and were calculated hourly. The upper envelope of the ratio was expected to be ranging between 1 to 1.1 to account for the rather frequent new particle formation events at MHD. The SMPS data were corrected on a daily basis to make sure the 84.15th quantile of $N_{cpc}/N_{10}$ were ranging from 1 to 1.1. The 84.15$^{th}$ quantile was used as it represents the upper limit of mean value plus 1 standard deviation in the normal distribution. ~~Considering the normal distribution of the log($N_{cpc}/N_{10}$), the value of 84.15$^{th}$ quantile of log($N_{cpc}/N_{10}$) represented the upper limit of 1 standard deviation over the mean value. The extra 10% of the uncertainty was allowed due to the relative board size range of CPC.~~"

*Lines 203-207: Why are 1.1*N30 and 1.2*N30 used as the limit?*

**Response:** The N30 number was chosen to represent the lowest size of the most soluble species (sea salt) activated at high supersaturation for comparison and cross-validation with CCN. The different coefficients (1.1 and 1.2) were due to SMPS counting statistics following Poisson distribution (Buonanno et al., 2009), meaning the uncertainty equals to the square root of the particle count resulting in different coefficients for low and high particle number events.

*Line 216: Why "uncertainty of the AMS based on κ to be lower than 20%"? How do you get 20%?*

**Response:** The main uncertainty of AMS was determined by collection efficiency, which is approximately 30-40% (Bahreini, R. et al). However, for determining the relative contribution of chemical species, which matters most for kappa calculation, the uncertainty is smaller. Moreover, it was demonstrated that the kappa values from AMS were usually within 20% from HTDMA derived GF (Ovadnevaite et al., 2017; Xu et al., 2020). Therefore, we considered the uncertainty of chemical composition derived kappa based on AMS was within 20%.

*4. Some of the statements and interpretations are too strong without supporting evidence. For example, in Line 311, Do you think wet removal is one of the "cloud processing" or not? I am guessing what you are trying to say is that higher concentration of accumulation might be due to strong condensation growth and/or free troposphere entrainment during wintertime.*

**Response:**

We would like to clarify that the accumulation mode of particles in the remote atmosphere is arising from cloud processing where activated Aitken mode particles can and do grow due to a multitude of processes: condensation of gaseous species, liquid phase reactions and coalescence of cloud droplets, all of which leaves aerosol particles larger after cloud evaporation then they were before entering the cloud. Cloud processing leaves the signature "Hoppel minimum" in aerosol size distribution separating Aitken mode from Accumulation mode particles. Coagulation of smaller particles cannot realistically happen in clean remote atmosphere where particle concentration is typically not exceeding several hundred of particles while condensation growth is rarely capable of growing Aitken mode particles to Accumulation mode sizes due to insufficient gaseous precursors in the generally clean remote atmosphere. The wet deposition by precipitation removes the particles from the cloud and atmosphere altogether, but those numbers are negligible due to the fact that cloud droplets are outnumbered by aerosol particles to the ratio of 20 or more. We chose to refer to "cloud processing" without going into specific details or processes.

*Lines 327-329: Based on your classification criteria, Polluted and Mix both feature higher BC mass, I presume the air masses during these periods are from the land. Why did Mix-H show greater similarity in number size distribution to Clean-H? Winds during Polluted-L are from 35° to 135°, whereas winds during Mix-L are from 135° to 190° and from 300° to 35°. It is not accurate to say the prevailing winds are similar during Polluted-L and Mix-L.*

**Response:** The Mixed cases not only include air masses from aforementioned sectors, but also include air masses from 190° to 300° degree clean sector with higher black carbon concentration (> 15 ng m-3). The Mixed-H was indeed similar to Clean-H (except for higher BC), because in the summertime, prevailing air masses from North Atlantic dominated at Mace Head. We have now added Figure S2 and S4 of backward trajectories clusters and wind rose plots.

We modified the sentences in Table 1 for clarification

Revised Table 1:

**Table 1: Classification criteria and total number of hours for each air mass category after data validation**

| Sector | Criteria | Level of biological acitvity | Month | Abbreviation | Duration (hours) |
|---|---|---|---|---|---|
| clean marine | BC < 15 ng $^{-3}$& WD from 190° to 300°& WS > 3 m/s | high (H) | May to Aug | Clean-H | 237 |
| clean marine | BC < 15 ng $^{-3}$ & WD from 190° to 300° & WS > 3 m/s | low (L) | Oct to Apr | Clean-L | 68 |
| polluted continental | WD from 35° to 135 ° | H | May to Aug | Polluted-H | 98 |
| polluted continental | WD from 35° to 135 ° | L | Oct to Apr | Polluted-L | 345 |
| mixed | All the data not included in clean or polluted sectors | H | May to Aug | Mix-H | 343 |
| mixed | All the data not included in clean or polluted sectors | B | Oct to Apr | Mix-L | 319 |

Minor comments:

*Line 152: Since you have not introduced the method F, it is better to delete this sentence here.*

The sentence is now deleted.

*Line 210: Please check the unit of BC in Table 1.*

Corrected.

*Line 306: "Fig. 2 shows" change to "Figure 2 shows".*

Corrected.

*Line 353: Did you use air mass back trajectory? As suggest in the major comments, the back trajectory is*

*important for your classification and data interpretation.*

**Response:** The backward trajectories were added to Figure S2 in the revised manuscript.

*Lines 366-368: In the classification standard, you only mentioned the wind direction at the measurement site. Wind direction cannot tell the air mass during Clean-L from the ocean rather than from continental. As I said in the major comment, the backward trajectory might be a better classification criterion than wind direction.*

**Response:** Indeed, wind direction alone is often insufficient in defining Clean sector from the Polluted one, but together with the conservatively set BC criterion allows for more efficient long-term data processing at minute time resolution leaving least ambiguity over the cleanness of the air masses. When in doubt, we certainly looked at the trajectories and many other auxiliary data to confirm the air mass type. See also O'Dowd et al. (2014) for details of air mass selection criteria and the representativeness of Mace Head observatory.

*Line 452: Change to "Fig. 5".*

Corrected.

Reference:

G. Buonanno, M. Dell'Isola, L. Stabile, A. Viola, Uncertainty budget of the SMPS-APS system in the measurement of PM1, PM2.5, and PM10. Aerosol Sci. Technol. 43, 1130–1141 (2009).

O'Dowd, C. D., Ceburnis, D., Ovadnevaite, J., Vaishya, A., Rinaldi, M., and Facchini, M. C.: Do anthropogenic, continental or coastal aerosol sources impact on a marine aerosol signature at Mace Head?, Atmos Chem Phys, 14, 10687 10704, https://doi.org/10.5194/acp-14-10687-2014, 2014.

Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K. J., Roberts, G., Ceburnis, D., Decesari, S., Rinaldi, M., Hodas, N., Facchini, M. C., Seinfeld, J. H., and C, O. D.: Surface tension prevails over solute effect in organic-influenced cloud droplet activation, Nature, 546, 637 641, https://doi.org/10.1038/nature22806, 2017.

Xu, W., Ovadnevaite, J., Fossum, K. N., Lin, C., Huang, R.-J., O'Dowd, C., and Ceburnis, D.: Aerosol hygroscopicity and its link to chemical composition in the coastal atmosphere of Mace Head: marine and continental air masses, Atmos Chem Phys, 20, 3777–3791, https://doi.org/10.5194/acp-20-3777-2020, 2020.