

Interactive comment on “Factors controlling marine aerosol size distributions and their climate effects over the Northwest Atlantic Ocean region” by Betty Croft et al.

Anonymous Referee #1

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The manuscript provides an overview of particle number and composition measurements over remote marine locations and aims at explaining sources of these particles by deploying the GEOS-Chem-TOMAS model. While it is a really unique set of measurements that is very difficult to obtain over remote locations, especially extending over various seasons, I feel that the model results are often over-interpreted. Particularly, for the cases where model simulations do not quite agree with the measurements, but important claims are made from these anyway. The manuscript has a potential, owing to the measurements, to bring valuable insights and provide new knowledge, however, several key issues must be resolved before publication.

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My main concern is that the paper seems to be biased to discussing and evaluating sources that were selected based on the literature, but not on the current measurements.

To name a few: new particle formation near the surface was not accounted for, nor primary marine sources were seriously considered. E.g. sea salt and sea spray, including organic matter, can be as small as 10 nm in diameter (de Leeuw et al., 2011), however, their effects to DRE and AIE were not evaluated, nor their contribution to vertical distributions analysed. Near surface or MBL NPF was completely omitted, while authors admit that MSOA NPF, which supposedly happens near the surface, can potentially have an impact (lines 275-277). The question is then why they were not included or evaluated? Without the proper evaluation, their role cannot be dismissed and conclusions stating that only cloud base or MBL top NPF are important are based on wrong assumptions.

Moreover, the conclusion on the dominant near MBL top NPF effect was based on very uncertain measurements near the ground. As the reliability of near ground measurements poses some questions, e.g. how the aircraft data were extrapolated to the ground level as, I assume, the airplane did not go down to 0m altitude? And, to my understanding from the methods section, the ship measurements were for particles >20nm in diameter? Therefore, Figure 4 showing N3 and N10 down to 0m as well as lines 406-408 are misleading as they do not represent the real N measurements.

Moreover, in high biological activity May/June period, the N3-N10 maximum extends over the broad amplitude range (~2km wide), as opposed to only the top of the MBL as stated by authors (408-412), therefore, the question is whether the drop in concentration that occurs at the very surface is due to measurement uncertainty? In which case, the NPF might have occurred over the whole boundary layer and not only at the MBL top, but was not detected due to these measurement limitations?

While, if indeed real, such strong gradient in number concentration would imply a con-

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stant source that occurs over larger geographical areas as these new particles are not mixed down into boundary layer within normal 15-20 min mixing, which would diminish the gradient if the source is not constant. The existence of such strong constant and wide source does not seem likely.

The statement on the number concentration maxima observed at 1 km (lines 30-32) is also not robust. The total number concentration, in Figure 3, shows 2 peaks, one at ~1km and other just below 2 km, with the latter being even stronger, so why 1 km maxima is highlighted and how the second maxima is explained? Is that the measurement uncertainty or just noise rather than the real maxima? Similarly, lines 396-397 point to one maximum in figure 3, which is quite subjective as there are many 'maximas' in that figure, but no explanation is provided.

Therefore, the statement 'that NPF near/above the MBL has a strong control on the development of the total particle number maxima near 1 km altitude...' (lines 464-467) is not exactly based on the measurements. Moreover, neither Base nor noABLNUC simulations agree with the measurements, actually, for this season, all N3-N10 simulations are very far from the measurements, so the claims on the processes influencing the number concentrations in the high biological activity season are not substantiated by the measurements or data in this paper. Similarly, the following statement on MBL-top NPF influencing the concentrations of near surface particles (lines 476-748) is not based on the findings as, without model simulations agreeing with the measurements, these are only speculations. There are other features in the simulations that were not properly discussed or explained, like high DMS contribution in winter (Figure 2 and Figure 7, lines 562-565). Provide quantitative (%) estimates when talking about differences between Base and noDMS. It is not so trivial to judge by just looking at the graphs. Also, high DMS effects (as well as MSOA) over continents are not discussed (figure 7).

Finally, the ship emission control does not look so modest to me (lines 593-594 and Figure 2), so, please provide a quantitative estimate of the difference. Also, it seems,

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that noSHIPS agrees better with the measurements than the BASE in figure 2? Discuss that in more details.

Also, why the ship emission effect (Figure 7 and 8) is the highest for the high biological period, discuss the observed seasonality in detail. How do you explain ship effects over continents? High AIE effect (lines 693-695) might as well point to over-prediction of ship influence rather than location-dependence? This would explain better agreement with measurements in Figure 2 when Ship emissions were not included?

Specific comments: Figure 4: noABLNUC is not visible in some panels, e.g. bottom and middle panels in the right column, please adjust colours or patterns.

Ship measurements do not cover particles smaller than 20nm, how can Figure 4 show the concentrations down to the ground level? Surely the aircraft could not have measured at these low altitudes. Were the measurements extrapolated then? How reliable are these extrapolations? Similarly, for Figure 5, how do composition measurements extend to the ground? Provide details on the extrapolation in the method section.

How the total number in figure 3 over August/September can be reconciled with Figure 4 data for the same phase for N10. N20 from fig 3 shows higher concentrations near the ground with reducing trend towards boundary layer top, which would resemble what is expected for a winter boundary layer with sea salt contributions, but N10 shows the opposite trend. Explain this in more details.

de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.: Production flux of sea spray aerosol, *Rev Geophys*, 49, RG2001, doi:10.1029/2010rg000349, 2011.

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