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Interactive comment on “A global process-based study of marine CCN trends and variability” by E. M. Dunne et al.

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Received and published: 7 November 2014

We thank the referees for their helpful comments, and present the following response for their consideration.

General comments:

As stated, both Northern Hemisphere boxes are downwind of North America, the north Atlantic box in the westerlies, the pacific box in the trade winds. Increasing wind speeds here also give shorter travel time from the continent, thus increasing the fraction of continental CCN.

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It is true that CCN concentrations within the regions examined may depend to some extent on wind speeds elsewhere. However, analyzing this effect is very complicated: the winds have not changed uniformly along the transport routes (see Fig. 1 in manuscript for linear wind speed trends); the transport routes themselves may have varied between different years; and part of the transport affecting surface concentrations in the study area is likely to have taken place at higher altitudes than the surface. The transport altitude, and thus deposition of pollution, may also vary between simulated years.

However, to estimate the role of continental transport in our simulations, we performed a trend analysis for the BC concentration within the two NH boxes (Figure 1 in this document). This analysis revealed that, in the Northern Equatorial Pacific, there is a great deal of scatter in carbonaceous aerosol compared to wind speed, with only a small decreasing trend apparent; while in the North Atlantic, accumulation-model carbonaceous aerosol mass decreases strongly with increasing wind speed (likely due to coagulation losses with coarse primary sea spray particles).

Specific comments:

Pg 15774 line 16-17 I assume GLOMAP can also be run on different resolutions? If so, change "It runs", with "It was"

GLOMAP can be run on different resolutions, but it rarely is. The line now reads, "It was run with a T42 spectral resolution..."

Pg 15775 The source function used in GLOMAP, from Mårtensson et al. 2003 has a temperature dependence in production. I cannot see that the effect of this is accounted for in any way. This needs to be addressed as it may well influence the

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production of CCN significantly.

Interannual variations in SST are not accounted for in GLOMAP, as a climatology of sea-surface temperature is used. Note also that the seasonal variation is removed in our analysis before calculating the trend. Figure 10 in Mårtensson et al. (2003) shows that the source function of primary sea spray is much more sensitive to a small change in wind speed than in temperature. We are therefore confident that any temperature effect would be very small - in any case, it is expected to be much smaller than the wind effect, which was not found to be very strong in our study.

The following paragraph has been added to the manuscript: “Interannual variations in sea-surface temperature are not accounted for in GLOMAP, as a climatology of sea-surface temperature is used and does not change from year to year. However, the ? parameterization is more sensitive to a small change in wind speed than in temperature, and CCN were not strongly affected by changes in wind speed.”

Pg 15776 line 22- 24 As production of marine CCN are highly non-linear, thus a monthly average wind speed may produce significantly different numbers of CCN. Taking a non-linear average would sort this as is suggested later on page 15780 line 19, the emission trend.

It should be stressed that the CCN production within the model is calculated every 30 minutes based on interpolation of 6-hourly wind speeds from the ECMWF reanalysis, and not using monthly mean wind speeds. The monthly means are used only in the trend analysis, as it is not feasible to generate model output on six-hourly timescales for long-term simulations of the type described in the paper (however, this was done for the two-month simulations used in Section 3.2 of the paper). It is possible as the reviewer suggests, that the same monthly mean wind speed can arise from different intra-monthly wind speed distributions. We mention this now in the manuscript, but expect it not to have a significant effect on the trend analysis.

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Figure 2 of this document shows (a) the monthly mean wind speed and mass flux for the Northern Equatorial Pacific over the full fifteen years and (b) the six-hourly mean wind speed and number flux for the Northern Equatorial Pacific for January 1990. The monthly mean values follow a power law of 3.26, which is closer to the expected value of 3.41 than the six-hourly value of 2.93. These figures will not be included in the manuscript.

Pg 15781-82 “The inclusion of nucleation scavenging also dampens the effects of other processes. This damping can be seen in the much greater absolute variation between peaks and valleys in the black lines compared to any of the other simulations” How does nucleation scavenging affect new particle formation, primary sea spray and DMS emissions? By visual inspection, this does not appear true in terms of the relative variability to the mean CCN concentrations, at least for the second month of the simulations. This statement needs some explanation.

Nucleation scavenging obviously does not affect sea spray or DMS emissions, as our model does not have a feedback from aerosol changes to atmospheric dynamics. It may, however, affect new particle formation by changing the background particle population onto which newly formed particles can coagulate or nucleating substances condense. The main point here is, however, that scavenging limits the contribution of particles from these sources to CCN concentrations. This is discussed further in our response to your comment on Pg 15786 line 20 below.

When we refer to dampening, we mean a limit on the extent to which processes other than nucleation scavenging can affect absolute CCN concentrations. For example, in Figure 4 (c), the maximum value of the black line (no nucleation scavenging) during the second month is approximately 350, and the minimum is approximately 200. The difference between the two values (the absolute variation between peaks and valleys) is therefore about 150 cm^{-3} , which is more than the maximum absolute

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concentration of CCN in any of the other simulations.

The following text has been altered in the manuscript: the sentences “The inclusion of nucleation scavenging also dampens the effects of other processes. This damping can be seen in the much greater absolute variation between peaks and valleys in the black lines compared to any of the other simulations.” now read “The inclusion of nucleation scavenging also dampens the extent to which other processes can affect absolute CCN concentrations. This damping can be seen in the much greater absolute variation between peaks and valleys in the black lines in Figure ?? compared to any of the other simulations. ”

Pg 15786 line 12-14 I would limit this statement to indirect effect as other climate effects than CCN such as the direct effect has not been examined.

The line has been altered to read “These results imply that in most marine regions the predicted changes in surface wind speed are likely to have only a small effect on future CCN, and the resulting aerosol indirect effect will therefore constitute only a minor climate feedback mechanism.”

Pg 15786 line 20 This dampening effect needs some more explanation. If I understand it correctly, the effect is that a higher percentage of CCN are removed by nucleation scavenging when there is above average CCN numbers and a lower fraction of CCN is removed when there is below average CCN numbers. Why? Alternate interpretations of the dampening would also need some physical explanation.

The dampening effect refers to a limitation on the range of absolute CCN concentration. In simulations which include nucleation scavenging, the absolute value of CCN never gets above 150 cm^{-3} , while simulations without it reach values of more than 400 cm^{-3} . The inclusion of nucleation scavenging therefore limits the maximum

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value of CCN and the number of CCN that can be introduced to the model by other processes.

The model represents our best available simulation of reality by incorporating mathematical representations of different physical processes. The value of a particular output (like CCN) will reach an approximate equilibrium level based on the interactions of those representations, with fluctuations based on the interactions of the processes. If a particular process is omitted, as is the case in the simulations described in Section 3.2, the model output will reach a different equilibrium value. Since nucleation scavenging is the most efficient aerosol removal process in the model, it also controls the level of this equilibrium value. The equilibrium value reached when nucleation scavenging is omitted is much higher than the “true” equilibrium value in a simulation where nucleation scavenging is taken into account (along with all other processes). Thus, nucleation scavenging simply reduces the amplitude of variation in CCN concentration causing a dampening effect.

References

Mårtensson, E. M., Nilsson, E. D., de Leeuw, G., Cohen, L. H. and Hansson, H.-C. (2003), “Laboratory simulations and parameterization of the primary marine aerosol production”, *J. Geophys. Res.*, Vol. 108, p. 4297.

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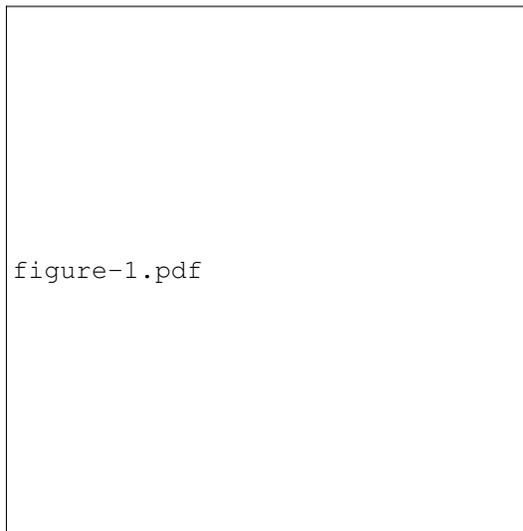
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Fig. 1. Trend analysis between wind speed and carbonaceous aerosol in the Northern Equatorial Pacific and North Atlantic.

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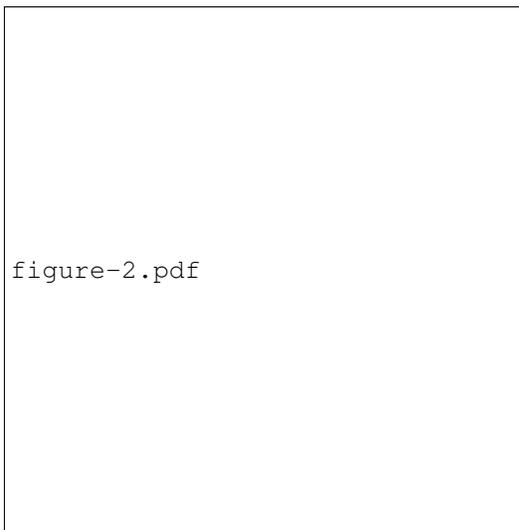
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Fig. 2. (a) Monthly mean mass fluxes of sea spray in the Northern Equatorial Pacific from 1990 to 2004 as a function of monthly mean wind speed. (b) Six-hourly number fluxes of sea spray in the Northern Equatorial Pacific in January 1990 as a function of six-hourly wind speed at 10m altitude. Both emissions fluxes fit perfectly to a power-law function of wind speed, although neither has exactly the theoretical exponent of 3.41.

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