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Response to Referees of *A global process-based study of marine CCN trends and variability*

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

1 Referee #1

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.

It is true that CCN concentrations within the regions examined may depend to some extent on wind speeds elsewhere. However, analyszing this effect is very complicated: the winds have not changed uniformly along the transport routes (see Fig. 1 in the 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 rou simulations, we performed a trend analysis for the BC concentration within the two NH boxes (Figure 1 in this document). This anal-

ysis 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 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 Mårtensson et al. (2003) 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 typie 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. 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.

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 of 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.

Here we talk about dampening variations in terms of 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 minumum 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 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 int eh 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 concenentrations. This damping can be seen in the much greater absolute variation between peaks and valleys in the black lines in Figure 4 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 faction of CCN is removed when there is below average CCN numbers. Why? Alternate interpretations of the dampening would also need some physical explanation.

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.

2 Referee #2

Section 3.3: When addressing the influence of continental aerosols, I had three additional questions:

1) Is the marine CCN continental aerosol influence a linear spectrum or are marine areas either pristine or not pristine with some tipping point in between?

The influence of continental aerosol on marine CCN is a spectrum, and will depend on the length and course of transport routes, and on meteorology along the transport pathway. For the purposes of this paper, we use low mass concentration of black carbon aerosol as a metric of how pristine a region is. As such, we do not have an exact numeric value to separate pristine areas from areas with strong continental influence.

2) Does each region have a similar value of low carbonaceous aerosol concentrations when other aerosol components correlate with marine CCN?

It would be very challenging to come up with a threshold value of carbonaceous aerosol below which the other components correlate with CCN, because the local CCN concentration also depends on transport of natural CCN from other regions, as well as dry and wet deposition of particles. Furthermore, sulphate has both continental and marine sources, and therefore it is not possible to distinguish completely between continental and marine aerosol sources.

3) Is there a trend in the 1990-2004 dataset in which marine regions have become more or less pristine?

Because the emphasis in these simulations was to examine long-term trends in wind-driven, marine aerosol, the same continental emissions were used for each of the fifteen years in the simulations. Only trends in aerosol removal processes due to changes in meteorology will affect the simulated concentrations of continental aerosol in marine regions. We found that no marine region in the model became more or less pristine over the 15-year period, although the same may or may not be true in reality.

Figure 1: I think that a global map of the linear trend in CCN concentration should be added as a part (b) to give spatial context to the Figure 3 CCN plots.

The figure is shown as Figure 3 of this document; however, given that one of the main findings of the paper is that linear trends are insufficient to determine the response of the aerosol system to wind speed, it will not be included in the revised manuscript.

Figure 3: I would suggest combining the left and right columns into one plot with a primary and secondary y-axis to improve comparison of the trends.

The revised Figure 3 is Figure 4 in this document.

Figure 5: Please describe how the accumulation-mode concentrations were norimalised.

The concentrations were scaled to be between the minimum and maximum values of CCN, as described in Eqn. 1. "Scaled" is a better term than "normalised", and will be substituted in the paper. Essentially, the limits of the mass line is confined to the range of the CCN line, purely for ease of

visual comparison.

$$m_{scale} = min(ccn) + [max(ccn) - min(ccn)] * \frac{max(mass) - m)}{max(mass) - min(mass)} \quad (1)$$

An error in the plotting subroutine meant that the lines were mis-scaled in the original draft. The new versions, which are properly scaled, have been included as Figure 5 of this document.

Figure 6-8: Please put the accumulation mode mass units in concentrations (eg. g m^{-3} or ng m^{-3}) more typical of other models and observations.

The units have been adjusted and the figures re-plotted - they are included as Figures 6-8 of this document.

Additional (supplemental?) figures: After reading about continental aerosol-influenced vs pristine environments, I think that there two additional figures would help inform the reader about the representativeness of the three regions:

1) A series of 4 global maps showing the Pearson coefficients between monthly mean CCN and monthly mean aerosol component mass for each grid cell;

2) I'm not sure of the metric, but I was wondering if it is possible to produce a global map showing the spatial extent of pristine regions predicted by the model?

The Pearson coefficient maps in Figure 9 of the supplement to this document show correlations at 915 hPa between concentrations of CCN and mass concentrations of each of the four aerosol components traced in the model. Where a particular component makes a strong contribution to CCN - sea spray in the Southern Ocean, for example - that component will show a strong correlation with CCN concentration. However, if CCN concentrations increase or decrease, mass concentrations of all aerosol components would also increase or decrease to a certain extent. As a result, there can be strong correlations between CCN concentrations and mass concentrations of multiple components, even when changes in a given component do not affect CCN concentrations. The maps therefore do not provide sufficient information to infer a causal link, and will be omitted from the revised manuscript.

Figure 10 of the supplement shows the concentration of black carbon in ng m^{-3} at 915 hPa. The concentration of BC is a good proxy for continental influence in these simulations, since marine carbonaceous aerosol is not included in these simulations. By comparing Fig. 10 with Fig. 9(d), we identify a region in the Southern Ocean (approximately 30-60S, 30-90E) where black carbon has a concentration of a few ng m^{-3} , and CCN concentrations correlate with marine aerosol - sea

spray and sulphate - and does not correlate with carbonaceous aerosol. However, while this map provides a guideline as to which regions have stronger or weaker continental influence, it cannot definitively map out pristine marine regions. For this reason, we will also omit this map from the revised manuscript.

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.

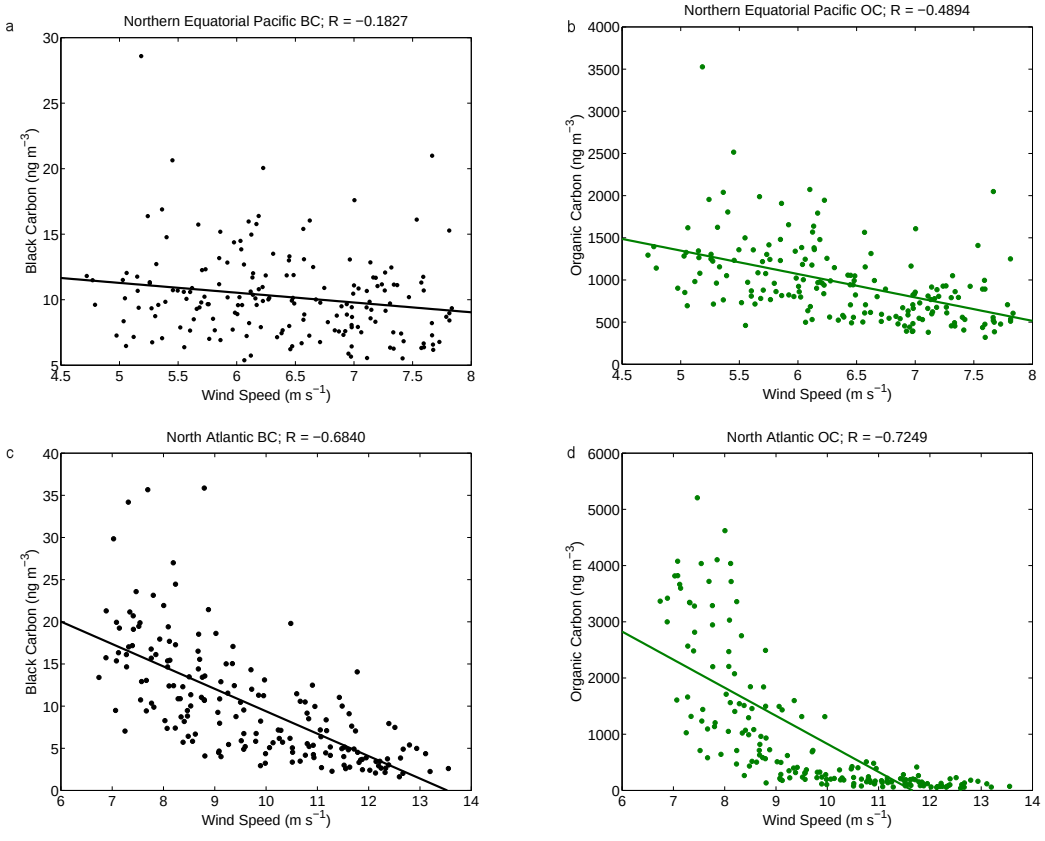

Fig. 1. Trend analysis between wind speed and carbonaceous aerosol in the Northern Equatorial Pacific and North Atlantic.

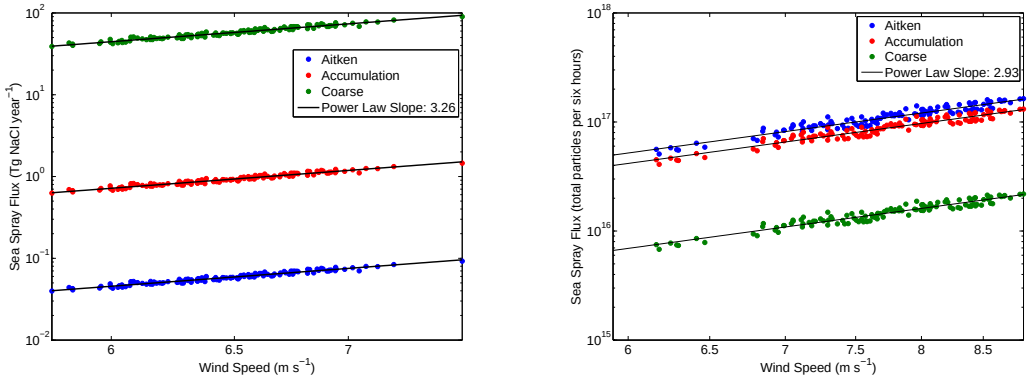

Fig. 2. (a) Monthly mean mass fluxes of sea spray in the Northern Equatorial Pacific from 1990 to 2004 as a function of monthly mean wind speed. (b) Six-hourly number fluxes of sea spray in the Northern Equatorial Pacific in January 1990 as a function of six-hourly wind speed at 10m altitude. Both emissions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

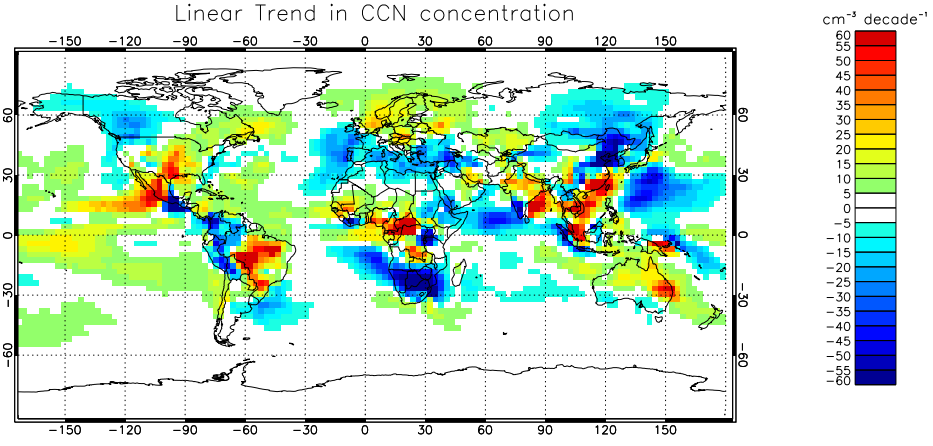

Fig. 3. Linear trend in CCN at 915 hPa for the years 1990-2004.

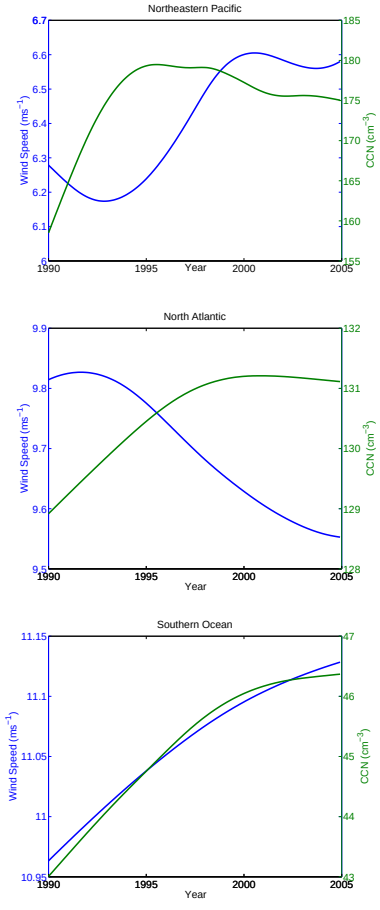

Fig. 4. Trends in wind speed (blue line) and CCN concentrations (green line) in the Northern Equatorial Pacific, North Atlantic, and Southern Ocean.

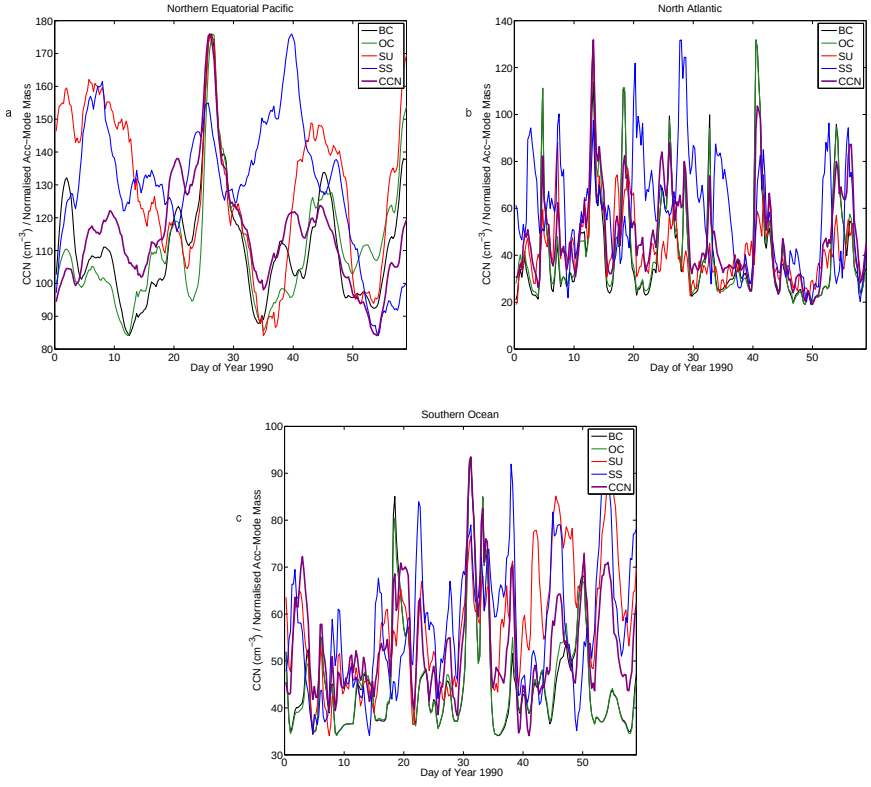

Fig. 5. A time series of normalised accumulation-mode mass of black carbon (black line), organic carbon (green line), sea spray (blue line), and sulphate (red line), overlaid with time series of CCN concentrations (purple line) in the three regions being examined.

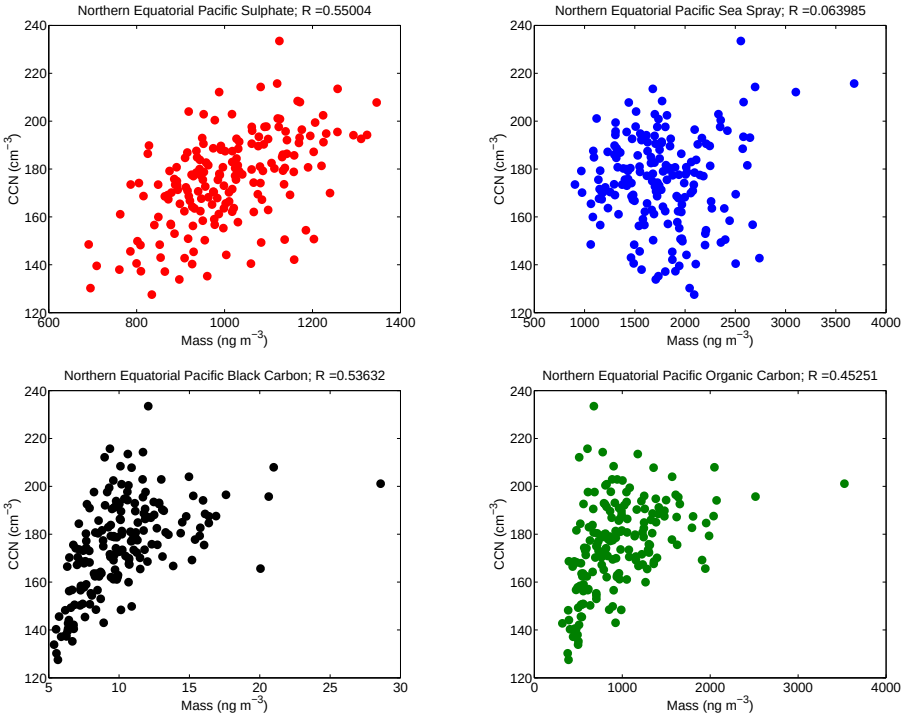

Fig. 6. Scatterplot of simulated monthly mean CCN concentration versus accumulation-mode mass of (a) sulphate, (b) sea spray, (c) black carbon, and (d) organic carbon, in the Northern Equatorial Pacific.

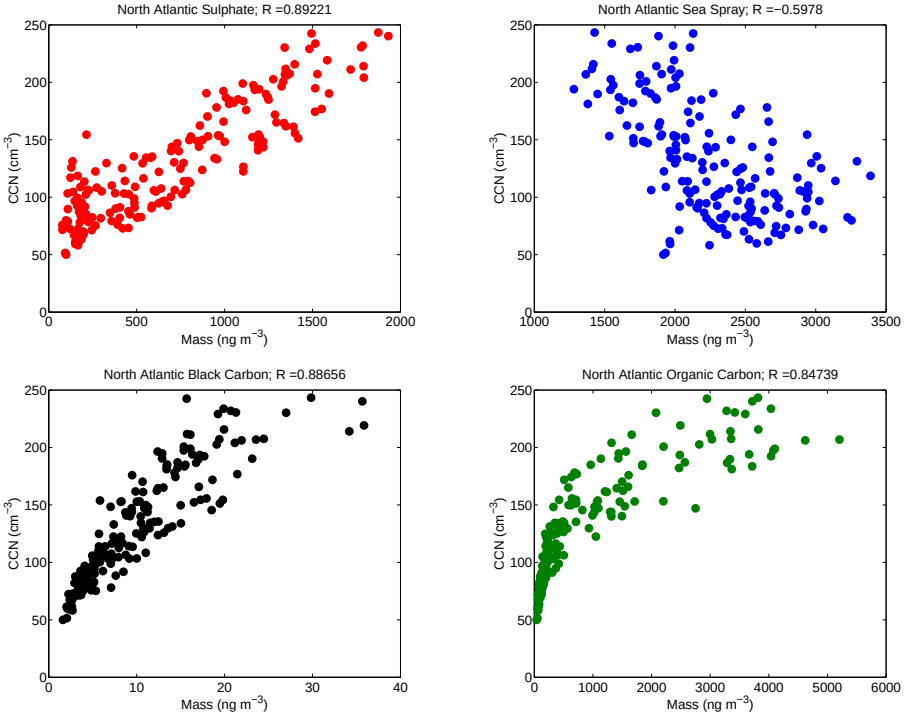

Fig. 7. As for Figure 6, but for the North Atlantic.

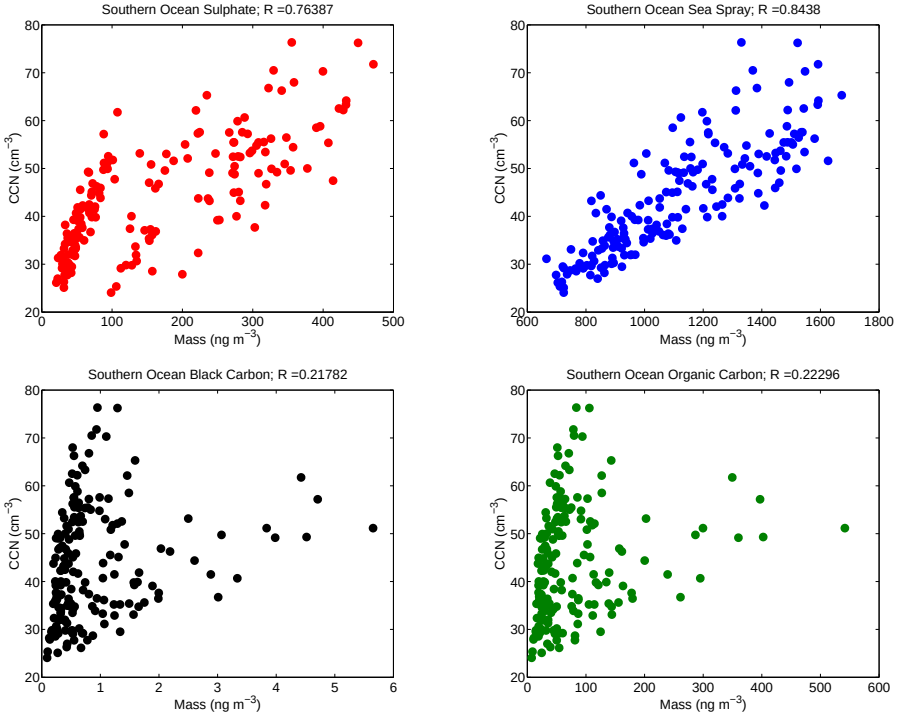

Fig. 8. As for Figure 6, but for the Southern Ocean.

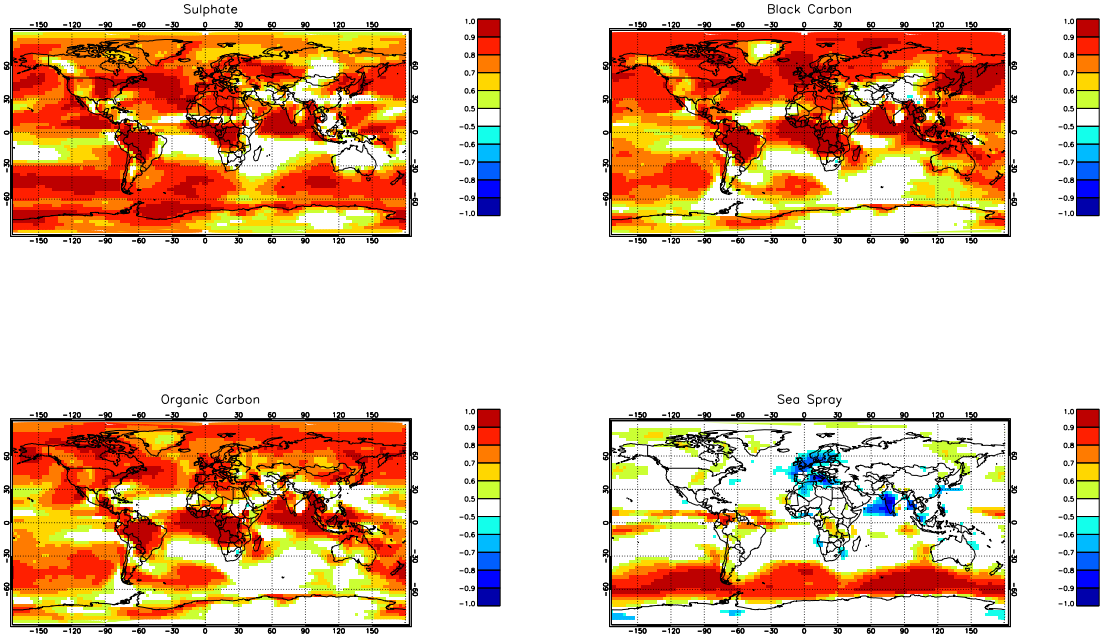

Fig. 9. Pearson coefficients between monthly mean CCN and monthly mean accumulation-mode mass for sulphate, black carbon, organic carbon, and sea spray.

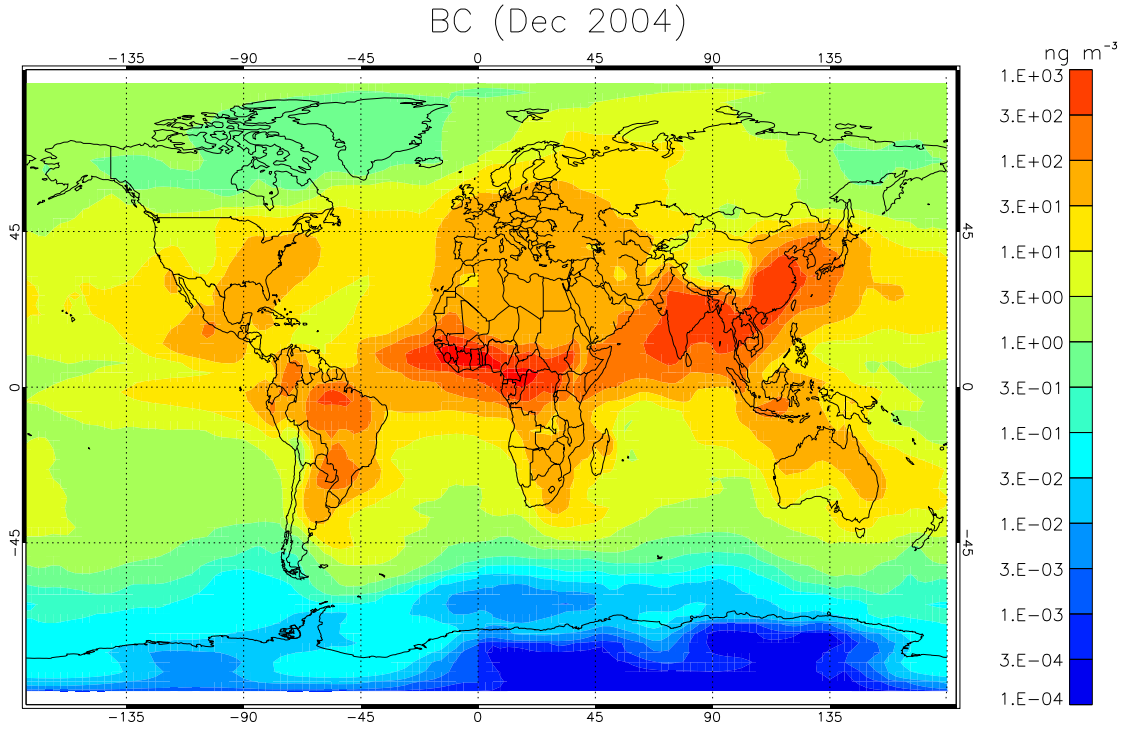

Fig. 10. Map of black carbon concentration (in ng m^{-3}) at 915 hPa in December 2004. Low concentrations of black carbon reflect a low influence of continental emissions.