

Interactive comment on “Decreasing particle number concentrations in a warming atmosphere and implications” by F. Yu et al.

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The authors thank the referee for taking the time to review the manuscript. The detailed comments are insightful and constructive. Our replies to these comments and actions taken to revise the paper are given below.

1. Statistical analysis of CN data.

We have added statistical analysis by giving the linear correlation coefficient (also called Pearson’s correlation coefficient) to illustrate the significance of trends.

Yes, we deseasonalized the data before calculating the trends. The trends given in the figure are the averages of 12 individual months. To look into the data from a different perspective, we provide the linear trends for annual mean CN (for all years having at

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least 10 months of data) in the revised Figure 4 but keep the average of monthly mean trends in Figure S1 of Supplementary Material.

Missing data. Unfortunately periods of missing data are common for many observations, especially for long-term measurements such as the CN data analyzed here. While the periods of missing data may increase the uncertainties, the derived linear trends should be still valid as long as there is no significant systematic shift in the measured values before and after the period of missing data. The question here is, as the referee has pointed out, was there a change in sampling technique, sampling lines or position of measurement inlet that might systematically affect the measured values? We could not find record to indicate such a change. It should be noted that measurements at NOAA background sites are intended to be long-term and instruments are relatively well maintained. While we agree with referee that “at Barrow the majority of the decline in CN concentrations appears to occur during a period when no observations are available”, we would like to point out: (1) Similar magnitude of change during a period of several years is not uncommon due to inter-annual variations; (2) No obvious change in observed CN values during the extended periods of missing data at MLO and SMO; (3) The decrease in CN from ~ 1990 to 1995 at Barrow coincides with the substantial reduction for SO₂ in North America associated with clean air act. As to separate CN trends for the 2 periods over which observations are available (1975–1990 and 1995–2010), both are positive. The first period is likely to be associated with increased SO₂ emission in North America during 80s while the second period may be a result of increased SO₂ emission in Asia during the last decade. As expected, anthropogenic emissions have large impacts on CN values at Barrow. The differences in the sign of the trends for different periods highlight the necessity to use long term data to derive meaningful long-term trends. It should be noted that the conclusions of this paper rely more on other NOAA sites (MLO, SMO, SPO) where effect of missing data appears to be smaller.

In summary, periods of missing data increase the uncertainties in our derived trends

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but our conclusions about the long-term decreasing trends of CN concentrations at the NOAA background sites should still be valid.

2-4. Model simulations and observed trends.

We agree with the referee that additional analysis and simulation are required in order to identify the most likely cause of declining CN concentrations. Following the referee's suggestion, instead of attributing the trends to DMS emission, we give a broad list of possibilities that might explain trends and that could be explored in future work. The abstract, text, and summary have been revised accordingly.

5. Yes, we only consider the effect of changing temperature in the particle formation in our sensitivity study. As we pointed out in the ACPD paper (page 27918, lines 4-8): "In the real atmosphere, global temperature change is inhomogeneous and has many other associated changes (meteorological fields, emissions, chemistry, etc.). The sensitivity study presented here is aimed to isolate and illustrate the impact of temperature change on nucleation and particle number concentrations." The impact of changing temperature on gas-phase reaction rates is not considered. We point this out explicitly in the revised manuscript.

6. As pointed out in the figure caption and in the text, Figure 6b is simply the scaled CCN change using the CCSM3 projected temperature changes. The purpose is to give "an example of how such warming might impact CCN abundances in different regions as a result of direct temperature effects on nucleation rates" as projected global warming has large spatial variations (Page 27963, Lines 14-16). We didn't say that Figure 6b is the "calculated future (2080-2099) changes in CCN concentrations". We agree with the referee that "Many other non-linear interactions would be expected in each of the links between T and indirect forcing". We still feel that Fig. 6b is useful to give order of magnitude of the effect of proposed positive feedback mechanism alone on CCN abundance in different regions. To address the referee's concerns, we make it clearer that Fig. 6b is the linearly scaled CCN changes and the real changes depend

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on many other non-linear processes which need further study.

Minor comments:

These are monthly mean CN data. Sorry for the typo. In the revised manuscript, we added the annual mean values. As to the trends in winter and summer, this is a good point. We agree that the contribution of particle formation and DMS emission to total particle number is expected to be smaller in winter than in summer. In the revised manuscript, DMS emission change is proposed as one of the possible reasons for the observed long-term trends, and further research is needed to understand how sensitive CN values are to DMS emission change and other processes.

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