

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

### **Anonymous Referee #2**

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This study presents long term (1975-2010) records of particle number concentrations observed at several sites around the world and uses a global aerosol model to explore observed trends over this period. The paper attempts to explain trends through changes to atmospheric temperature and changes to dimethyl sulfide emissions.

This is an interesting study and one of the first to explore impacts of changes to temperature on particle formation and atmospheric aerosol. However, there are several aspects of this paper that, in my opinion, means that it is not publishable in its present form. Most importantly the link between trends in particle number concentrations and DMS emissions which is a major conclusion of the paper is not demonstrated. Either the authors need to include new analysis to demonstrate the DMS-aerosol link or this conclusion needs to be removed from the paper. The authors could instead discuss a

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number of possibilities that might explain trends in CN concentrations (e.g., changes in natural and/or anthropogenic aerosol emissions, changes to climate) that could be explored in future work. I outline major comments to the paper below.

1) There is no statistical analysis of the reported trends in observed particle number (CN) concentration.

Given the strong temporal variability (both seasonal and interannual) in observed CN concentrations it is important that the authors demonstrate the significance of the reported trends. No such statistical analysis is presented. These trends in CN concentrations are the foundation of the paper and so they need to be solid. Did you deseasonalize the data before calculating trends? This would be standard practice but does not appear to have been done here. Furthermore, you calculate linear regressions across periods that include substantial chunks of missing data. Reporting these linear regressions can be misleading. For example, at Barrow the majority of the decline in CN concentrations appears to occur during a period when no observations are available. Was there a change in sampling technique, sampling lines or position of measurement inlet that might explain this change? What are the separate CN trends for the 2 periods over which observations are available (1975-1990 and 1995-2010). Are these trends for these periods significant? I have similar concerns for data from other sites including MLO and SMO. A more exhaustive analysis of the observations is required to convince the reader that trends are statistically significant.

2) The model simulation does not capture the reported observed trends.

Simulated changes in CN are almost an order of magnitude smaller than observed trends. This suggests that other variables (assumed to be constant in this study) are more important in driving the observed trends. Obvious candidates are changes to anthropogenic aerosol emissions and precursors, changes to natural aerosol emissions (including, but not limited to DMS) and changes to climate. Since the authors make no attempt to explore these different drivers it is not possible to single out one as the

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dominant cause. It is not clear to me why the authors decide that changes to DMS emissions are the most likely candidate. Additional analysis is required here if the authors want to identify the most likely cause of declining CN concentrations. Else the authors need to discuss a broad range of possibilities that might explain trends and that could be explored in future work.

### 3) Changing anthropogenic emissions

The authors assume that anthropogenic emissions of aerosols and aerosol precursors do not change over the study period. However, in reality substantial regional and global changes in anthropogenic emissions have occurred. Most obvious and well documented are changes to anthropogenic sulfur emissions. These are discussed briefly by the authors but then not treated. Ignoring these changes is a serious simplification. Changes in anthropogenic emissions may be the dominant cause for the observed changes in CN concentrations. Long-range transport of anthropogenic pollutants at regional and global scales are well documented in the literature and so even “remote” sites may be heavily influenced by anthropogenic pollution.

If the authors want to make claims about which source (e.g., DMS versus anthropogenic) is most likely to explain CN trends this needs to be explored in detail using the model. Else the paper needs to discuss a wide range of potential causes that can be explored in future work.

### 4) No evidence is provided to support the postulated DMS-based climate feedback.

The authors postulate that changing DMS emissions, driven by changes in climate, are responsible for much of the observed trends in CN concentration (that they can not explain by changes to temperature alone). If the authors were able to demonstrate such a DMS-based aerosol feedback process this would be a very important result. However, no evidence is provided to support this link. The authors do not complete any model runs with modified DMS emissions nor do they provide any evidence that DMS emissions have actually declined. How sensitive are simulated CN and CCN

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concentrations to changing DMS emissions? What change in DMS emissions would be required in your model to match the observed change in CN concentrations? Is such a change in DMS reasonable over the 1975-2010 period? Is the CN at “remote” sites much more sensitive to DMS emissions than other natural or anthropogenic emissions? This is assumed by the authors but not demonstrated. Without such analysis the author’s conclusions are very speculative and unfounded. Furthermore, it should be noted that even if a climate-DMS-CN interaction was demonstrated this does not demonstrate that a feedback upon climate occurs. This requires several additional steps in the feedback cycle to be proven. Other natural aerosol emissions (sea salt, wildfire) and precursors (BVOCs) are driven at least in part by climate and therefore may have changed over the study period. Changes to any one of these sources may be just as likely to be responsible for any possible trend in CN. Why did you single out DMS from all these potential drivers? Can the authors discount changes to any other natural aerosol emission?

5) Climate driven changes to aerosol are not accounted for. Do you only account for changing temperature in the particle formation mechanism? It is not clear whether you include the impact of changing temperature on gas-phase reaction rates. Other important changes to climate (e.g., rainfall, boundary layer depth, wind speed etc) that may have occurred are not explored but may be important.

6) CCN changes under climate change.

This is an important result and needs to be well founded. The authors need to give more information on how they calculated future (2080-2099) changes in CCN concentrations. This is not well explained at present. Did you extrapolate from the change in CCN from your simulation of a 1K increment? If so, is the T-nucleation rate – CN concentration – CCN concentration – indirect climate forcing system linear? There are a number of reasons to think that it is very non-linear. For example, Figure 1 shows that the relationship between T and nucleation rate is not linear. Many other non-linear interactions would be expected in each of the links between T and indirect forcing. It is

therefore not appropriate to make the simple scaling that you have applied here. In my opinion the analysis here is not sufficient to support this important conclusion. Either more analysis if required or this aspect of the paper should be removed.

#### Minor comments

Figure 4. Are these annual mean values as stated in the caption? Or are they monthly mean values? If they are monthly mean values then winter time CN concentrations appear to be declining as fast as summer time concentrations. Is this true? If so, does this support your hypothesis that trends are driven by changes to particle formation and DMS emissions? I would expect the contribution of particle formation and DMS emission to total particle number to be smaller in winter than in summer.

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