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Atmospheric Chemistry and Physics Discussions

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

J. Pierce (Referee)

jeffrey.robert.pierce@gmail.com

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Review of "Decreasing particle number concentrations in a warming atmosphere and implications" by Yu et al.

This manuscript presents the interesting and perhaps overlooked possibility that aerosol number concentrations may decrease in the future due to increasing temperatures. This hypothesis is driven by aerosol nucleation being less favorable with increasing temperature (all else being fixed). The authors test the changes in the aerosol (and CCN) concentrations using GEOS-Chem with online aerosol microphysics and show the effect on CCN to potentially be important. The authors explore the only possible observational evidence (that I can think of) for whether or not this phenomena may be occurring, long-term CN measurements.

The work is very interesting and is certainly within the scope of ACP. I have, however, several important issues that should be relatively easy to address in the paper. Once these have been address, I recommend publication.

General issues:

- The authors show 5 nucleation schemes and their temperature dependence. Nucleation rates are generally a strong function of temperature for these schemes. However, in field studies in the continental boundary layer, nucleation has not always shown to be greatly affected by temperature (e.g. Mikkonen et al., 2011 and Hamed et al., 2007 – though in the latter paper it could be that higher temps correlate w/ other variable changes that make nucleation more favorable). None of the nucleation schemes in the paper include all nucleating species (e.g. Amines) and thus might not capture the right temperature dependence. Obviously the nucleation must be temperature dependent to some degree. However, the strength of the conclusions of the paper depends on how well the temperature dependence is known. Please modify the text and conclusions to reflect the uncertainties here.

- I have 2 worries about the CN data: (1) There are occasionally jumps in the data even when there were not instrument changes (e.g. Barrow in the early 90s after a 3 year gap, Bondsville for $\sim$2 years around 2006, MLO around 1995 after a 3 year gap). The measurements may be influenced by losses in sampling lines and local sources (e.g. generators). Therefore there may be changes in CN due to other other changes at the sampling site. Is there any evidence of these in the data records? Please comment on these possibilities in the paper. (2) How confident are you that the reduction in anthropogenic emissions has negligible effect on CN at the remote sites? There doesn't seem much of a basis to entirely throw out the possibility of anthropogenic influence at these locations. Given a typical SO2 lifetime of $\sim$3 days (and aerosol lifetimes of $\sim$1 week) it certainly seems like anthropogenic SO2 from SH industrial centers could influence SH remote sites to at least some degree. GEOS-Chem simulations could test this. Without additional evidence, anthropogenic emissions are as likely as reverse

CLAW in explaining the trends at the remote sites. Please make this uncertainty in the influence in anthropogenic emissions much more clear in the paper. Stronger claims could be made in a future paper where changes in anthropogenic and DMS emissions are tested in GEOS-Chem.

- Have ships measured oceanic DMS concentrations over the past 30 years? This would give some direct evidence of whether or not the reverse CLAW is happening.

Specific points:

P27918 L3: So this 1 deg C increase ONLY affects the nucleation rates, not the chemistry and biogenic VOC emissions? I deduced this from the text, but it would be good if it was explicitly stated.

P27921 L25: Dal Mason should be Dal Maso.

Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Dal Maso, M., Kulmala, M., Cavalli, F., Fuzzi, S., Facchini, M. C., Decesari, S., Mircea, M., Lehtinen, K. E. J., and Laaksonen, A.: Nucleation and growth of new particles in Po Valley, Italy, Atmos. Chem. Phys., 7, 355-376, doi:10.5194/acp-7-355-2007, 2007.

Mikkonen, S., Korhonen, H., Romakkaniemi, S., Smith, J. N., Joutsensaari, J., Lehtinen, K. E. J., Hamed, A., Breider, T. J., Birmili, W., Spindler, G., Plass-Duelmer, C., Facchini, M. C., and Laaksonen, A.: Meteorological and trace gas factors affecting the number concentration of atmospheric Aitken (Dp = 50 nm) particles in the continental boundary layer: parameterization using a multivariate mixed effects model, Geosci. Model Dev., 4, 1-13, doi:10.5194/gmd-4-1-2011, 2011.

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