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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions from DMS and ship emissions to CCN observed over the summertime North Pacific" *by* L. Phinney et al.

L. Phinney et al.

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Response to Reviewer #2

We would like to thank Reviewer #2 for thoughtful and constructive comments on our manuscript. Please find below the responses to the reviewer's comments.

1: The authors make a strong point that during their measurements, there was one day during which marine boundary layer (MBL) nucleation contributed to CCN formation and one day during which organic ship emissions contributed to CCN. How about rest of the days? Apparently, sulfur compounds deriving from DMS oxidation played a central role in providing CCN, but where the seed particles were coming from. Were they i) particles nucleated in the free troposphere and entrained from there into the MBL, ii) primary particles emitted by the ocean (sea salt and organics), or iii) long-range trans-

ported anthropogenic particles? The authors should discuss this issue explicitly and address whether their measurements could give any hints on it.

AUTHOR RESPONSE:

This is an interesting and of course important question. Based on the amount of sulphate, our results suggest that the aerosol other than two events we focus on (ship emissions and nucleation) was at least a day distant of the seeds, and we can not delineate whether the sources of the seed particles are primary or nucleation. Since nucleation is not a primary process, the fact that we found only one instance suggests that the conditions in our sampling area were generally not favourable for this process. One of those conditions may have been insufficient SO_2 relative to the surface area of the aerosol, something that is difficult to assess on a larger scale. We have no evidence from the MOUDI samples (Phinney et al., 2006) that sea salt was a seed for particles smaller than about 0.5 μm. Primary organics from the ocean are a possibility, but primary organics from ship emissions might be a larger source. There are also other possible sources, including those you have mentioned, i.e. entrainment into the boundary layer from the free troposphere, primary ocean emissions (sea salt measurements are discussed and related to wind speed in Phinney et al., 2006), long range transport, as well as diffused and aged anthropogenic emissions from ships. Though we present case studies of specific particle events, which we explain with our measurements, our experiment was not designed specifically to identify all the sources of the particles, so it is difficult to do so with the data we collected.

2: The study suggests that secondary aerosol formation (via addition of sulfur species into pre-existing smaller particles) plays an important role in the CCN budget of a remote MBL. The author could discuss briefly how/whether differs from CCN sources in a continental boundary layer (e.g. primary vs. secondary aerosol sources).

Combustion sources, whether on land or over water, produce many small carbonaceous particles that can serve to determine the number concentrations of particles and

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CCN. There is no reason to expect that this process would be significantly different over land. This process is consistent with the discussion in Dusek et al., 2006, showing the size distribution of particles to be the most important factor for determining the number of CCN (Dusek et al, 2006; *Size matters more than chemistry for cloud-nucleating ability of aerosol particles*, Science 312 (5778), pp 1375-1378).

3: I am slightly puzzled with the CCN measurements? It remains unclear how quantitative they are. Do real CCN concentrations depend linearly on deltaV? Are CCN values at different supersations comparable to each other (i.e. if deltaV is higher by a certain factor at 0.34% supersaturation as compared with 0.19% saturation, are real CCN concentrations higher by the same factor)? I do not know wheter issues have been addressed in the paper by Shantz et al. (2008), but they should be briefly mentioned here as well.

The following sentence has been removed (p315 line 9):

"The CCN observations are reported here as deltaV, the difference between the voltage signal from the scattered light and the baseline voltage. The calibration factor for converting deltaV to number concentration varies with the size and composition of the particle as well as the supersaturation. This issue is discussed further in Shantz et al., 2008."

And the following explanation has been added in its place:

"The CCN number concentration is approximately linearly dependent on deltaV (the difference between the signal from the scattered light and the baseline voltage) and can thus be calculated by multiplying deltaV by a constant. Laboratory calibration of the CCN counter for sulphate particles ranging in size from 75 nm to 240 nm found the following relationships between CCN number concentration and deltaV, independent of particle size:

$CCN_{0.19\%}$ = deltaV*185 (6)

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$CCN_{0.35\%}$ = deltaV*130 (7)

for CCN activated at 0.19% and 0.35% supersaturations, respectively. These equations are used to calculate CCN number concentration from deltaV in this paper."

4: I doubt that particles in a remote marine boundary layer would grow by condensation larger than 1000 nm diameter (page 321, line 4).

Removed the phrase "...while the particles on the upper end grow out of the Q-AMS detection range $>$ 1000 nm)."

5: I can not follow the discussion on page 321 (lines 22-28). How would particle number size distributions alone tell anything about effective CCN radius?

This sentence has been changed to "This suggests that the source of the higher CCN/CN ratio is greater aerosol activation, rather than a shifted size distribution."

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 309, 2009.

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