

Interactive comment on “Observations of sulfur dioxide uptake and new particle formation in a midlatitude cumulus cloud” by J. R. Peter et al.

Anonymous Referee #1

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The relative importance of in-situ formation of new particles vs entrainment from the free-troposphere into the clean marine boundary layer (MBL) is still far from clear, with considerable disagreement among different groups in the community. It is known with little doubt that following precipitation and associated reduction of aerosol surface area, new nucleation can occur in the MBL. However, an ability to quantify the new nucleation rate, and therefore the relative importance of the source, is still lacking.

The amount of observational work that can cast light on this issue is actually quite small, and so studies of the type presented by Peter and collaborators are badly needed. What Peter et al. attempt to do is to construct a cloud-scale budget on the number concentration of aerosols to assess the likely impact that the clouds themselves have on the number concentration in the clean MBL. They conclude that there

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is a source of new particles within the cloud system. Their lack of evidence for new particle formation in the regions *adjacent* to the clouds, leads them to speculate that the new source must be *within* the cloud itself. Unfortunately, it is widely recognized that CN counters are unable to reliably measure particles within clouds due to droplet shattering on the inlet. Thus, the evidence for new nucleation is inferred only directly. Budget analysis is a potentially promising way to assess the potential for new nucleation in the MBL. Although I think the paper is interesting, I have some major issues with the study as it stands that must be addressed before I can have confidence that new nucleation occurred in this case.

Chiefly, I have two concerns. The first concerns the methodology used:

I cannot understand Fig. 4, which would appear to imply that, for an in-cloud residence time of 30 minutes (typical for stratocumulus-topped boundary layers) all aerosol particles larger smaller than about $r=50$ nm would be scavenged after a single traverse through cloud. And yet, we *almost always* observe a persistent and active Aitken mode population in the clean MBL with a mean radius of typically 25 nm. Also, I don't understand how one can obtain $CN_{predicted}/CN_{observed}$ as a function of radius r from the observations when no size distribution was measured at small r . Maybe I'm missing the point of this figure. I would like the authors to clarify what they have done. In addition, the authors need to give me details about the parameters used in their model.

The second concern is about the physics that would allow nucleation to occur *within* cloud:

Calculations (e.g. Capaldo et al. 1999), and even a cursory examination of the form of the nucleation rate (e.g. Jaeger-Voirol and Mirabel 1989) suggest that new nucleation would take place in the high relative humidity regions with strong sunlight either just above cloud or adjacent to the cloud. There was no evidence for new particle formation in the regions adjacent to the cloud in this study. The high surface area (of cloud droplets) within the body of the cloud itself would be expected to remove excess sul-

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furic acid rapidly and quench nucleation. The timescale for removal of excess sulfuric acid is roughly equal to $1/(10 \cdot A)$ where A is the aerosol-or-cloud particle surface area ($\text{m}^2 \text{m}^{-3}$). Thus, for a very low aerosol surface area ($10^{-5} \text{m}^2 \text{m}^{-3}$), a long timescale of several hours could allow H_2SO_4 to increase to levels where new nucleation can occur. In clean marine clouds, where A is typically $0.01\text{--}0.1 \text{m}^2 \text{m}^{-3}$, the timescale for removal of H_2SO_4 is 1-10 seconds. It is therefore difficult to understand how sulfuric acid concentrations could build up significantly to the levels required to nucleate new particles. At the least, the authors need to comment on this, and give some reason why nucleation would be favored within cloud rather than out of cloud.

Capaldo, K. P., P. Kasibhatla, and S. N. Pandis, 1999: Is aerosol production within the remote marine boundary layer sufficient to maintain observed concentrations? *J. Geophys. Res.*, 104, 3483-3500. Jaeker-Voirol, A. and P. Mirabel, 1989: Heteromolecular nucleation in the sulfuric acid-water system. *Atmos. Environ.*, 23, 2053-2057.

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