Atmos. Chem. Phys. Discuss., 11, C3428–C3430, 2011
www.atmos-chem-phys-discuss.net/11/C3428/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "In situ observations of new particle formation in the tropical upper troposphere: the role of clouds and the nucleation mechanism" *by* R. Weigel et al.

Anonymous Referee #3

Received and published: 16 May 2011

In this paper, the authors present the data of ultrafine particle formation (UPF) observed in the continental tropical Upper Troposphere (UT) and Tropical Tropopause Layer (TTL) during several field campaigns, and investigate whether observed concentrations of newly formed particles can be reproduced by a aerosol model. The paper provides some interesting observational results, and is well written and suitable for ACP, although some clarifications/additional information are needed to improve the paper. I recommend the publication of manuscript in ACP after the following comments are properly addressed and reflected in a revised version.

Major comments:

1. As the authors have acknowledged, one limitation of their modeling approach is that the mixing of air masses with surrounding air, which are well-known to occur significantly in the convective outflows, was not considered. One question is that how this limitation may affect the conclusion of the modeling results that "predicted concentrations of ultrafine particles in reasonable agreement with the in situ observations." As can be seen from Figs. 3 and 10, high concentrations of Nuf exit in limited areas and are essentially zero in many areas. If the MAIA model allows the dilution (say by a factor of 10 or more) of freshly nucleated particles (Nuf), the simulated Nuf at t=0 in Fig. 6 may be much smaller than the observed values.

2. Figure 6. Based on modeling results, most of Nuf at t=0 were formed ~2-6 days ago. Can you still called these particles freshly nucleated? In discussing Fig. 2, the authors mentioned that many nucleation events were observed over periods of 15 min or less. If these Nuf particles were formed 2-6 days ago, why they didn't spread to more wide regions? In a few days, these particles might have been transported far away from cloud outflow regions. Base on Figs. 3 and 10, high concentrations of Nuf are limited to the cloud outflow regions. Could these observations suggest the observed Nuf (Figs. 3, 6, and 10) were formed on the day of the measurements instead of being formed 2-6 days ago somewhere else)?

3. Figure 6. The influences of initial SO2 concentrations and surface areas on the predicted Nuf are presented. Both SO2 concentrations and surface areas affect new particle formation by controlling sulfuric acid vapor concentrations ([H2SO4]). [H2SO4] is a more direct and important parameter. In addition, it is hard to infer the nucleation rates from Fig.6. I would suggest the time series of modeled [H2SO4] along with nucleation rates be presented in an additional figure or in supplementary materials. Such additional information will be very useful to the interested readers.

4. p9278, lines 14-15. Why neutral nucleation dominates at night? What is [H2SO4] at the night? How do you separate ion versus neutral nucleation? Provide time series of [H2SO4] and nucleation rates (both ion and neutral) will help to clarify the picture.

ACPD
11, C3428–C3430, 2011

5. p9281, line 23. How do you maintain the constant surface area densities? The nucleated particles coagulate and grow and will contribute to the surface area densities. Since MAIA is a size resolved and kinetic model, it will be useful if the authors can provide a figure showing the evolution of particle size distribution (dN/dlogDp versus time) for a representative case.

6. Section 3.3. Please give a brief description of size ranges and number of bins used in the model.

Minor comments:

1. p9252, lines 8-11. Not up to date and might be not true anymore. Should check and discuss more recent global NPF studies using various schemes for example: Yu, F., G. Luo , T. Bates , B. Anderson , A. Clarke , V. Kapustin , R. Yantosca , Y. Wang , S. Wu: Spatial distributions of particle number concentrations in the global troposphere: Simulations, observations, and implications for nucleation mechanisms, J. Geophys. Res., 115, D17205, doi:10.1029/2009JD013473, 2010.

2. p9252, line 20. Why at ocean surface? DMS is emitted from ocean and then gradually oxidized to become SO2.

3. p9256, lines 9-11. Should provide some representative references for this statement.

4. p9279, last paragraph. The picture will be clearer if the [H2SO4] values were given.

5. p9286, line 4 and line 14. Not “recent new” or “freshly” based on Fig. 6 (formed 3-6 days ago).

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 9249, 2011.