

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

J. R. Peter et al.

Received and published: 6 March 2007

We would like to thank the reviewer for their constructive comments of our manuscript. Below are our responses to the comments, and details of changes we have implemented.

1.1

The question as to how we how we constructed the $CN_{predicted}/CN_{observed}$ ratio was also raised by Referee 2 and has been answered in point 2.9 in response to Anonymous Referee #2.

We have added the following paragraph,

“Since we only have two size “bins,” provided by the differential concentration of the CN counters and the ASASP, it was necessary to make assumptions about the form of the

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

aerosol number size distribution $n_a(r_a, t)$. We assumed the aerosol size distribution to be monomodal and its number concentration specified by whether it was in the size range of the first size bin (determined by the differential concentration of the TSI-3025 and TSI-3010) or the second size bin (determined by the differential concentration of the TSI-3010 and the ASASP). In effect, this specifies the maximum and minimum particles that could be left in a size bin. If all particles are considered to be of the size specified by the left hand size of the size bin (i.e. their smallest possible value) then they will have the largest possible diffusion coefficient and be subject to the largest possible scavenging. Conversely, if all particles are the size specified by the upper limit of the size bin, then the particles will have the smallest diffusion coefficient and be subject to the least amount of scavenging.”

The reviewer also asks how the calculations, which show that most particles of radius less than 50 nm should be scavenged after an in cloud residence time of about 30 minutes, can be reconciled with observations that show a consistent Aitken mode of typically 25 nm? The only way to reconcile these points is if there is a continual source of Aitken mode particles. We have attempted to show in the paper that clouds may provide this source. We have shown that there may have been a source of new particles within the cumulus, however, the same process may occur within stratocumulus clouds. A sampling strategy to measure the possibility of new particle formation within stratocumulus clouds is more problematic than for cumulus clouds due to the dynamic differences that exist between stratocumulus and cumulus clouds. As a result, it is not possible to construct a mixing diagram for stratocumulus, such as Fig. 1, on which the subsequent analysis is reliant. This is a topic that requires further investigation.

The reviewer asks for parameters used in the model. We have stated all the necessary equations (Eqs. (5)-(10)). The only other information that we did not supply was the assumed density of the particles, for which we used the density of ammonium sulfate. Assuming the composition of the aerosol, especially since we are concerned with fine mode particles, to be ammonium sulfate is perfectly reasonable. Modification of the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

particle density has no impact on the conclusions for all realistic values of the density. We also assumed an accommodation coefficient of unity.

The following has been added to the text (p. 14, line 3),

Equations 9 and 10 were solved with the following assumptions: (1) the aerosol particles were composed of ammonium sulfate; (2) once a CN encountered the surface of a droplet its probability of sticking was unity (i.e. an accommodation coefficient of one); (3) a stationary droplet size distribution.

1.2

The reviewer states that in-cloud, the sulfuric acid concentration would be quenched before being able to reach the necessary acid supersaturation required for new particle formation. We agree, and suggested that (as proposed by Hegg (1991) that the necessary sulfuric acid concentrations could be produced by the increased hydroxyl radical production occurring in (and near) cloud. This is entirely the same mechanism as previously proposed to explain the observed increase in new particle concentration encountered in cloud outflow, however, we suggest that the same mechanism could be occurring in cloud.

We do propose the increase in hydroxyl radical production as a mechanism for new particle formation within cloud on page 7485. The nucleation is favored inside cloud because the actinic flux will be greater there than outside of cloud. Furthermore, detailed quantum mechanical modelling studies Ianni (1999) and Ianni (2000) have shown that the spontaneous nucleation of new particles in the $\text{H}_2\text{SO}_4\text{—H}_2\text{O}$ system is thermodynamically unstable due to the high dehydration rate. Within cloud, where the environment is supersaturated, there is an abundance of water molecules and nucleation is energetically favourable.

The following text has been added (p. 16. 3rd paragraph)

Furthermore, detailed quantum mechanical modelling studies Ianni (1999) and Ianni

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

(2000) have shown that the spontaneous nucleation of new particles in the H_2SO_4 — H_2O system is thermodynamically unstable due to the high dehydration rate. Within cloud, where the environment is supersaturated, there is an abundance of water molecules and nucleation may be energetically favourable.

Hegg, D.: Particle production in clouds, *Geophys. Res. Lett.*, 18, 995–998, 1991.

Ianni, J. and Bandy, A.: A density functional theory study of the hydrates of $\text{NH}_3 \cdot \text{H}_2\text{SO}_4$ and its implication for the formation of new atmospheric particles, *J. Phys. Chem.*, 103, 2801–2811, 1999.

Ianni, J. and Bandy, A.: A theoretical study of the hydrates of $(\text{H}_2\text{SO}_4)_2$ and its implications for the formation of new atmospheric particles, *J. Mol. Struct.—THEOCHEM*, 497, 19–37, 2000.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 6, 7471, 2006.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)