Atmos. Chem. Phys. Discuss., 3, S1369–S1371, 2003
www.atmos-chem-phys.org/acpd/3/S1369/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Köhler theory for a polydisperse droplet population in the presence of a soluble trace gas, and an application to stratospheric STS droplet growth" *by* H. Kokkola et al.

Anonymous Referee #1

Received and published: 22 August 2003

The MS presents an interesting aspect of the microphysical behaviour of atmospheric aerosols. Kevin effect prevents the small particles to be activated at small cooling rates. The presence of trace gases may change the activation behaviour of aerosol particles. These phenomena are well described under tropospheric condition for mono-dispersed particles distribution (Laaksonen et al, 1998). The present MS applied this theory to polydispersed particle populations and also under non-equilibrium conditions.

The activation of CCNs is an important issue for the cloud properties (liquid cloud) in the troposphere. The influence of semi-soluble trace gases may change the number density of the liquid clouds and there their radiative and chemical properties.

However, the authors were not able to address this problem properly:

1) The first part of this MS (section 4-5) is devoted to tropospheric conditions. However, the readers can not get the information by how much the presence of the trace gases may change the cloud properties (see below).

2) The authors applied this model to a stratospheric case, leading to a wrong conclusion (see below).

Therefore, substantial modification of the MS is required for publication. The example shown here (STS in the stratosphere) is not justified. An application for tropospheric clouds which shows the effects of the trace gases would contribute the understanding of the cloud activation and make this MS acceptable for the publication in ACP.

Major comments:

1) Figure 5 shows the particle radius, above which they may activated. I suggest that the size bins without HNO3 should also be included in this figure. Then, the effect of trace gases will be better illustrated. 2) The second issue is the implication for the real atmosphere. By how much the cloud droplet number density would change in the presence of trace gas (closed system) at certain meteorological conditions? An concrete example is highly welcome at this point. 3) The authors concludes that the measured large HNO3-rich particles by Voigt et al (2000) could be liquid particles which have overcome the Köhler maxima. However, the agreement was achieved by using a cooling rate of 0.3/day = 0.0125 K /hr. The measurements of Voigt et al. were performed on January 25 2000 at downwind of a strong mountain wave activity. The cooling rates in such events are at least several Kelvin per hour (peak values of several 10 Kelvin per hour). The cooling rate used in the model simulation is 2-3 orders of magnitude to small. The observed PSC particles are formed clearly in the Lee wave events, which are ˜ 2 hours before the observations (airborne lidar measurements show no cloud upwind of the mountain wave). However, at a cooling rate used in model, a temperature decrease of 3 K (the transition from an background aerosol to STS) requires a cooling

ACPD

3, S1369–S1371, 2003

with constant rate for 10 days. That is also far from the observation (Voigt et al., 2000). With the cooling rate in typical Lee waves (several - several 10 K / hr), nearly 100 % of the aerosol particles will be activated, i.e., no splitting of the size distribution can be obtained, in contrast to the conclusion of the present MS. Also it is mentioned by the authors that the particles observed by Voigt et al. shows depolarisation, indicating solid particles. In summary, the large particles observed by Voigt at el. can not explained by liquid particles due to preferential growth of large particles. I doubt that such small cooling rate 0.3/day (including adiabatic process) exists in the atmosphere over tens of days at all in the Arctic stratosphere.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3241, 2003.