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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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.

H. Kokkola et al.

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We thank the referee for the comments, and believe that our revisions due to the comments will improve the paper.

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).

   - Although the conditions at which the calculations were done correspond to the troposphere, the purpose of Sections 4 and 5 is not to explore how tropospheric cloud properties may change, but to give a general description of the effect of a soluble trace gas on the equilibrium and non-equilibrium evolution of a polydisperse droplet population as a function of RH. The effects of HNO_3 and ammonia on cloud properties have been explored in a number of papers by Kulmala and coworkers e.g. Kulmala et al. (1993), Laaksonen et al. (1997) and Kulmala et al. (1998), and by Hegg (2000).

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 to the understanding of the cloud activation and make this MS acceptable for the publication in ACP.

– With tropospheric clouds, cooling rates are usually too fast for the splitting of the size distribution described in this MS to have any effect on the activation. Rather, the enhanced activation considered by Kulmala et al. (1993), Laaksonen et al. (1997), Kulmala et al. (1998), and Hegg (2000) can be understood by noting that smaller droplets with higher surface-to-volume ratio collect relatively more soluble gases during the limited activation time than do larger droplets, enhancing effectively their water-soluble mass. This leads to the enhanced activation.

There are two atmospheric processes that come into mind regarding the near-equilibrium splitting caused by soluble gases. These are radiation fog formation under polluted conditions, and STS droplet growth under conditions of seasonal cooling. We chose to study the latter in the present MS because it is a "cleaner" example in the sense that in the stratosphere, there is only one soluble gas (HNO_3) condensing, which is probably not the case with most pollution fogs. We believe that the example we have chosen is justified (see below).

Major comments:

1) Figure 5 shows the particle radius, above which they may activated. I suggest that the size bins without HNO_3 should also be included in this figure. Then, the effect of trace gases will be better illustrated.

   – Figure 5. shows the size class under which the gap appears in the size distribution below 100 % RH. Particles above this size class are not still not yet activated (in the traditional sense). This gap will not appear in the size distribution under 100 % without HNO_3 present in the system.

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.

   – The effect of trace gases on droplet activation has been previously studied (see above). In this manuscript we are trying to decribe the exact mechanisms concerning the effect of semi-volatile species on polydisperse droplet populations.

3) The authors concludes that the measured large HNO_3-rich particles by Voigt et al (2000) could be liquid particles which have overcome the Khler 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).

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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 up-wind 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 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.

– In hindsight, we admit that this part of the manuscript was not well written. We are not trying to say that we think that the particles observed by Voigt et al. (2000) were in the liquid state, although Fig. 8 and the last sentence of the manuscript may create such an impression. Rather, the point we want to make with the comparison is that the splitting of the size distribution may have occurred already before the mountain wave, and that the freezing to NAT may then have been favored in the larger mode (thus the sentence on p. 3253 "The calculations show that under certain conditions the liquid phase molar ratio of H_2O:HNO_3 decreases close to 3 and this could indicate that such droplets can form NAT). The seasonal cooling rates measured in the Arctic stratosphere can be extremely low Tabazadeh et al. (2000) and in the simulation presented in the paper, the splitting occurs already after 5 days. During the splitting, the H_2O:HNO_3 molar ratio is at its lowest and after

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that the droplets are diluted with respect to HNO_3. Our calculations do not remove the possibility that the bimodality of the size distribution measured by Voigt et al. were caused by the freezing itself, however, they offer an alternative explanation. We will rewrite Sections 6-8 to make these points clearer.

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