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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.

Anonymous Referee #2

Received and published: 13 August 2003

Review of the paper: Köhler theory for polydisperse droplet population... by Harri Kokkola et al, MS-NR: 2003-045

General:

The paper gives a comprehensive theoretical description of the behavior of a polydisperse aqueous droplet population in open and closed binary systems (H20 + HNO3). The most interesting and new property of such systems is their ability to split their size distribution for sufficiently high RH (but still smaller than 100%) and high partial pressures of HNO3. The presented paper is easy to read and the figures are all appropriate

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Discussion Paper

and clear. In particular the sections 1-5 make an important contribution to understanding of condensation processes on polydisperse droplets population in closed systems and should be published in ACP, even if some specific comments need some clarification (see below).

However, beside a small number of minor points, I feel that there are 2 serious critical points in the application of this theory on the condensation processes in the stratosphere:

- The main criticism is based on the fact that growth of NAT and STS aerosols in the stratosphere, in particular of large particles, is mainly determined by sedimentation processes which are completely neglected in the presented paper. If sedimentation processes are taken into account, then closed systems with limited amount of HNO3 and water may behave like open system because falling particles (e.g. NAT rocks) experience continuously changing ambient conditions. Thus, probably not (only) the splitting of the distribution function but much more selective falling of the aerosols causes the formation of the bimodal size distribution.
- The theory presented in sections 1-5, in particular the approximations (3) and (4) are valid only for high relative humidities. The typical conditions in the stratosphere are different, mostly with moderate relative humidities. Here the question arises if the presented theory (based on equation (3) and (4)) is still applicable.

To discuss these points I expect that some additional comments should be made and, presumably, parts of the manuscript have to be rewritten.

Specific comments:

- page 3, par 2: “In the closed system ....”
  Intuitively, one expects that the equilibrium saturation ratio derived for a closed

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system approaches the traditional Köhler equation if the number density of droplets decreases (i.e. every droplet contains a sufficiently amount of HNO3 that can condensate on it). Thus, it is not clear why a closed system should behave as an open system if the droplet size growths.

- page 3, par 2: "...the closed system Köhler curve crosses a succession of open system curves corresponding to a smaller and smaller concentration. For droplets larger than about 1 mi the crossing points reside on the unstable (decreasing) side of the open system curves, however, the resulting closed system Köhler curve represents stable equilibrium"

  This sentence needs a little bit more explanation. One method to understand this effect is to consider the Helmholtz free energy ΔF for open and closed systems, see e.g. Konopka and Vogelsberger, JGR, 1997, Vol. 102., p. 16057

- page 4, par 2 and 3: Explanation of the splitting effect in equilibrium
  These 2 paragraphs are not the explanation but rather a description of the splitting effect. To explain this effect (at least in equilibrium) one needs an energetical discussion (e.g. in terms of the Helmholtz free energy ΔF) answering the question why the splitted distribution is more favorable than an unsplitted one. Even if a complete energetical discussion is not possible in this paper, some remarks are desirable.

- page 7, par 1: "fairly high number density"
  here some numbers should be given

- page 4 first par in section 5
  replace "considered" through "considered as"

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

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