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Köhler theory for a polydisperse droplet population in the presence of a soluble trace gas, and an application to stratospheric STS droplet growth

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Abstract. We consider the equilibrium behavior of a polydisperse aqueous droplet population as a function of relative humidity (RH) when a soluble trace gas, such as nitric acid, is present in the system. The droplet population experiences a splitting when the RH is increased sufficiently. This splitting is not related to the traditional Köhler activation of cloud droplets, as it may occur at relative humidities below 100%. Remarkably, the splitting always takes place in such a way that the largest size class of the (discretized) droplet population starts taking up the soluble trace gas efficiently, growing steeply as a function of RH, and forcing the smaller droplets to shrink. We consider this behavior in terms of open and closed system Köhler curves (open system referring to one in which the trace gas concentration remains constant and closed system to one in which the gas concentration decreases as a result of uptake of the trace gas). We show how the open and closed system Köhler curves are related, and that the splitting of the population can be explained in terms of closed system curves crossing the Köhler maxima of the open system curves. We then go on to consider time-dependent situations, and show that due to gas-phase mass transfer limitations, the splitting of the size distributions moves toward smaller sizes as the rate of RH increase becomes more rapid. Finally, we consider stratospheric supercooled ternary solution droplet populations, and show that the splitting described using the new theory may lead to formation of bimodal size distributions in the stratosphere.

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