



Evaporation of cloud particles: solutions to the population balance equation and formulation of memory based bulk schemes (MBBS, v1.0)

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Abstract. One of the main cloud processes is the annihilation of particles by evaporation. If the saturation ratio drops below the equilibrium level, water evaporates and diffusion transports the water vapor through the environment of the particles. While other cloud processes are investigated carefully, evaporation is somewhat neglected, and the process' formulation in models is quite simplified. In this contribution, we investigate evaporation using the population balance equation (PBE) for mass distributions. A simplified regime is chosen which allows analytical treatment of the evaporation process. The theoretical results justify the inclusion of memory variables in evaporation modeling. We develop a family of Memory-Based Bulk Schemes (MBBS) that improve the representation of number concentration loss during evaporation by tracking memory variables of the particle mass distribution. We focus on number concentration because, unlike the mass concentration, its evaporation rate depends on a point-wise value of the distribution and therefore cannot be captured reliably by standard moment-based closures. Comparisons against the full PBE solutions for idealized downdraft and oscillatory scenarios show that the higher-complexity MBBS variants, in particular the two-memory-variable scheme, substantially improve the representation of number concentration during evaporation.

1 Introduction

Evaporation is a key process for the evolution of clouds, the annihilation of particles at subsaturated conditions is crucial for the macroscopic structure of clouds as well as for the interaction with radiation. While water particles grow by diffusion of water vapor in air under conditions above equilibrium (partial vapor pressure is larger than the saturation vapor pressure of the respective stable phase) evaporation is the complementary process with phase transition. The water molecules as released to the gas phase are transported to the environment by diffusion. Thus, a single particle is shrinking until it is completely evaporated. There might be some issues for applying the continuous diffusion equation for very small particles, since for a cluster of few molecules the continuous approach of diffusion cannot be applied in a strict sense. However, in practice the loss of mass can be described quite well with this approach, which is also mathematically well-posed (Baumgartner and Spichtinger, 2018a). For an ensemble of particles, the treatment of evaporation is more challenging: For huge ensembles of water particles, it is not possible anymore to treat individual particles, thus we have to use statistical approaches, which in turn translate into the representation of a cloud in respective (mathematical) models on distinct scales. On scales with cloud volumes large enough



for a description with a size or mass distribution, assuming a large and homogeneously distributed population of particles, the evolution of the cloud can be determined by a Boltzmann-type equation, the so-called population balance equation (PBE). In such a formulation, evaporation is treated as a transport in the mass space (i.e. the internal coordinate of the distribution); the loss of particle is given by the boundary condition at $m = 0$. While the time evolution of a size or mass distribution would be the appropriate representation of a cloud with homogeneously distributed particles, many weather and climate models use a representation by general moments of such distributions, with equations formally derived from the PBE; usually, equations for the zeroth moment (number concentration n) and the first moment (mass concentration q) of the mass distributions are used. This type of cloud schemes is often called bulk scheme. While for higher moments the forcing terms can be expressed by functions of moments (mostly power laws) this is generally not possible for the zeroth moment. By definition, the loss of particles (i.e. the decrease in the number concentration) is a boundary condition, which depends on the distribution itself rather than on the integrated properties (as moments). Thus, the evaporation of number concentration cannot be treated correctly in bulk models, if the information about the lower boundary of the distribution is not taken into account. Similar results were already stated in the study by Gierens and Bretl (2009).

In standard bulk models we find different approaches to treat the loss of number concentration in terms of function of the available cloud variables (e.g. number and mass concentration, respectively). We find two typical and rather extreme cases.

The first approach, mostly used for liquid clouds, leaves the number concentration unchanged during the evaporation process ($\frac{dn}{dt} = 0$), with changes of the total mass due to the growth law by diffusion. Once, the mass concentration falls below a certain threshold, the cloud variables are set to zero, i.e. $n = q = 0$. This procedure can be motivated or justified by the assumption of a homogeneously mixed cloud with a quite narrow cloud droplet distribution. The evaporation process shifts the distribution in mass space towards the boundary $m = 0$. If the distribution is quite narrow, nothing happens until the distribution meets the boundary in mass space $m = 0$; then the distribution is shifted over the boundary very fast, so this instantaneous switching to "no cloud" is meaningful or at least explainable.

In the second approach it is assumed that the loss rate of number concentration due to evaporation is somewhat proportional to the loss rate of the mass concentration. Thus, we end with a rate of evaporation

$$\left. \frac{dn}{dt} \right|_{\text{evaporation}} = \frac{n}{q} \left. \frac{dq}{dt} \right|_{\text{evaporation}}, \quad (1)$$

see, e.g., model descriptions in Morrison and Gettelman (2008) and Morrison et al. (2005) and others. There are some generalizations of this general concept, taking into account that due to the shift towards the boundary we lose first small particles (see, e.g., discussions in Harrington et al., 1995; Spichtinger and Gierens, 2009; Gierens and Bretl, 2009); however, the general concept of describing the number concentration rate by functions of moments only remains the same.

In both cases, the number concentration of the cloud particles is treated with systematic errors (or biases). In the first case, there is an overestimation of the number of cloud particles, while in the second case, the number is underestimated. This could have severe impact, e.g., on the radiation, leading to too many (and too small) cloud particles or even too few (and too large) cloud particles, respectively. This issue is maybe also critical for the investigations of tropical anvil clouds in the climate context (e.g. Gasparini et al., 2019), since it might change lifetime and microphysical properties of thin clouds.



In this study we give a physically correct and consistent description of evaporation as treated by the PBE (Sec. 2); actually,
60 we can derive analytical solutions, going beyond the investigations by Gierens and Bretl (2009), since we can treat a fully
coupled system with time dependent forcings. We further investigate evaporation in bulk model schemes in Sec. 3, since these
are commonly used in many different models on various scales. Starting from the consistent PBE model and the benchmark so-
lutions, we investigate the question if and how some of the features of the correct treatment of evaporation can be implemented
into existing bulk model schemes. This leads naturally to the formulation of the Memory-Based Bulk Schemes (MBBS), which
65 are able to treat evaporation of cloud particles in a physically meaningful way by incorporating memory of the evolving par-
ticle size distribution. We show the performance of the different MBBS variants for various idealized scenarios in Sec. 4. We
conclude our investigation in Sec. 5 with a short discussion and an outlook to further applications, both for the analytical PBE
approach and for the MBBS.



2 Population balance equation

70 We consider an arbitrary conservation law for an extensive physical quantity U with a density u

$$\partial_t u + \nabla \cdot \mathbf{J}_u = h_u \quad . \quad (2)$$

where J_u represents the flux density of this variable in an arbitrary coordinate space ξ ; h_u denotes the sum of sources and sink terms. In a purely deterministic approach the flux in the conservation law is given by

$$\mathbf{J}_u = \frac{d\xi}{dt} u. \quad (3)$$

75 As already discussed by Hulburt and Katz (1964) when studying the dynamical evolution of particle populations it is useful to define such a conservation law for the number of particles, i.e., u is set to be a density of particles in the phase space ξ . This set of coordinates can be split into external $\mathbf{x}$ and internal $\mathbf{r}$ coordinates such that $\xi = [\mathbf{x}, \mathbf{r}]$. External coordinates represent the position of the particle population whereas internal coordinates characterize the properties of the particles in a polydisperse population. Usual choices for internal coordinates are mass m , temperature T , velocity v , chemical composition ν etc.

80 In cloud hydrometeor modeling we consider the particle size distribution (PSD) f normalized to number concentration hydrometeors per kilogram of dry air. Therefore, we set $u = \rho f$ with ρ the density of the dry air. We also consider the mass of the hydrometeors to be the only relevant internal coordinate $\mathbf{r} \rightarrow m$, i.e., we rather consider a particle mass distribution. The function $f(t, \mathbf{x}, m)$ is defined for m in the open interval $(0, \infty) \subset \mathbb{R}$. The resulting conservation law for the PSD is

$$\frac{\partial(\rho f)}{\partial t} + \nabla_x(\mathbf{v} \rho f) + \frac{\partial}{\partial m} \left(\frac{dm}{dt} \rho f \right) = h_{\rho f} \quad (4)$$

85 which is commonly known as the population balance equation (PBE) for cloud hydrometeors. The $h_{\rho f}$ term usually represents nucleation and/or processes such as collisions and breakage of hydrometeors. The distribution f is normalized by the total number concentration of particles n . As for usual probability density functions we can formulate the general moments of the distribution as

$$\mu_k(t, \mathbf{x}) := \int_0^\infty m^k f(t, \mathbf{x}, m) dm \quad (5)$$

90 for a real number $k \in \mathbb{R}$, if the integral is converging. Note, that we integrate over positive masses only. Using this definition, we can relate the total number and mass concentrations to the general moments, i.e.

$$n(t, \mathbf{x}) = \mu_0(t, \mathbf{x}) = \int_0^\infty f(t, \mathbf{x}, m) dm, \quad q(t, \mathbf{x}) = \mu_1(t, \mathbf{x}) = \int_0^\infty m f(t, \mathbf{x}, m) dm \quad (6)$$

2.1 Diffusive Regime

We study a particular regime of the PBE on which the diffusional interaction between the hydrometeors and the surrounding
 95 vapor field is the only relevant process. This allows to have an analytically tractable evaporation process.



To do so we work in the frame of reference of the air parcel, effectively adopting a Lagrangian perspective. Generally, we would expect motion of hydrometeors relative to the center of gravity of the parcel. However, since we are interested in the evaporation regime only, we neglect sedimentation of particles; this is a meaningful approximation as sedimentation is relevant for large hydrometeors for which evaporation is not. Nucleation and collisions terms are also neglected, i.e., we set $h_{\rho f} = 0$.

100 The resulting PBE describes the dynamics in a zeroth dimensional box model:

$$\partial_t f + \partial_m \left(\frac{dm}{dt} f \right) = 0 \quad (7)$$

The continuity equation for the density for dry air

$$\partial_t \rho + \nabla_x (\mathbf{v} \rho) = 0 \quad (8)$$

has also been used to derive the PBE in the diffusive regime (Eq. 7). We use Maxwell's law for diffusional growth to set an
 105 advection velocity in mass space

$$\frac{dm}{dt} = C(S - 1)m^{1/3} \quad (9)$$

with $C = C(T, p)$ a thermodynamic function which acts as an effective diffusive constant dependent on the atmospheric conditions, and S the saturation ratio for the considered hydrometeor type over a flat surface of the respective stable water phase. The definitions of these thermodynamic functions can be found in Appendix A. The derivation of Maxwell's growth law can be
 110 found in wide variety of atmospheric physics textbooks (e.g., Lamb and Verlinde, 2011, chapter 8). Its basic assumptions are the spherical symmetry of the hydrometeors, which is somewhat less suitable for the ice phase, steady state profiles for the temperature and water vapor density fields around the hydrometeors (i.e. $T(t, \mathbf{x}) = T(\mathbf{x})$, $\rho_v(t, \mathbf{x}) = \rho_v(\mathbf{x})$), and equilibrium condition between the bulk phase of the hydrometeors and its surrounding vapor, which allows the use of the Clausius-Clapeyron equation at all times. In this study we also omit ventilation and kinetic corrections in the growth term in order to keep the solutions
 115 suitable for analytical studies.

With the mass advection "velocity" $\frac{dm}{dt}$ specified, the PBE to solve in the diffusive regime is

$$\partial_t f + \partial_m \left(C(S - 1)m^{1/3} f \right) = 0, \quad f(0, m) = f_0(m), \quad f(t, 0) = f(t, \infty) = 0, \quad (10)$$

where initial and boundary conditions have been stated. The solution of this PBE can be found analytically using the method of characteristics for an arbitrary prescribed $S = S(t)$. The detailed step by step calculation can be found in the Appendix B,
 120 using a general growth exponent $\dot{m} \sim m^b$ with $b < 1$. In the main study, we restrict ourselves to spherical hydrometeors, i.e. $b = \frac{1}{3}$.

For the explicit solution, we have to introduce the functions

$$I(t) := \int_0^t (S(\tau) - 1) d\tau, \quad I_{min}(t) := \min_{0 \leq \tau \leq t} I(\tau). \quad (11)$$

The function $I(t)$ will be referred to as the saturation memory. The function $I_{min}(t)$ is defined as the minimum value of $I(t)$,
 125 considering all of the previous times $0 \leq \tau \leq t$. By definition, this function has the property $I_{min}(t) \leq 0, \forall t$.



Depending on the prescribed saturation ratio profile $S(t)$, the ODE in Equation (9) can have multiple solution for a given initial condition since $m^{\frac{1}{3}}$ is not locally Lipschitz continuous near $m = 0$. Physically, however, we have an additional condition: Once a hydrometeor has completely evaporated, i.e., it reached mass $m = 0$, it cannot grow again to positive masses just by a small increase in S (instead one really needs a nucleation event with much higher saturation ratios, which we neglect here).
 130 This translates into the condition that if $m(t^*) = 0$ for some $t^* \geq 0$, then $m(t) = 0$ for all $t \geq t^*$. Under this additional physical condition, the ODE from Eq. (9) with initial condition $m(0) = 0$ does have a unique solution for all positive times $t > 0$ and this solution is given by

$$m(t) = \begin{cases} \left(m_0^{2/3} + \frac{2}{3}CI(t)\right)^{3/2} & \text{if } m_0^{2/3} + \frac{2}{3}CI_{min}(t) > 0 \\ 0 & \text{otherwise,} \end{cases} \quad (12)$$

where the condition $m_0^{2/3} + \frac{2}{3}CI_{min}(t) > 0$ ensures that $m(\tau) \neq 0$ at all previous times $0 \leq \tau \leq t$. Explicitly, the largest initial
 135 mass $m_{0,*}$ that has evaporated until time t can be tracked by the minimum value $I_{min}(t)$ of the saturation memory by

$$m_{0,*}(t) := \left(-\frac{2}{3}CI_{min}(t)\right)^{3/2}. \quad (13)$$

With the characteristic curves given as Eq. (12), the formal solution for the mass distribution satisfies

$$m^{1/3}f(t, m) = m_0^{1/3}f_0(m_0) \Leftrightarrow f(t, m) = \left(\frac{m_0}{m}\right)^{1/3}f_0(m_0) \quad (14)$$

for the initial condition $f(0, m) = f_0(m)$. Following the characteristics with initial value m_0 we obtain a valid mass $m > 0$
 140 only if we start with an initial value $m_0 > m_{0,*}$; otherwise we end in $m = 0$ at a certain time $\tau < t$, and the expected number of particles should be zero (i.e. $f(\tau, 0) = 0$). This condition $m_0^{2/3} > m_{0,*}^{2/3} = -\frac{2}{3}CI_{min}(t)$ results into the condition

$$m^{2/3} = m_0^{2/3} + \frac{2}{3}CI(t) > m_{0,*}^{2/3} + \frac{2}{3}CI(t) = -\frac{2}{3}CI_{min}(t) + \frac{2}{3}CI(t) = \frac{2}{3}C(I(t) - I_{min}(t)) \quad (15)$$

for a valid mass coordinate of the mass distribution. Thus, we can formulate the resulting PSD for the hydrometeors in the diffusive regime as

$$145 \quad f(t, m) = \begin{cases} m^{-1/3} \left(m^{2/3} - \frac{2}{3}CI(t)\right)^{1/2} f_0\left(\left(m^{2/3} - \frac{2}{3}CI(t)\right)^{3/2}\right) & m^{2/3} > \frac{2}{3}C(I(t) - I_{min}(t)) \\ 0 & m^{2/3} \leq \frac{2}{3}C(I(t) - I_{min}(t)) \end{cases} \quad (16)$$

for the initial condition $f(0, m) = f_0(m)$.

To illustrate how the solutions look like we consider the simple case of a constant saturation ratio $S = S_0 = \text{const}$. This case was already studied by Gierens and Bretl (2009), where the change of shape of the distribution in supersaturated and subsaturated regimes was analyzed. In Fig. 1 we show an exemplary case for the evolution of the distribution $f(t, m)$ and the
 150 new introduced functions $I(t)$ and $I_{min}(t)$, respectively. The plots are shown in arbitrary units and the initial distribution is set to a lognormal distribution.

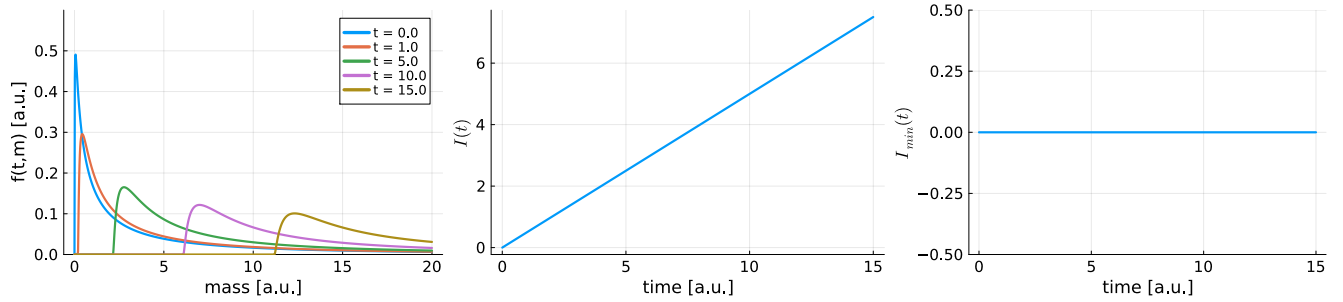


Figure 1. Snapshots of the distribution function $f(t, m)$ at certain times along with the time evolution of $I(t)$ and $I_{min}(t)$ for an idealized growth case, i.e. $S > 1$. The simulated regime consists of an initial lognormal distribution of hydrometeors experiencing a constant supersaturated environment. The units remain arbitrary. The advection of the mass distribution can be seen in the left panel, whereas the saturation memory is increasing (middle panel). There is no evaporation happening in this case, thus $I_{min}(t) = \text{const.} = 0$.

We see in this idealized test case the shifting of the distribution towards larger masses following the characteristic curves. Since the saturation ratio is constant, we simply have $I(t) = (S_0 - 1)t$ and $I_{min}(t) = 0$ which indicates that no hydrometeors have been annihilated by evaporation. To further illustrate the power of this exact solution we consider the more complex but
 155 still analytically tractable case of an oscillating saturation ratio, i.e. $S(t) = 1 + s_0 \cos(\omega t)$. Simple integration of this saturation ratio yields a saturation memory $I(t) = \frac{s_0}{\omega} \sin(\omega t)$. In Fig. 2 the time evolution of the mass distribution is represented. In addition, Fig. 3 shows the respective time evolutions of the normalized number concentration (relative to the initial value n_0), the saturation ratio S , the saturation memory $I(t)$ and its minimum value $I_{min}(t)$, respectively.

The oscillation of the distribution around the same mass values makes the visualization in this case a bit harder, therefore
 160 snapshots at different times of the mass distribution together with the initial distribution are shown in Fig. 2. Still, we can appreciate how initially the distribution moves to larger masses (e.g. until $t = \frac{\pi}{2\omega}$). At later times (e.g. until $t = \frac{3\pi}{2\omega}$) the mass is accumulating near the boundary $m = 0$ and evaporation is taking place. Finally, the distribution moves back again to larger masses (e.g. until $t = \frac{5\pi}{2\omega}$). This last time is particularly illustrative, as one can observe the cutoff in the distribution caused by the evaporation of hydrometeors. The location in mass space can be calculated with $I(t)$ and $I_{min}(t)$ by means of Eq. (13).

165 As stated previously, in this case the saturation memory $I(t)$ is simply given by a sine function. More interesting is the function $I_{min}(t)$, which can be calculated explicitly, yielding

$$I_{min}(t) = \begin{cases} 0 & \text{for } 0 \leq t < \frac{\pi}{\omega} \\ \frac{s_0}{\omega} \sin(\omega t) < 0 & \text{for } \frac{\pi}{\omega} \leq t < \frac{3\pi}{2\omega} \\ \frac{s_0}{\omega} \sin\left(\frac{3\pi}{2}\right) = -\frac{s_0}{\omega} & \text{for } t \geq \frac{3\pi}{2\omega} \end{cases} \quad (17)$$

Thus, mass is only evaporated in the interval $\frac{\pi}{\omega} \leq t < \frac{3\pi}{2\omega}$, where the function $I_{min}(t)$ decreases.

In the general case of a cloud, the saturation ratio is not simply a continuous function of time $S(t)$ but its dynamical evolution
 170 is usually coupled to the distribution function itself. For an air parcel, vertically moving with a velocity $w(t)$, a differential

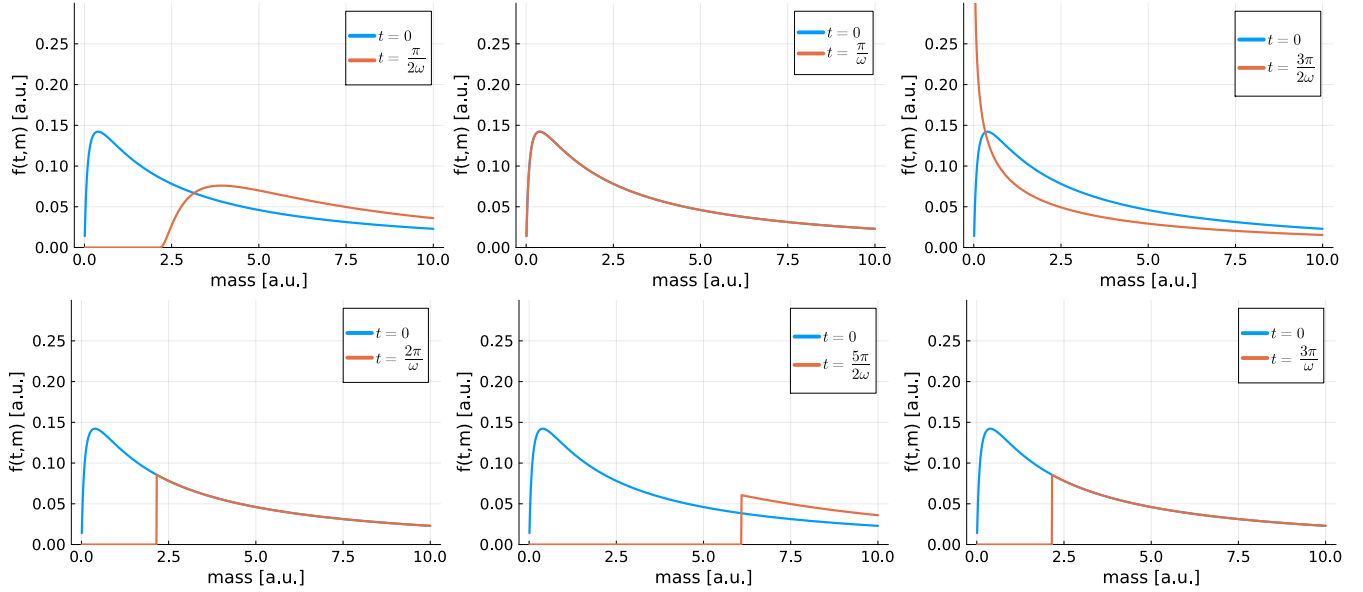


Figure 2. The time evolution of the mass distribution is shown in case of an oscillating saturation ratio. The blue curve indicates the initial distribution in all panels, the red curve displays the mass distribution at the respective time. The shift of the distribution beyond the boundary $m = 0$ leads to evaporation of particles, visible as a cutoff on the left hand side in the following evolution.

equation for the saturation ratio can be given by

$$\frac{dS}{dt} = AwS - B \frac{dq}{dt} \quad (18)$$

where q denotes the total cloud mass concentration, and $A = A(T, p)$ and $B = B(T, p, S)$ are thermodynamic functions that parametrize the strength of the adiabatic cooling/warming, and the strength of the hydrometeor-vapor field coupling, respectively. Their exact definitions along with the derivation of the differential equation can be found in the Appendix A. This differential equation includes the total mass concentration q . This means that the characteristic curves depend on a global property of the distribution rather than a point-wise value. There are some investigations about the existence, uniqueness and regularity of the weak solutions to such problems (Keimer and Pflug, 2017). We apply the general framework of the method of characteristics even for this very general case of a cloud in the spirit of the investigations by Keimer and Pflug (2017), extending previous analytical investigations (as, e.g., in Gierens and Bretl, 2009).

By inspection of Eq. (16) we see that the problem is fully solved by specifying an initial distribution of masses $f_0(m)$ and then obtaining $I(t)$ (and thus $I_{min}(t)$). The latter can be attained by using the relation between the saturation ratio and memory in Eq. (18) yielding

$$\frac{d^2 I}{dt^2} = Aw(\dot{I} + 1) - B \frac{dq}{dt}(I, \dot{I}, I_{min}). \quad (19)$$

This second order ODE must be solved numerically as no analytical solution exists for the general cases.

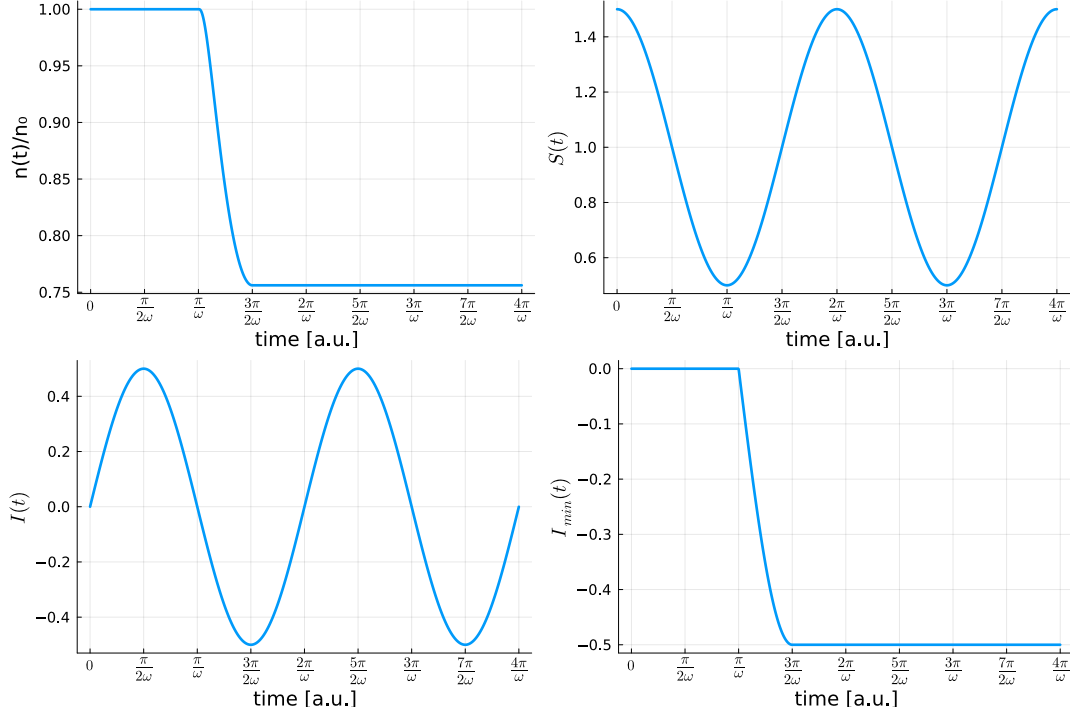


Figure 3. For the oscillating case the relevant variables number concentration n (relative to the initial value n_0), saturation ratio S , saturation memory I and its minimum I_{min} , respectively, are shown. The evaporation event takes place in the time interval $[\frac{\pi}{2\omega}, \frac{3\pi}{2\omega}]$ with $S < 1$, shifting the distribution towards smaller masses; however, the number concentration is only changed from $t = \frac{\pi}{\omega}$ on, since at this time the distribution reaches the boundary $m = 0$ first. This change in n can clearly be seen in the change of I_{min} during this time interval, indicating the annihilation of cloud particles.

If the vertical velocity of the air parcel is set as a constant, and also B is approximated as a function $B(T, p)$, a first integration of this ODE can be performed resulting in

$$\frac{dI}{dt} = S_0 - 1 + Aw(I + t) - B(q(I, I_{min}) - q_0) \quad (20)$$

reducing one order of the respective ODE. The exact definitions of $q(I, I_{min})$ and $\frac{dq}{dt}(I, \dot{I}, I_{min})$ can be found in the next section.

We acknowledge that the final calculation of the distribution, i.e., the solution of the PBE, relies on the numerical integration of an ODE. However, its functional form is completely explicit and the mathematical complexity is drastically reduced. Moreover, no closure approximations need to be performed as compared to usual methods of moments and the accuracy of the solution will solely depend on the ODE solver.



195 2.2 Moment transport in the diffusive regime

In this section we analyze how the bulk properties of the distribution i.e. its general moments as defined in Eq. (5) are transported in this dynamical regime. We focus mainly on the zeroth and first moments, i.e. number and mass concentration, respectively. The dynamical evolution of the k -th moment of the PSD can be obtained by integrating the mass power m^k over the PBE (7), i.e.,

$$200 \quad \frac{d}{dt} \int_0^\infty m^k f dm + \int_0^\infty m^k \partial_m \left(\frac{dm}{dt} f \right) = 0. \quad (21)$$

The first term is simply the time derivative of the integral moment $\mu_k(t)$. The second term can be rewritten using integration by parts, thus obtaining the following evolution equation for the k -th moment:

$$\frac{d\mu_k}{dt} = k \int_0^\infty m^{k-1} \frac{dm}{dt} f(t, m) dm - \lim_{m \rightarrow \infty} \left[m^k \frac{dm}{dt} f(t, m) \right] + \lim_{m \rightarrow 0^+} \left[m^k \frac{dm}{dt} f(t, m) \right]. \quad (22)$$

The first term in the right hand side relates the evolution of the k -th moment to integral properties of the PSD. The other two
 205 terms are related to fluxes across the outer boundary at $m \rightarrow \infty$ and the inner boundary at $m \rightarrow 0^+$. Usual mass advection rates have the shape $\frac{dm}{dt} \sim m^b$ which yields a null flux across the outer boundaries for distribution functions that fulfill the regularity condition

$$\lim_{m \rightarrow \infty} [m^p f(t, m)] = 0, \quad \forall p \in \mathbb{R}. \quad (23)$$

The usually chosen distributions for cloud hydrometeor modeling (e.g. gamma, lognormal etc.) fulfill this regularity condition.
 210 If any of these distributions is prescribed as the initial condition of the PBE, the transported distribution at time t will also fulfill the regularity condition. This can be seen by considering the distribution transport along the characteristics given by

$$m^{1/3} f(t, m) = m_0^{1/3} f_0(m_0). \quad (24)$$

The flux across the inner boundary represents the particles that leave the domain $(0, \infty)$ across the $m = 0$ value due to the continuous advection in mass space. In our problem this represents the annihilation of the hydrometeors due to the interaction
 215 with a subsaturated environment. It is worth to mention that we are not evaluating the value of the PSD at zero size as no masses of zero size can be found; the distributions are defined for m in the open interval $(0, \infty)$. We are considering the value of the flux across our absorbing boundary at zero size via the limit $m \rightarrow 0^+$ of some distribution property. These limits are key to calculate some bulk quantities, in fact if we look at the evolution of the number concentration ($\mu_0 := n$), we obtain

$$\frac{dn}{dt} = \lim_{m \rightarrow 0^+} \left[\frac{dm}{dt} f(t, m) \right] = \begin{cases} C(S-1) \left(-\frac{2}{3} CI(t) \right)^{1/2} f_0 \left(\left(-\frac{2}{3} CI(t) \right)^{3/2} \right) & I(t) = I_{min}(t) \\ 0 & I(t) > I_{min}(t) \end{cases} \quad (25)$$

220 We see that the evaporation of the number concentration of hydrometeors is exactly given by the number concentration flux $\frac{dm}{dt} f(t, m)$ across the inner boundary at $m = 0$, and **no integral property** of the distribution influences the rates. The fact of



defining the evaporation of the number of particles by a limit of the distribution function is a well known fact in the population balance community that can be found in the literature (Ramkrishna, 2000) as well as the in corresponding publications (Yuan et al., 2012) in order to treat evaporation and dissolution kinetics problems.

225 The ODE of the number concentration can be solved analytically, yielding the following closed form solution

$$n(t) = \int_{m_{0,*}(I_{min}(t))}^{\infty} f_0(m_0) dm_0 = n_0 - F_0(m_{0,*}(I_{min}(t))), \quad F_0(m_{0,*}(I_{min}(t))) = \int_0^{m_{0,*}(I_{min}(t))} f_0(m) dm, \quad (26)$$

with the largest evaporated mass $m_{0,*}(t)$ as defined in the previous section in Eq. (13). We can obtain the ODE for the mass concentration by setting $k = 1$ (while $\mu_1 = q$) in the moment transport equation (22), which yields

$$\frac{dq}{dt} = C(S-1) \int_0^{\infty} m^{1/3} f(t, m) dm = C\dot{I} \int_{m_{0,*}(I_{min}(t))}^{\infty} \left(m_0^{2/3} + \frac{2}{3}CI(t) \right)^{1/2} f_0(m_0) dm_0. \quad (27)$$

230 As we can see, the evolution of the mass concentration solely depends on integral properties of the distribution, and fluxes across boundaries have no contribution. The mass concentration can also be calculated at each time via the first moment of the distribution

$$q(t) = \int_{m_{0,*}(I_{min}(t))}^{\infty} \left(m_0^{2/3} + \frac{2}{3}CI(t) \right)^{3/2} f_0(m_0) dm_0 \quad (28)$$

using the full solution of Eq. (16).

235 Note that in our formulation of the system (PBE with coupled differential equation for saturation ratio), we set the environmental conditions to constant values p, T , i.e. the vertical motion of the parcel only affects the saturation ratio via the vertical velocity but there is no change in temperature and pressure, respectively. This approach was already used in former investigations (e.g. Spreitzer et al., 2017; Bergner and Spichtinger, 2026). The system could be extended to the more general case, using additional ODEs for the evolution of temperature and pressure, respectively. However, for our investigations we control
 240 the saturation ratio directly in order to determine changes for different models in a controlled environment. We will apply the same strategy for the bulk models as defined in the next section.



3 Bulk schemes

Now we investigate the formulation of cloud processes in so-called bulk models. These bulk models are in essence the formulation of a system of ODEs for certain bulk or averaged properties of the PSD as well as for the thermodynamic fields of interest, such as temperature T , pressure p , mass concentration of water vapor q_v etc. The ODEs for the bulk quantities are coupled to those of the thermodynamic fields via physical arguments.

The evolution equations of the bulk properties, i.e. the general moments μ_k , as defined in Eq. (5), are derived from the PBE as moment transport equations (see Sec. 2.2) up to a certain order K . Unfortunately, this mathematical machinery leads in general to a non-closed system of ODEs. As we have seen the evolution of $n = \mu_0$ depends on a point wise value of the distribution flux, and the mass concentration $q = \mu_1$ depends on a non-integer moment of the distribution $\dot{q} \propto \mu_{1/3}(t)$. Therefore a mathematical closure is always needed if a solvable bulk scheme is to be formulated. In practice, we close the system by prescribing a certain (mostly analytical) type for the mass/size distribution with general (analytical) expressions for general moments, and fixing certain parameters of the chosen functional type (e.g. lognormal or gamma distribution).

As we will see in the results section, the fact that the evolution of $q = \mu_1$ depends on an integral property of the distribution leads to a better agreement of standard closure approaches with the analytical solution of the PBE; this closure is explained in the first subsection. On contrary, the closure for the number concentration $n = \mu_0$ as an ODE in the subsaturated regime leads to more complications, and all of the newly proposed strategies try to approximate this boundary flux term in meaningful manner.

We want to remark here that the presented MBBS do not correspond to the "ultimate solution" for treating the evaporation term of the number concentration. Rather, they represent different strategies that can be taken into account for a better representation of the hydrometeor number, e.g., for determining radiative properties or other variables, sensitive to the total number of cloud particles. We leave it to the community to decide which MBBS variant is most appropriate depending on the desired regime or accuracy. We also encourage the community to try to develop more suitable parameterizations that are also consistent with the memory tracking analytical solutions.

All presented bulk schemes are formulated with some common properties, which makes it easier for comparing the respective behavior:

- The only tracked bulk quantities are mass and number concentration, and the environmental conditions are only represented by the saturation ratio S , mimicking similar studies performed in the past (e.g. Bergner and Spichtinger, 2026; Spreitzer et al., 2017; Wacker, 1995).
- The initial prescribed distribution is always considered to be a lognormal. This choice is motivated by the fact that improved bulk schemes are especially needed for treating ice clouds with slow sublimation processes; for ice clouds, lognormal distributions are a quite common choice (e.g. Schröder et al., 2000). As it will become clear, the newly proposed schemes can be equivalently formulated for another distribution type, e.g., (generalized) gamma distribution as it is also common, especially for liquid clouds (Seifert and Beheng, 2006).



- 275 – The simulations always include a subsaturated regime ($S < 1$), where evaporation of mass and/or number concentration may occur. As can be seen from literature, and also in our examples, bulk schemes work quite well for predicting the time evolution of the mass concentration q .

It follows from the solutions of the PBE, evaporation or sublimation of cloud particles is most important for small particles, i.e. distributions close to the boundary $m = 0$. Thus, in the following we will concentrate on the regimes of small particles.

280 3.1 Standard approaches for bulk models

First, we present the closure approximation to formulate an ODE for the mass concentration $q = \mu_1$. This standard calculation has been performed in many bulk models in the past (see, e.g., Spichtinger and Gierens, 2009). The moment transport equation yields an exact ODE for the mass concentration

$$\frac{dq}{dt} = C(S - 1) \int_0^\infty m^{1/3} f(t, m) dm = C(S - 1) \mu_{1/3}(t). \quad (29)$$

- 285 This equation is still exact, since the distribution can change its shape; the moment $\mu_{1/3}(t)$ can be calculated at each time t , if the integral converges. For the change to a bulk scheme, we make some closure assumptions. The closure approximation relies on considering that both, the distribution type and width, remain fixed during the time evolution of the cloud, whereas the mean mass $\bar{m} = \frac{q}{n}$ is allowed to change. As stated previously, we only consider evolutions of lognormal distributions throughout this paper. This closure approximation for the lognormal distribution results into

$$290 \quad \frac{dq}{dt} \approx C(S - 1) \int_0^\infty m^{1/3} f_{\log}(t, m) dm = r_0^{-1/9} C(S - 1) n \bar{m}^{1/3} = r_0^{-1/9} C(S - 1) n^{2/3} q^{1/3} \quad (30)$$

with the fixed parameter r_0 related to the fixed distribution width, as defined in the Appendix C. For determining the ODE for the number concentration $n = \mu_0$, we are facing the issue of being rather a boundary condition than an integral property.

- There are two general approaches for imposing a closure condition to the number concentration evaporation term in classical bulk models, at least to our knowledge. First, it is assumed that all particles just shrink, and there is no loss of particles until they reach a threshold mean mass (or the mass completely evaporates, i.e. $q = 0$). This hypothesis is sometimes called "homogeneous mixing of cloud particles" (see, e.g., Grabowski, 2007; Jarecka et al., 2009, 2013), and is rooted in turbulence research of liquid clouds. This approach is particularly suitable for modeling cloud droplets, as their distributions can be very narrow (e.g. Khain et al., 2015; Noppel et al., 2010), and, more important, diffusion is relatively fast in warm cloud environments ($T \sim 270 - 290$ K and $p \sim 800 - 1000$ hPa) due to larger diffusion coefficients and higher water vapor concentrations, respectively.

In Fig. 4 we show a simulation of the number and mass concentration of liquid cloud droplets at $T = 280$ K and $p = 800$ hPa, the initial cloud droplet distribution is set to a lognormal distribution with $m_m = 1.0 \times 10^{-12}$ kg and $\sigma_m = 2.0$ (corresponding to $r_0 \sim 1.62$), prescribing the initial number concentration as $N_0 = 100 \text{ cm}^{-3} = 10^5 \text{ L}^{-1}$. The initial saturation is set to equilibrium $S = 1$, the system experiences a slow downdraft of $w = -0.1 \text{ m s}^{-1}$. Since downdrafts in or around liquid

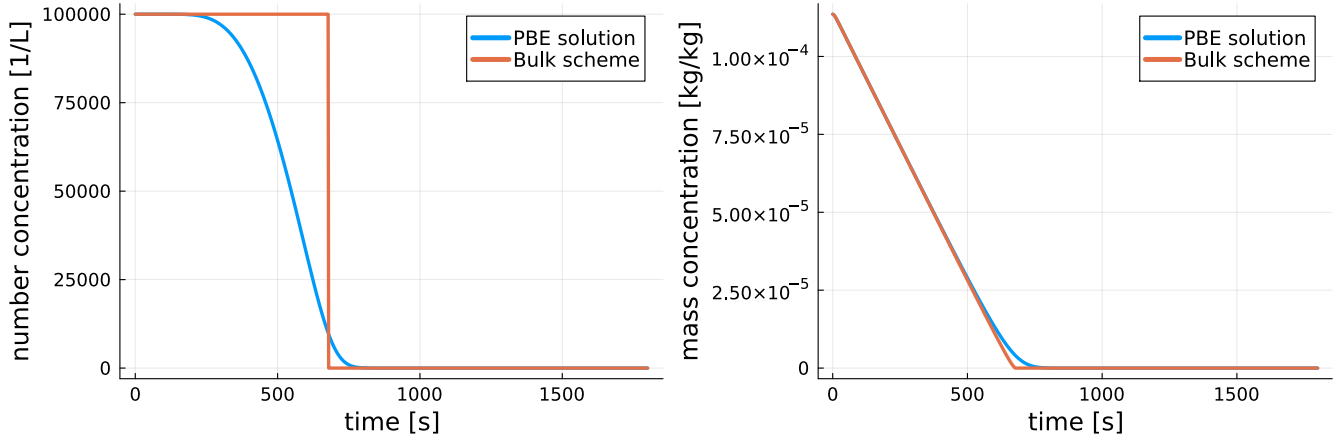


Figure 4. Comparison between the PBE solution and the two moment bulk scheme. The evaporation rate is chosen as a step function for the number concentration. A lognormal closure is set for the mass concentration. The environmental conditions are $T = 280$ K and $p = 800$ hPa. The simulation is performed for a downdraft on an air parcel with vertical velocity $w_0 = -0.1 \text{ m s}^{-1}$ which at first was at equilibrium with the environment $S(t=0) = 1.0$. The parameters of the chosen lognormal distribution are $\sigma_m = 2.0$ and $m_m = 1.0 \times 10^{-12}$ kg, the initial number concentration is set to $N_0 = 100 \text{ cm}^{-3} = 10^5 \text{ L}^{-1}$. While the agreement for the total mass is almost perfect, there are deviations between the solutions for the number concentrations.

clouds can be much faster (Berg et al., 2017; Hernández Pardo et al., 2025), this is a lower bound and constitutes the "worst case" scenario; for larger absolute values of w the evaporation is much faster.

Both, the results from the analytical solution of the PBE as well as the described closure approximations for the moments are shown in the two panels of the figure. The closure approximations for cloud droplets ("all or nothing") fit the analytical solution quite well due to the fast diffusion times, and narrow distributions. Note, that we chose one of the worst cases; if the distribution is more narrow and the downdraft is faster, the agreement becomes much better. However, even in this "worst" case, the "all or nothing" approach is a meaningful approximation for the evaporation of liquid cloud droplets. We omit the study of the raindrop evaporation as raindrop distribution dynamics are mainly influenced by collision and collection terms (e.g. Beheng, 2010), making the diffusive regime not a meaningful approximation.

For regimes with evaporation as a slow process, as e.g. in case of pure ice clouds at lower temperatures (Spichtinger and Gierens, 2009; Bergner and Spichtinger, 2026), this approximation might not be appropriate. Therefore, there must be used another closure approximation for the loss of total number concentration. Generally, one can assume that the loss of number concentration is somewhat proportional to the loss of total mass concentration. For simplicity this can be formulated as a discrete process (as it is treated in numerical models anyway): within one timestep Δt , there is a loss of mass Δq , which is related to the loss of number concentration via

$$\Delta n = n \cdot \left(\frac{\Delta q}{q} \right)^\alpha \quad (31)$$

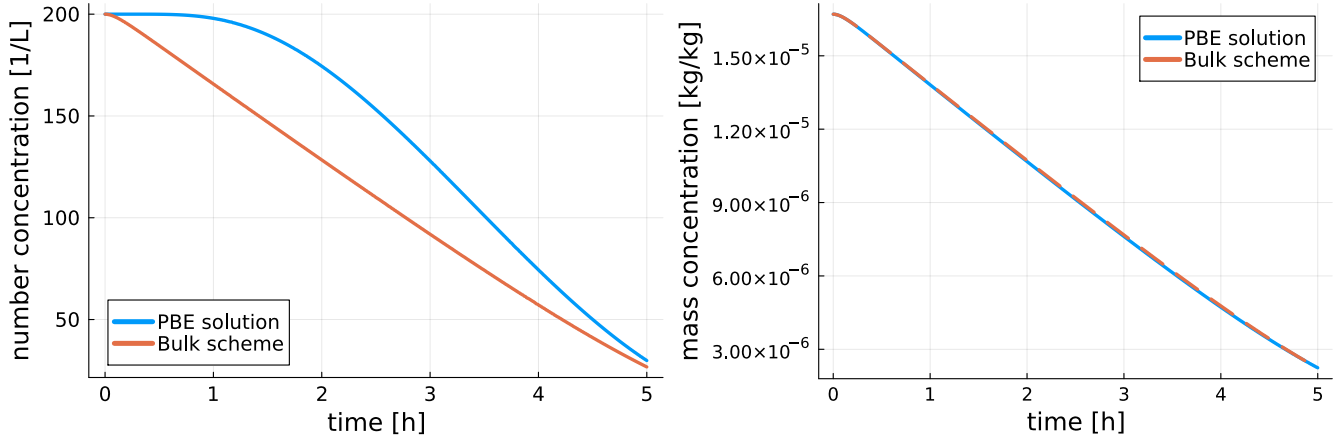


Figure 5. The time evolution of number concentration (left panel) and mass concentration (right panel) are shown for the PBE solution (blue) and the simulation using the classical bulk scheme (red) as represented in Eqs. (33)-(35). The environmental conditions are $T = 220$ K and $p = 200$ hPa. The simulation is performed for a downdraft on an air parcel with vertical velocity $w_0 = -0.01 \text{ m s}^{-1}$ which at first was at equilibrium with the environment $S(t=0) = 1.0$. The parameters of the chosen lognormal distribution are $\sigma_m \approx 2.85$ ($r_0 = 3.0$) and $m_m = 1.5 \times 10^{-11}$ kg, the initial number concentration is set to $N_0 = 0.2 \text{ cm}^{-3} = 200 \text{ L}^{-1}$. While the agreement for the mass concentration is (almost) perfect, there is a strong deviation of the bulk solution from the PBE solution. Even the slope of the curves is different, showing a linear decrease for the bulk model, whereas the PBE solution shows an s-shape behavior.

with an exponent $\alpha \geq 1$ (see Harrington et al., 1995; Spichtinger and Gierens, 2009), whereas the loss Δq is determined by the closure approximation as stated above, i.e. calculated by the integrated mass equation for $q = \mu_1$. An exponent $\alpha = 1$ leads to the common proportionality

$$\frac{dn}{dt} = \frac{n}{q} \frac{dq}{dt}, \quad (32)$$

325 which is used in many models, especially when treating sublimating ice clouds (see Gasparini et al., 2019; Morrison et al., 2005, and others). This relation implies a constant mean mass $\bar{m} = \frac{q}{n}$ during the evaporation process. These closure approximations result in the following system of ODEs

$$\dot{q} = r_0^{-1/9} C(S-1) n^{2/3} q^{1/3} \quad (33)$$

$$\dot{S} = AwS - Br_0^{-1/9} C(S-1) n^{2/3} q^{1/3} \quad (34)$$

$$330 \quad \dot{n} = r_0^{-1/9} C(S-1) n^{5/3} q^{-2/3}, \quad (35)$$

when treating evaporation explicitly. For the ODE determining the variable S we use the formulation of Eq. (18), inserting the explicit expression $\dot{q}$. The width of the lognormal distribution is represented by the constant r_0 .

In Fig. 5 the time evolution of the total number (left panel) and mass (right panel) concentration are represented, using the "standard" bulk scheme (red line) of eqs. (33)-(35) and the analytical solution of the PBE (blue line). The simulation is



performed at $T = 220\text{ K}$ and $p = 200\text{ hPa}$, the initial ice crystal distribution is set to a lognormal distribution with $m_m = 1.5 \times 10^{-11}\text{ kg}$ (corresponding to a mean radius $r_m = 20\text{ }\mu\text{m}$) and $\sigma_m \approx 2.85$ (corresponding to a width parameter $r_0 = 3.0$), with a total number concentration of $N_0 = 0.2\text{ cm}^{-3} = 200\text{ L}^{-1}$. Equivalently to the cloud droplet case the system is initialized at the equilibrium $S = 1.0$ and it is forced with a slow downdraft of $w_0 = -0.01\text{ ms}^{-1}$, which is a typical value for synoptic scale downdrafts (e.g. around high pressure systems, see Holton et al., 1995; Spichtinger et al., 2005). While the evolution of the total mass concentration of the bulk model is in perfect agreement with the PBE solution, for the total number concentration there is a huge deviation between both solutions. An important feature of the PBE, i.e., the delayed decrease of the number concentration, is completely missed by the bulk scheme. Actually, the number concentration is reduced as soon as the water vapor in the air parcel is subsaturated. However, for a distribution far away from the boundary $m = 0$, evaporation must first shift the distribution towards the boundary until there is loss of particles. This feature must be implemented into the bulk schemes for a more realistic treatment of evaporation.

3.2 General strategy for MBBS formulation

As we have seen, although the evaporation of liquid cloud droplets in standard bulk schemes via "all or nothing" formulations does not exactly match the analytical PBE solution, their discrepancies are less critical due to the fast diffusion timescales at warm temperatures. The same is not true for the ice particles, where diffusion is a slow process for temperatures way below the melting point. Thus, the upcoming discussion and bulk scheme modification will only concern the ice phase; one should have in mind the cold temperature regime $T < 240\text{ K}$, where liquid water cannot exist. Thus, we also exclude the mixed-phase from our investigations, because of the additional (e.g. Baumgartner and Spichtinger, 2018b). Nevertheless, the approaches presented here might be extended to mixed-phase clouds. Furthermore, since the closure approximation for the mass concentration agrees almost perfectly with the full PBE solution, modification will only be proposed for the number concentration. To describe the general strategy to modify the number concentration rate $\frac{dn}{dt}$ we first rewrite its analytical expression given by Eq. (25) as

$$\frac{dn}{dt} = -f_0(m_{0,*}(t)) \frac{dm_{0,*}}{dt}, \quad \frac{dm_{0,*}}{dt} = -C m_{0,*}^{1/3} \Xi(I_{\min}(t)), \quad \Xi(I_{\min}(t)) = \begin{cases} 0 & I_{\min}(t) < I(t) \\ S-1 & I_{\min}(t) = I(t) \end{cases} \quad (36)$$

where we remind the definition of the largest initial mass, which is evaporated, i.e. $m_{0,*}(t) = \left(-\frac{2}{3} C I_{\min}(t)\right)^{3/2}$. Note, that even for values $\Xi \neq 0$ the initial value problem $m_{0,*}(t_0) = 0$ is not unique (since the right hand side of the ODE is not Lipschitz continuous), i.e. the onset of evaporation might not be unique. However, we can use the same arguments as in Hanke and Porz (2020), that the physical conditions lead to a unique solution, determining the onset and also the further evolution of evaporating masses. The right hand side allows piecewise continuous functions via the function Ξ , and we are searching for continuous solutions representing the lower bound of evaporating mass bins. Therefore, any general improvement of the number concentration evaporation has to tackle the following two contributions:

- The evaporation term depends strongly on the shape of the initial distribution, i.e. $\dot{n} \propto f_0(m_{0,*}(t))$, as can be seen from the PBE solution. Thus at $t = 0$ the number concentration must fulfill the condition $\dot{n}(0) = 0$ when a lognormal



distribution is prescribed as initial distribution. Contrary, standard bulk schemes predicts $\dot{n}(0) = r_0^{-\frac{1}{9}} C(S-1) n_0^{5/3} q_0^{-2/3}$. As can be seen in Fig. 5, this ansatz predicts a large initial evaporation rate for all particles, which is not always true.

- The switching between subsaturated and supersaturated regimes must be implemented in a consistent way, i.e. evaporation only takes place when $I_{min}(t)$ decreases; otherwise, we do not lose particles (i.e. n). This implies that evaporation of ice crystals should not start only when $S < 1$ but a "harder constraint" must be implemented. The accuracy of this constraint depends on how precisely the memory of the distribution is tracked.

3.3 Logistic Bulk

Here, we describe our first modification of the evaporation rate. In order to not increase the computational complexity of the bulk scheme, we try to find a functional approximation such that $\dot{n} = F_n(n, q, S)$; thus, we do not increase the amount of variables in the system of ODEs. The strategy consists in proposing a meaningful shape for the function F_n with some free fitting parameters. The parameters are found by using least squares with the data obtained from the analytical solutions. The data is generated in constant subsaturation regimes.

In order to capture the monomodal structure of the considered distributions we propose the fit for the distribution shape in terms n, q

$$\frac{1}{n_0} \frac{dn}{dt} \approx K_n \left(\frac{n}{n_0} \right)^a \left(\frac{q}{q_0} \right)^b \left(1 - \left(\frac{n}{n_0} \right)^a \left(\frac{q}{q_0} \right)^b \right) \frac{dm_{0,*}}{dt} \quad (37)$$

where K_n, a, b are the fitting parameters. As a functional form we choose a generalized logistic growth law, which leads to the time evolution of the total number concentration with an sigmoidal shape (with an inflection point). The values n_0, q_0 denote initial values values of n, q , i.e. these values must be stored for the new scheme. It is noteworthy that by the definition this ansatz fulfills $\dot{n}(0) = 0$ and $\dot{n} = 0$ whenever $n, q = 0$. For a lognormal distribution with σ_m, m_m the following fitting parameters are obtained

$$K_n = \frac{y(\sigma_m)}{m_m^{2/3}}, \quad y(\sigma_m) = 2.25 \times (\log \sigma_m)^{-0.711} \quad (38)$$

$$a = a(\sigma_m) = 0.062 + 1.091\sigma_m - 0.264\sigma_m^2 \quad (39)$$

$$b = b(\sigma_m) = 0.004 \times \sigma_m^{4.47} \quad (40)$$

which are fitted for the range of interest $\sigma_m \in [1.1, 7.0]$. With the shape of distribution already approximated we just need to set an approximated switching condition, i.e. expressing the function $\frac{dm_{0,*}}{dt}$. A way to do so without increasing the amount of variables is the following expression:

$$\frac{dm_{0,*}}{dt} \approx C(S-1)H(1-S)H\left(\frac{q_0}{n_0} - \frac{q}{n}\right) \quad (41)$$

with $H(x)$ the Heaviside step function. This ansatz prevents the system of ODEs to reduce the number concentration if the current mean mass is larger than the initial mean mass of the system $m_0 = \frac{q_0}{n_0}$. The newly proposed number concentration



395 evaporation rate is expressed as:

$$\dot{n} = n_0 C(S-1) \frac{y(\sigma_m)}{m_m^{2/3}} \left(\frac{n}{n_0}\right)^{a(\sigma_m)} \left(\frac{q}{q_0}\right)^{b(\sigma_m)} \left(1 - \left(\frac{n}{n_0}\right)^{a(\sigma_m)} \left(\frac{q}{q_0}\right)^{b(\sigma_m)}\right) H(1-S) H\left(\frac{q_0}{n_0} - \frac{q}{n}\right). \quad (42)$$

This adapted bulk scheme will be referred to as the **logistic bulk scheme** due to the functional form of the ansatz. The main advantages of such a scheme is that we keep a structure similarly to the standard bulk schemes and do not increment the numerical complexity of the system of ODEs by adding more variables. However, we have the drawback that we need to perform empirical fits of the distribution function with a rather arbitrary ansatz, which has to be redone every time the distribution is to be changed, and it is not guaranteed that this procedure works sufficiently well for all distribution types.

In the upcoming bulk schemes modifications we circumvent this tedious process of fitting for every single distribution type; however, we pay the price of adding more variables to the system of ODEs.

3.4 One memory variable scheme

405 In this new modification of the bulk scheme we introduce the variable $m_{0,*}$ to the system of ODEs. As described in the previous sections, this variable tracks the largest evaporated mass (bin). Since the derivative $\frac{dm_{0,*}}{dt}$ depends on the saturation memory function I and its minimum I_{min} , the switching condition must be approximated again. However, the distribution shape can be captured exactly. Approximating the switching condition via the mean mass condition as before yields the following two ODEs

$$410 \quad \dot{n} = C(S-1) m_{0,*}^{1/3} f_0(m_{0,*}) H(1-S) H\left(\frac{q_0}{n_0} - \frac{q}{n}\right) \quad (43)$$

$$\dot{m}_{0,*} = -C(S-1) m_{0,*}^{1/3} H(1-S) H\left(\frac{q_0}{n_0} - \frac{q}{n}\right) \quad (44)$$

which are now added to the standard bulk scheme. In this modified bulk scheme, one additional variable is used to track the lower bound of the distribution, where evaporation takes place exactly. The switching condition is still implemented in a heuristic manner. This modification will be referred to as **one memory variable scheme**.

415 3.5 Two memory variable schemes

If we want to have the expression for the number concentration rate exactly as it is, then we have to include the saturation memory function I as one of our new variables. Its ODE is by definition

$$\dot{I} = S - 1. \quad (45)$$

Now, the piecewise jump between regimes can be tracked without approximations with the variable I_{min} , which satisfies the ODE

$$\dot{I}_{min} = (S-1) H(1-S) H(I_{min} - I). \quad (46)$$



The full but extended system of ODEs can be written as

$$\dot{q} = r_0^{-1/9} C(S-1)n^{2/3}q^{1/3} \quad (47)$$

$$\dot{S} = AwS - Br_0^{-1/9} C(S-1)n^{2/3}q^{1/3} \quad (48)$$

$$\dot{n} = C(S-1) \left(-\frac{2}{3} CI_{min} \right)^{1/2} f_0 \left(\left(-\frac{2}{3} CI_{min} \right)^{3/2} \right) H(1-S)H(I_{min}-I) \quad (49)$$

$$\dot{I}_{min} = (S-1)H(1-S)H(I_{min}-I) \quad (50)$$

$$\dot{I} = S-1. \quad (51)$$

We have introduced two extra variables to track both, the largest evaporated mass (bin), and the piece-wise jump between regimes. This regime will be referred to as the **two memory variable scheme**. It is noteworthy that no approximation has been made in this last case in order to treat the number concentration evaporation rate. The only closure approximation that remains here is the one performed for the mass concentration evolution.

The assumption of a constant shape parameter r_0 of the lognormal distribution during evaporation/diffusion is however not exactly true. In contrast, we know from our analytical solutions of the PBE, and also former investigations (Gierens and Bretl, 2009), that the shape parameter r_0 and thus the width of the lognormal distribution is changing drastically during the process of evaporation (and also during growth). If we want to reduce the error of the two memory variable scheme further, we can take into account the change of r_0 . Thus, we can replace r_0 in Equations (47)-(51) by $r_{\text{fit}}(\frac{q}{q_{\text{init}}})$ for an empirically determined function r_{fit} given by

$$r_{\text{fit}}(x) = \begin{cases} 2 + 8.3 \cdot \exp(-\log(8.3) \cdot x^{2.6}) & \text{for } x \leq 1 \\ 3 - \frac{4}{\pi} \arctan(3 \cdot (x-1)) & \text{for } x > 1, \end{cases} \quad (52)$$

and where q_{init} denotes the initial mass concentration. The shape parameter is fitted to numerically calculated data points as follows: For constant updraft and downdraft and different initial conditions with number concentrations $10 \text{ L}^{-1} - 500 \text{ L}^{-1}$, initial mean radii $5 \mu\text{m} - 40 \mu\text{m}$ and an initial saturation ratio of $S = 1$, we calculate the full PBE solution over a time of 8 h starting with a lognormal distribution with initial shape parameter of $r_0 = 3$. For equally distributed specified times during that time interval, we consider the resulting distribution and fit the lognormal distribution with shape parameter r'_0 which minimizes the error between the distributions as measured by the integral over the absolute difference. For an example case of $T = 200 \text{ K}$, $p = 300 \text{ hPa}$, and a vertical velocity of $w_0 = \pm 0.01 \text{ m/s}$ the numerically determined shape parameters in dependence of the ratio of the mass concentration $q(t)$ at the considered time t and the initial mass concentration q_0 are displayed by the gray dots in Fig. 6.

The empirical fit works equally well for different temperatures $T = 190 - 240 \text{ K}$, pressures $p = 150 - 300 \text{ hPa}$, and constant low vertical velocities w_0 with $|w_0| \leq 0.1 \text{ ms}^{-1}$, respectively. For non-constant vertical velocities the empirical fit does not match as good as in the case of constant vertical velocities; however, the use of the empirical fit still increases the accuracy of the 2-memory variable scheme as opposed to the version of the same scheme with constant r_0 as illustrated by the examples in the following Sec. 4. This modification will be referred to as **two memory variable scheme with fitted r_0** .

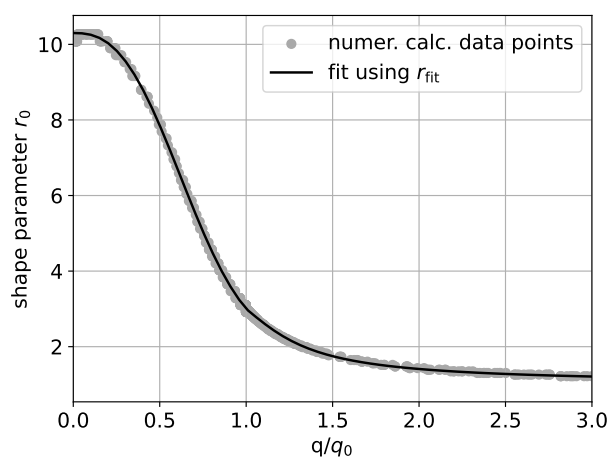


Figure 6. Shape parameter fitting: The gray dots denote numerically calculated shape parameters r_0 for settings with different initial mean radii between $5\mu\text{m}$ and $40\mu\text{m}$ and initial number concentrations between 10L^{-1} and 500L^{-1} depending on the ratio of the mass concentration $q(t)$ at specified times between $t = 0\text{h} - 8\text{h}$ and the initial mass concentration q_0 . Only data points of cases with $q(t) > \frac{q_0}{100}$ are included. The black line indicates the fitted shape parameter as given by Equation (52).



4 Comparison of different approaches

In this section, we compare the different schemes presented in Section 3 with the full solution of the PBE calculated using the equations from Sec. 2 in different exemplary settings. The different schemes are the classical bulk scheme from Sec. 3.1, the logistic scheme from Sec. 3.3, the 1-memory variable scheme from Section 3.4, and the two versions of the 2-memory variable scheme from Sec. 3.5 with and without fitted shape parameter, where the version without the fitted shape parameter is labeled as "2-memory var. scheme" in the plots, while the one with fitted shape parameter is labeled as "2-memory var. scheme, with fitted r_0 ".

In all examples, we only consider the processes of evaporation/diffusion, while other processes such as, e.g., sedimentation are not included. We consider examples of a slow downdraft, an oscillation of the vertical velocity around 0m/s, a slow downdraft with a superimposed oscillation of the vertical velocity, and a slow updraft with a superimposed oscillation of the vertical velocity. In all examples, we set $T = 220\text{K}$, $p = 200\text{hPa}$, $n_{\text{init}} = 200/\text{L}$ as the initial number concentration, and $S = 1$ as the initial saturation ratio. Furthermore, we consider in each example two initial conditions for the initial mass concentration q_{init} corresponding to initial mean radii of $r_{\text{init}} = 5\mu\text{m}$ and $r_{\text{init}} = 20\mu\text{m}$, i.e., small and medium sized ice crystals (cf., Fig. 7(c) in Krämer et al. (2020)).

4.1 Example of a slow downdraft

We consider the example of a slow downdraft of $w_0 = -0.01\text{m/s}$. The time evolutions of the number concentration (left column) and mass concentration (right column) calculated with the different schemes and the full PBE solution are displayed in the plots in Fig. 7, the different modes are shown in the upper ($r_{\text{init}} = 5\mu\text{m}$) and lower ($r_{\text{init}} = 20\mu\text{m}$) row of the figure.

In case of the small ice mode (i.e. small mean initial radius, left column of Fig. 7) the removal of particles (i.e. of number concentration) starts quite fast; after about 6 minutes the PBE solution shows a remarkable decrease in number concentration. The classical bulk scheme depletes the number concentration from the very beginning, as expected from theory. However, all other extended bulk schemes follow the PBE solution very accurately; just the logistic scheme starts a bit earlier. All schemes produce very similar values of (very low) number concentrations after about 24 minutes. For the mass concentration the picture is a bit different. The PBE solution is only matched by the corrected 2-memory scheme. All other schemes produce a stronger evaporation, leading to relevant differences after about 15 minutes. Note, that these differences in masses occur in a regime where the number concentration is quite small already, i.e. most of ice particles have already evaporated.

In case of the ice crystal mode with larger particles (right column of Fig. 7), the removal of ice particles is strongly delayed for the analytical PBE solution, i.e. remarkable changes are present after about one hour. Again, we see the strong difference in the classical bulk scheme, which produces a strong decrease of number concentration from the very beginning. We clearly see that even the shape of the time evolution is not correct, producing an almost linear decay in contrast to the s-shaped solutions by the PBE. Here, we also see differences in the solutions of the "corrected" bulk schemes. While the logistic scheme is underestimating the PBE solution, the 1-memory and 2-memory schemes overestimate the number concentrations. Only the 2-memory scheme with fitted r_0 matches the PBE solution almost perfectly.

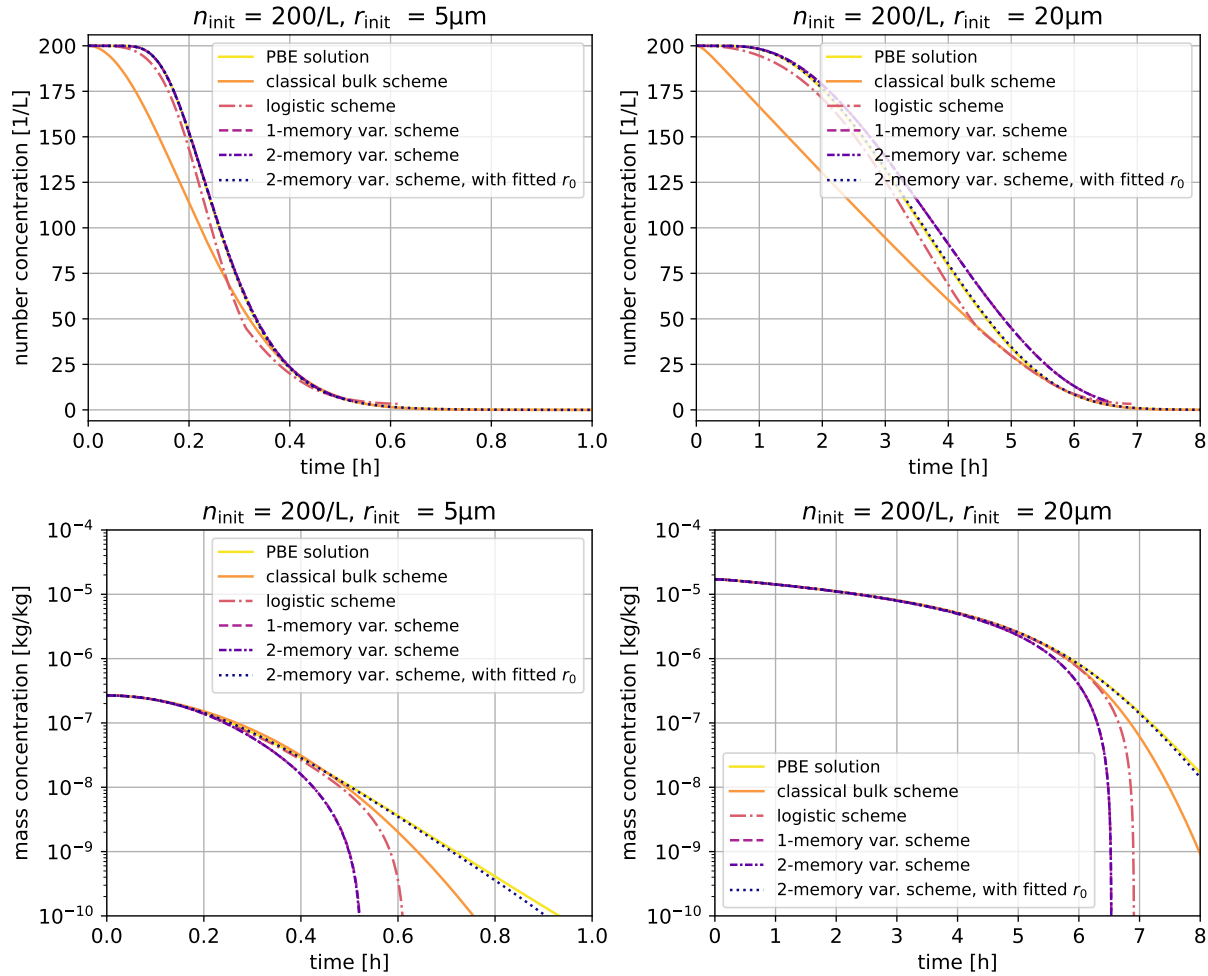


Figure 7. Time evolution of the number concentration (top) and mass concentration (bottom) for a slow downdraft of $w_0 = 0.01 \text{ m/s}$ for ice crystals with small mean initial radius of $5 \mu\text{m}$ (left) and medium initial radius of $20 \mu\text{m}$ (right).

While for the number concentration the differences are quite visible, for the change in the mass concentration we find a different picture. Actually, all schemes are almost indistinguishable until about 5 hours; afterwards, they produce different values, only the 2-memory scheme with fitted r_0 matches the PBE solution.

We can conclude that for small downdrafts it seems that all new schemes produce better results than the classical bulk scheme; the differences and the performance of the schemes clearly depend on the initial condition of the ice crystal mode.



4.2 Example of an oscillation in the vertical velocity

We consider the example of an oscillation in the vertical velocity of the form

$$w(t) = w_{amp} \cos\left(\frac{2\pi t}{\tau}\right) \quad (53)$$

with an amplitude of the oscillation $w_{amp} = 0.1 \text{ m/s}$ and period length $\tau = 1800 \text{ s}$. This can be seen as a simple parameteri-
 495 zation of a gravity wave propagating through an existing ice clouds, e.g., for anvil clouds produced by a thunderstorm. This
 approach of mimicking a gravity wave was used in former investigations (Spichtinger and Krämer, 2013). Again, the time
 evolutions of the number concentration and mass concentration calculated with the different schemes and the full PBE solution
 are displayed in the plots in Fig. 8.

This scenario is quite sensitive to the respective scheme, since the oscillation shifts the prescribed ice mode periodically in
 500 the mass space. For the PBE the general evolution is quite clear. In a first decrease of saturation ratio below equilibrium (i.e.
 $S < 1$) the ice particle distribution is shifted towards the boundary at $m = 0$. All particles up to the mass $m_{0,*}$ are evaporated,
 thus changing the number concentration to a lower value $n_1 < n_0$. In the following evolution the number concentration does
 not change anymore: The distribution is just periodically shifted back and forth in mass space, but it is not shifted further to the
 left, so no particles are evaporated (via the boundary condition). In contrast, the mass of the ice particles changes periodically.
 505 Having in mind this scenario of the full PBE solution, it is quite clear that the change of the number concentration must depend
 crucially on the initial distribution, centered at a mean mass/radius. For a small initial radius, more ice particles are lost than for
 an initially larger radius. This is exactly what we see in the time evolution of the PBE solution for the number concentration.
 In the upper left panel of Fig. 8 the initial distribution centered at a small radius $r_{init} = 5 \mu\text{m}$ is shifted towards the boundary
 $m = 0$, and a large fraction of the particles is lost by evaporation. Afterwards, the number concentration remains constant. A
 510 similar but different behavior is seen for an initially larger ice mode, centered at a radius $r_{init} = 20 \mu\text{m}$, see upper right panel of
 Fig. 8. The number concentration of the PBE is only reduced marginally (hard to see), and then stays constant until the end of
 the simulation. The subsaturation driven by the oscillation is not sufficient to shift the whole distribution far enough to smaller
 masses, such that there is a noticeable change in n .

While the analytical PBE solution produces a meaningful result, the bulk schemes have some problems to represent the
 515 correct features of the scenarios. The classical bulk scheme starts earlier than all other schemes to lose particle (numbers).
 In addition, by definition the scheme cannot keep track on the lower boundary. Thus, the scheme evaporates particles if the
 saturation ratio falls below equilibrium, producing a stepwise behavior. This can be very nicely seen in the case of the large
 ice mode (orange curve in the upper right panel of Fig. 8). For the extended bulk schemes including some kind of memory the
 results are closer to the analytical PBE solution. In case of the small ice mode (left panel) all schemes are quite close to the PBE
 520 solution; they show the strong decrease in number concentration around $t \sim 0.5 \text{ h}$, and afterwards the number concentration
 remains constant. In case of the larger ice mode (right panel) the logistic scheme and the 1-memory scheme have some issues,
 producing some more (small) steps until they stay at a constant value. The two 2-memory schemes perform very well and stay
 close to the PBE solution.

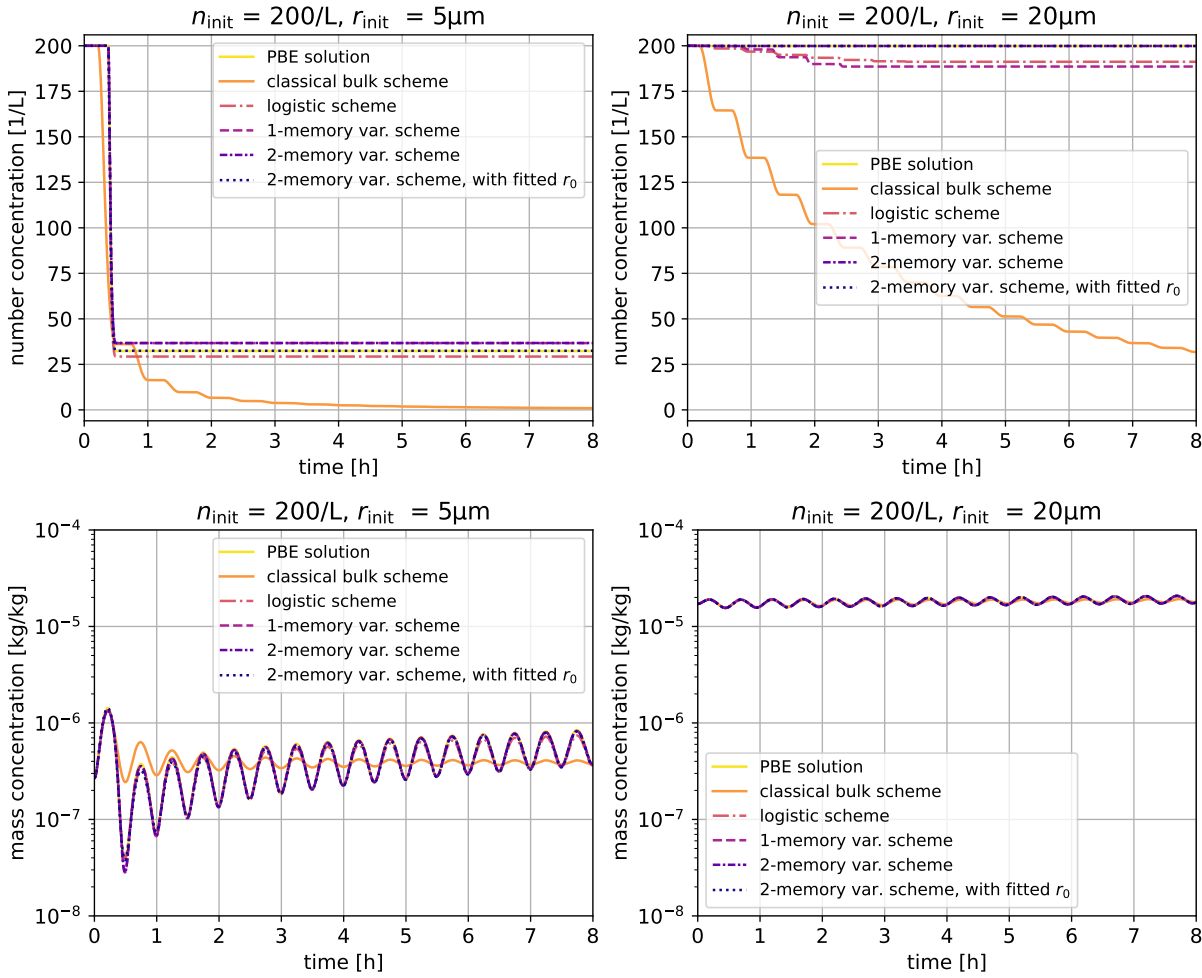


Figure 8. Time evolution of the number concentration (top) and mass concentration (bottom) for a vertical velocity of the form $w(t) = w_{\text{amp}} \cos\left(\frac{2\pi}{\tau}t\right)$ with amplitude $w_{\text{amp}} = 0.1 \text{ m/s}$ and period length $\tau = 1800 \text{ s}$ for ice crystals with small mean initial radius of $5\mu\text{m}$ (left) and medium initial radius of $20\mu\text{m}$ (right).

For the change in the mass concentration, only the classical bulk scheme shows some differences for the small ice mode; in
 525 all other cases the schemes are very close to the analytical PBE solution.

Note that the ansatz of the cooling term in $\frac{dS}{dt} = AwS$ as in Eq. (18) leads to a model that is not mass conserving, but where
 a slight accumulation of mass occurs over time. This is also visible in the very slightly increasing mass concentration in this
 example displayed the bottom row of Fig. 8. However, the increase in the average mass concentration is only very minor here
 and not substantial in the considered time interval. Actually, the bias is present in all considered schemes.



530 4.3 Example of a slow downdraft superimposed with an oscillation

We consider the example of a vertical velocity of the form

$$w(t) = w_0 + w_{amp} \cos\left(\frac{2\pi t}{\tau}\right) \quad (54)$$

with $w_0 = -0.01$ m/s, an amplitude of the oscillation $w_{amp} = 0.1$ m/s and period length $\tau = 1800$ s, combining the two former cases as described in Secs. 4.1 and 4.2. Again, the time evolutions of the number concentration and mass concentration
 535 calculated with the different schemes and the full PBE solution are displayed in the plots in Fig. 9.

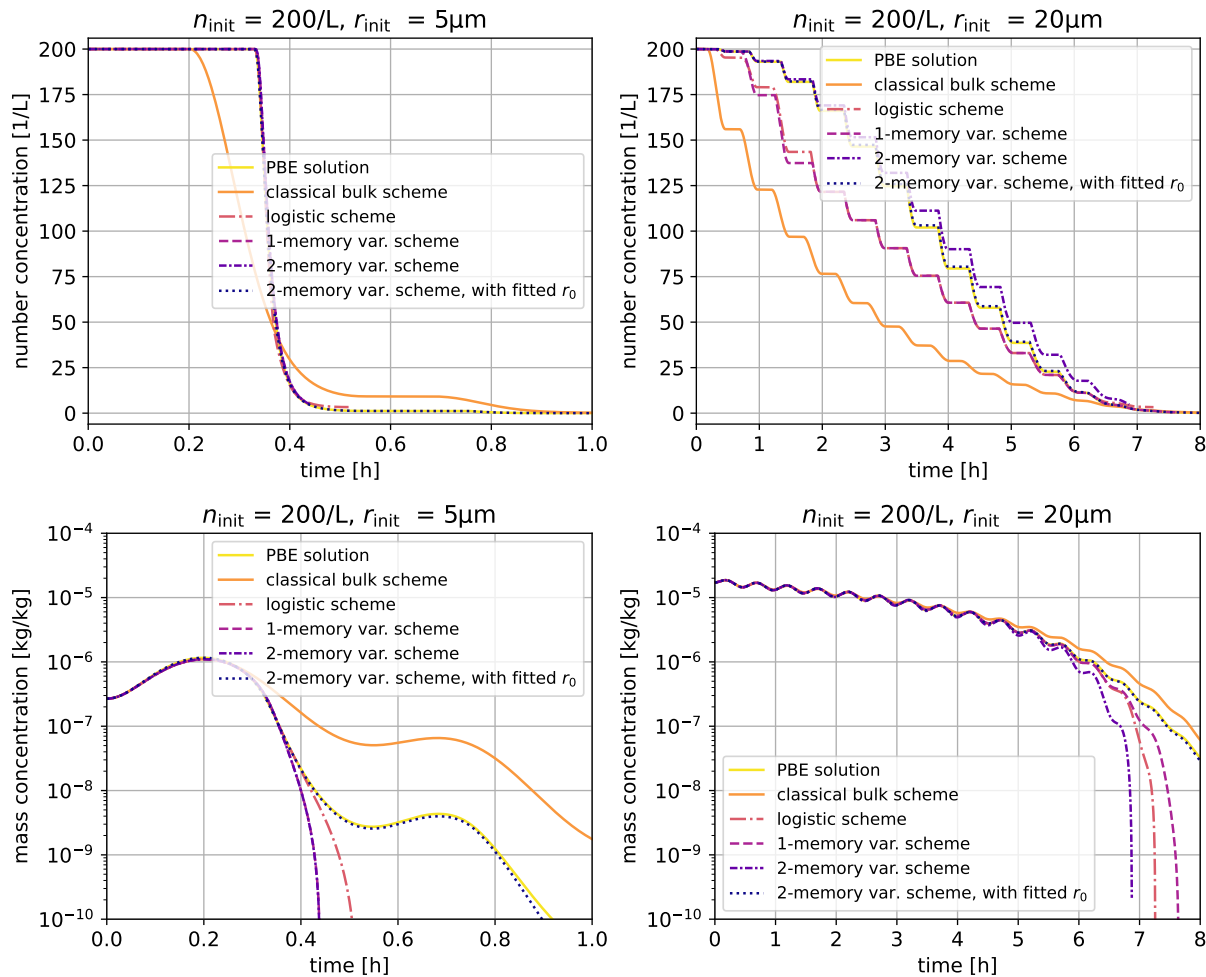


Figure 9. Time evolution of the number concentration (top) and mass concentration (bottom) for a vertical velocity of the form $w(t) = w_0 + w_{amp} \cos\left(\frac{2\pi}{\tau}\right)$ with large scale downdraft $w_0 = -0.01\text{ms}^{-1}$, amplitude $w_{amp} = 0.1\text{ms}^{-1}$, and period length $\tau = 1800$ s for ice crystals with small mean initial radius of $5\mu\text{m}$ (left) and medium initial radius of $20\mu\text{m}$ (right).



The combination of the two scenarios (downdraft + oscillation) shows features of both single scenarios; again, the choice of differently large ice modes as initial conditions has a strong impact on the solutions, and thus on the final choice of a meaningful bulk scheme. We first discuss the theoretical behavior of the PBE solution. Generally, we would expect a slow shift of the mass distribution towards the boundary $m = 0$ as induced by the slow downdraft, superimposed by an oscillatory behavior of the distribution in the mass space. Thus, it can be expected that in contrast to the purely oscillating case in Sec. 4.2 with only one evaporation event, there should be more than one stepwise change in the number concentration, since the mean state of the oscillation is shifted by the superimposed downward motion.

In case of the small ice mode (left panel) the gravity wave superimposed to the downdraft leads to an initially supersaturated regime ($S > 1$) until $t \sim 12$ min; thus the mass concentration increases and the number concentration stays constant (in the PBE solution). Afterwards the particles start to evaporate, i.e. the distribution is shifted towards smaller masses. At about $t \sim 21$ min the distribution reaches the boundary $m = 0$, thus ice particles are lost. Since the gravity wave is leading to a strongly subsaturated regime, the decrease of number concentration is quite fast until the regime becomes supersaturated again ($t \sim 30$ min). Afterwards, a second oscillation is reducing the ice particles to $n \sim 0$, i.e. (almost) all particles are lost/evaporated at the end of the simulation. Between the two oscillations as producing the subsaturation, the number concentration remains constant and we see some increase in the mass concentration. The oscillations can be clearly seen in the time evolution of the mass concentration.

In case of the large ice mode, the behavior is slightly more complicated. We see several stepwise changes in the number concentration driven by the supersaturation as induced by the oscillating vertical velocity. The slow downdraft is shifting the mean state slowly towards the boundary $m = 0$, so the oscillation becomes more effective in evaporating ice crystals. Actually, we see the stepwise behavior in the number concentration until the end of the simulation at $t = 8$ h. In the mass concentration the oscillation can be clearly seen. In contrast to the analytical solution of the PBE, we find crucial differences in the resulting solutions of the various bulk models. As expected by design, the classical bulk model reduces the number concentration as soon as $S < 1$. However, the resulting solution show different behavior for the two cases. In case of the small ice mode, the reduction starts earlier but stays constant at a higher level than the PBE solution. In addition, the mass concentration is less reduced than in the PBE case, producing a small amount of ice crystals at the end of the simulations. In case of the large ice mode, the classical bulk scheme is overestimating the reduction of ice particles (i.e. number concentration) tremendously. Also the extended bulk schemes partly show some deviations and issues. While for the small ice mode case, the number concentration is met by all schemes quite well, only the 2-memory scheme with fitted r_0 can reproduce the PBE solution correctly; the other schemes crucially underestimate the masses, leading to an cloud free environment far to soon. For the large ice mode, the logistic scheme and the 1-memory scheme underestimate the number concentration, while the 2-memory scheme overestimates the number concentration, respectively. However, one can state that all extended schemes perform (qualitatively) much better than the classical bulk scheme. For the mass concentration, all schemes (maybe also except of the classical bulk scheme) are very close until the last part of the simulations at low number concentrations. Here, all schemes finally diverge, only the 2-memory scheme with fitted r_0 can match the PBE solution.



570 As expected, we see a superposition of the issues as found for the single cases, i.e. pure downdrafts and pure oscillations, respectively.

4.4 Example of a slow updraft superimposed with an oscillation

Finally, we consider the example of a vertical velocity of the form

$$w(t) = w_0 + w_{amp} \cos\left(\frac{2\pi t}{\tau}\right) \quad (55)$$

575 with $w_0 = 0.01 \text{ ms}^{-1}$, an amplitude of the oscillation $w_{amp} = 0.1 \text{ ms}^{-1}$ and period length $\tau = 1800 \text{ s}$. Again, the time evolutions of the number concentration and mass concentration calculated with the different schemes and the full PBE solution are displayed in the plots in Fig. 10.

Since we superimpose a slow upward motion, the removal of ice particles by evaporation is only possible via the oscillation, i.e. in case of a subsaturation as driven by the downdraft in the gravity wave. In addition, the mean of the oscillation is shifted
 580 towards larger (mean) masses. Thus, we expect that later oscillations in the subsaturated range are weaker in terms of reducing the number concentrations than the earlier ones; it might also occur that the slow growth of ice particles is sufficient to shift that distribution away from the boundary $m = 0$ that later subsaturated events are not able to evaporate many ice crystals or even any at all.

In our scenarios of small and large ice modes, we actually experience the latter case. For the PBE solution we see an initial
 585 increase in the mass concentration (i.e. growth of ice particles) followed by oscillations in the mass concentration. However, the distribution is moved towards larger particles, thus there is only a slight change in the number concentration in the first subsaturated event (which barely can be seen in the figure). Afterwards, the downward motion is not strong enough to shift the mass distribution beyond the boundary $m = 0$, thus the number concentration remains constant. This behavior is represented in all extended bulk schemes, and also the mass concentration is well represented. However, for the classical bulk scheme,
 590 subsaturation leads immediately to loss of ice particles, thus there is a huge deviation of the number concentration as compared to all other schemes. Surprisingly, the mass concentration is represented quite well in the classical bulk scheme. However, one should keep in mind, that for other properties relying on the mean mass of ice crystals (as, e.g., scattering properties) this mismatch in number concentration with a "correct" mass concentration certainly leads to errors in these quantities.

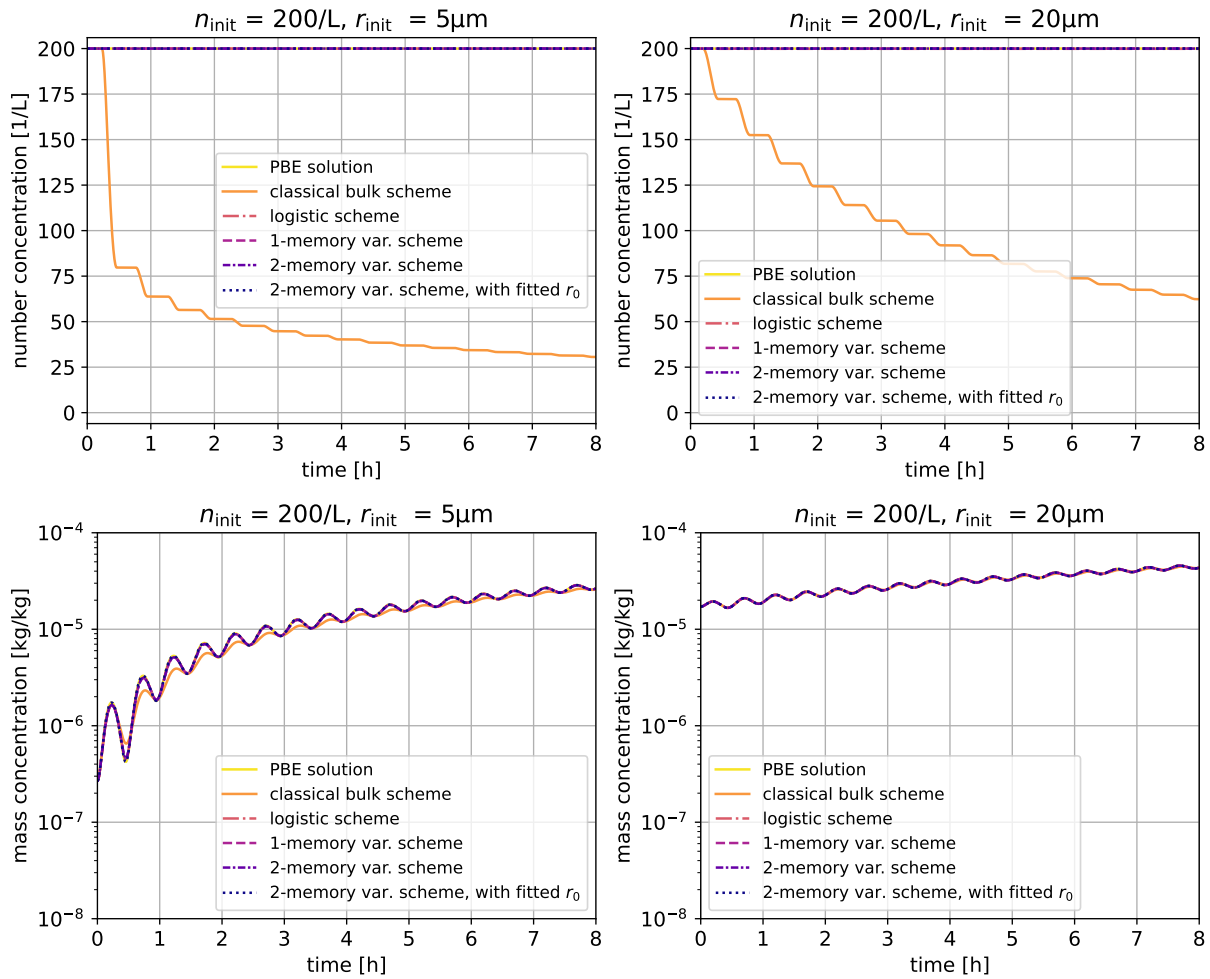


Figure 10. Time evolution of the number concentration (top) and mass concentration (bottom) for a vertical velocity of the form $w(t) = w_0 + w_{amp} \cos\left(\frac{2\pi}{\tau}t\right)$ with large scale updraft $w_0 = 0.01 \text{ ms}^{-1}$, amplitude $w_{amp} = 0.1 \text{ ms}^{-1}$, and period length $\tau = 1800 \text{ s}$ for ice crystals with small mean initial radius of $5 \mu\text{m}$ (left) and medium initial radius of $20 \mu\text{m}$ (right).



5 Discussion and conclusions

595 We analyzed the evaporation process of cloud hydrometeors populations. In order to have a simplified analytical treatment of this phenomenon, we considered a physical regime where the transport in mass space is the only relevant term in the population balance equation (PBE). The "velocity" through mass space was assigned using Maxwell's law for diffusional growth at fixed temperature and pressure. In Sec. 2.2 we investigated the moment transport in this diffusive regime, paying particular attention to the number n and mass q concentration. We showed how the functional form of this two rates is inherently
 600 different. Whereas the mass concentration depends on integral properties of the distribution function, the evaporation rate of number concentration solely depends on a point value of the same distribution. This is a general feature, no matter which mass advection rate is prescribed. Both of these terms yield a not fully determined ODE if the distribution function is not known at all times. Therefore, standard bulk schemes derived from the PBE need some closure approximation for both terms. We observed how the usual closure approximation for the mass concentration fits very well to the that obtained by solving the
 605 PBE.

Our hypothesis to explain such a good agreement is that both the analytical solution and the approximation depend on integral properties of the distribution function. The same does not hold true for the number concentration. The dependency of the rate on a point value of the distribution makes the approximation via moments unsuitable. Although the shape of the number concentration profile is not well captured by any of the usual approximations, standard bulk schemes do not perform bad when
 610 diffusion is fast (see evaporation of cloud droplets in Sec. 3) or when ice crystals are really small in a purely evaporating regime (see the example of slow downdraft in Sec. 4.1). On the other hand, the bulk scheme representation of the number concentration works poorly when for example ice crystals are of a moderate size (see again the slow downdraft in Sec. 4.1) or when there is a switching between supersaturated and subsaturated regimes (see all of the oscillatory examples in Sec. 4).

The general strategy to improve the number concentration representation consists in either better fitting the original distribution shape, having a meaningful switching condition or a combination of both. Throughout Sec. 3 we have presented some possible modifications based on the found analytical solution. These modifications constitute what we refer to as the Memory-Based Bulk Schemes (MBBS): three bulk formulations of increasing complexity — a logistic scheme without additional variables, a one-memory-variable scheme, and a two-memory-variable scheme — that progressively improve the representation of cloud particle number concentration during evaporation. We want to emphasize that none of these schemes is expected to be a
 620 definitive number concentration representation. They should be interpreted as a guide on how to improve this mathematically complex evaporation term, and we encourage the community to improve on them from both a theoretical and practical point of view.

Modifications of the growth law can also be considered. For example, in the Appendix B the solution was presented for general shapes of particles which yield growth rates $\dot{m} \sim m^b$, $0 < b < 1$. In general the method of characteristics can be used
 625 to solve this quasi-linear PDE for all the transports with the mass rate

$$\frac{dm}{dt} = \alpha(m)\gamma(t)$$



and $\frac{1}{\alpha(m)} > 0, \forall m > 0$; $\alpha(m)$ is expected to be continuous. This makes it suitable for further mass growth modification such as corrections for the kinetic or ventilation regimes (Lamb and Verlinde, 2011). Temporal variations in the temperature and pressure fields can also be introduced by using two extra ODEs to solve. This would increase the complexity of the analytical treatment without qualitatively changing the solutions as temperature and pressure dependencies only contribute to the growth constant $C = C(T, p)$ in Maxwell's law. The issue on how to couple the evaporation dynamics with other relevant hydrometeors processes such sedimentation or nucleation still remains open work.



Appendix A: Thermodynamic functions

For the formulation of the growth/evaporation process we use Maxwell's law, as derived by the electrostatic analogon (see, e.g., Lamb and Verlinde, 2011) from a system of partial differential equations, including diffusion of water vapour and heat conduction (Baumgartner and Spichtinger, 2018a). Here, we assume spherical particles, i.e. there is a clear mass-length relation as given by $m = \frac{4}{3}\pi\rho_p r^3$. Since we are interested in the evaporation process, which in turn leads to small particles, this assumption is quite justified, even in case of considering ice particles. Here, ρ_p denotes the density of the respective phase (liquid water or hexagonal ice) of the cloud particle.

The growth law can be formulated as

$$\frac{dm}{dt} = 4\pi D_v G_v (S - 1) r \quad (\text{A1})$$

whereas r can be seen as capacity, if the electrostatic analogon is applied rigorously.

For the impact of latent heat release, changing the temperature of the particle we use the Howell factor (Howell, 1949)

$$G_v = \left[\left(\frac{L}{R_v T} - 1 \right) \frac{L}{T} \frac{D_v}{K_T} + \frac{R_v T}{p_s} \right]^{-1} \quad (\text{A2})$$

including the specific enthalpy of sublimation L (i.e. latent heat), the ideal gas constant of water vapor R_v , the diffusion constant of water vapor in air, and the specific heat conductivity of air K_T , respectively. For the latent heat we use the temperature dependent formulation by Murphy and Koop (2005, their equations (5) or (9), depending on the respective phase liquid water vs. hexagonal ice).

The diffusion coefficient can be formulated via the experimental fit (Hall and Pruppacher, 1976)

$$D_v = 2.11 \cdot 10^{-5} \text{m}^2 \text{s}^{-1} \left(\frac{T}{T_0} \right)^{1.94} \left(\frac{p_0}{p} \right), \text{ using } T_0 = 273.15 \text{K}, p_0 = 101325 \text{Pa}. \quad (\text{A3})$$

For the conductivity we use the formulation

$$K_T = \frac{a_K T^{b_K}}{T + T_K \cdot 10^{\frac{c_K}{T}}}, \text{ using coefficients } a_K = 0.002646 \text{W m}^{-1} \text{K}^{1-b_K}, b_K = \frac{3}{2}, T_K = 245 \text{K}, c_K = -12 \text{K}. \quad (\text{A4})$$

The saturation ratio for hydrometeors is defined as

$$S := \frac{p_v}{p_s(T)} = \frac{pq_v}{\epsilon p_s(T)} \quad (\text{A5})$$

using the partial water vapor pressure p_v and the saturation vapor pressure $p_s(T)$ of the respective stable phase, which is determined by the Clausius-Clapeyron equation

$$\frac{dp_s}{dT} = \frac{L p_s}{R_v T^2}. \quad (\text{A6})$$

The saturation ratio can be reformulated, using the ideal gas laws for water vapor and air together with the water vapor mass concentration $q_v = \frac{M_v}{M_a}$ and the ratio of the specific gas constants, leading to the ratio of molar masses of water and air

$$\epsilon = \frac{M_{mol,v}}{M_{mol,a}} \approx 0.622. \text{ For the saturation vapor pressures we use the fits by Murphy and Koop (2005).}$$



Using the spherical shape, we can finally reformulate the growth equation as

$$\frac{dm}{dt} = C(S-1)m^{1/3}, \quad C(T,p) = 4\pi D_v G_v \left(\frac{3}{4\pi\rho_p} \right)^{1/3}, \quad (\text{A7})$$

as it is used in the main text.

For the treatment of the time evolution of evaporation, we have to formulate a differential equation for the saturation ratio.

665 The time evolution of the saturation ratio is given by

$$\frac{dS}{dt} = \frac{\partial S}{\partial T} \frac{dT}{dt} + \frac{\partial S}{\partial p} \frac{dp}{dt} + \frac{\partial S}{\partial q_v} \frac{dq_v}{dt} \quad (\text{A8})$$

with the respective partial derivatives

$$\frac{\partial S}{\partial T} = -S \frac{L}{R_v T^2}, \quad \frac{\partial S}{\partial p} = \frac{S}{p}, \quad \frac{\partial S}{\partial q_v} = \frac{p}{\epsilon p_s(T)} = \frac{S}{q_v} \quad (\text{A9})$$

where the Clausius-Clapeyron equation has been used in the first equation. The total derivatives of the thermodynamic variables

670 are

$$\frac{dT}{dt} = -\frac{g}{c_p} w - \frac{L}{c_p} \frac{dq_v}{dt}, \quad \frac{dp}{dt} = \frac{dp}{dz} \frac{dz}{dt} = -g\rho_a w, \quad \frac{dq_v}{dt} = -\frac{dq}{dt}, \quad (\text{A10})$$

here, we assume adiabatic changes in temperature and pressure, respectively, and hydrostatic balance of air $\frac{\partial p}{\partial z} = -g\rho_a$ with the air density ρ_a . For the change in temperature we take into account latent heat release due to phase changes.

Using these derivatives, the evolution equation of S becomes the ODE

$$675 \quad \frac{dS}{dt} = A(T,p)wS - B(T,p,S) \frac{dq}{dt} \quad (\text{A11})$$

with the thermodynamic functions

$$A(T,p) = \left[\frac{L}{c_p R_v T^2} - \frac{1}{R_a T} \right] g, \quad B(T,p,S) = \left[\frac{L^2}{c_p R_v T^2} S + \frac{p}{\epsilon p_s(T)} \right] \quad (\text{A12})$$

For our investigations with ice particles, we assume low temperatures $T < 240\text{K}$; in this regime, we can neglect the first term of the function B , thus we can write

$$680 \quad B(T,p,S) \approx \frac{p}{\epsilon p_s(T)}, \quad (\text{A13})$$

see also the estimation of errors in Bergner and Spichtinger (2026). For the liquid cloud regime, an approximation

$$B(T,p,S) = \left[\frac{L^2}{c_p R_v T^2} S + \frac{p}{\epsilon p_s(T)} \right] \approx \left[\frac{L^2}{c_p R_v T^2} + \frac{p}{\epsilon p_s(T)} \right] \quad (\text{A14})$$

with $S \approx 1$ might be appropriate.



Appendix B: Solution Diffusive Regime (Method of Characteristics)

685 In this section we present the analytical solution of the PBE in the diffusive regime via the method of characteristics. The corresponding PBE is

$$\partial_t f + \partial_m \left(\frac{dm}{dt} f \right) = 0; \quad (\text{B1})$$

note here that the term $\frac{dm}{dt}$ describes a velocity in mass space, thus there is advection through the mass space. For this rate, we choose the following form

$$690 \quad \frac{dm}{dt} = C(T, p)(S - 1)m^b, \quad 0 < b < 1, \quad (\text{B2})$$

with $C(T, p)$ a thermodynamic function which corresponds to that given by Maxwells law when $b = 1/3$. The avoidance of exponents $b \geq 1$ is because the exponential or super-exponential rates do not predict annihilation of particles in finite time. Using this mass advection rate the PBE can be written as

$$\partial_t f + C(S - 1)m^b \partial_m f = -bC(S - 1)m^{b-1} f, \quad (\text{B3})$$

695 assuming a time-dependent function $S(t)$. This is a quasi-linear PDE which can be solved via the method of characteristics (see, e.g., Nandakumaran and Datti, 2020), which in essence consists of solving the PBE along a certain set of curves, parametrized with a variable τ referred to as the characteristic curves. The method allows us to map this class of PDEs to a system of ODEs which defines the characteristic curves, which for our case are given by the equations

$$\frac{dt}{d\tau} = 1, \quad \frac{dm}{d\tau} = C(S - 1)m^b, \quad \frac{df}{d\tau} = -bC(S - 1)m^{b-1} f. \quad (\text{B4})$$

700 The solutions of the system then satisfy

$$t - t_0 = \tau, \quad (\text{B5})$$

$$m^{1-b} - m_0^{1-b} = (1 - b)C(I(t) - I(t_0)), \quad (\text{B6})$$

$$m^b f(t, m) = m_0^b f_0(t_0, m_0), \quad (\text{B7})$$

where we have defined the so called saturation memory function

$$705 \quad I(t) = \int_0^t d\tau (S(\tau) - 1). \quad (\text{B8})$$

Since we want to prescribe the distribution at $t = 0$ we choose our characteristic curves to start at $t_0 = 0$ and therefore $t = \tau$ and the function $f(0, m_0) = f_0(m_0)$ is the initial distribution function. Inverting the map $m_0(t, m)$ along the characteristics yields the final result

$$f(t, m) = m^{-b} (m^{1-b} - (1 - b)CI(t))^{\frac{b}{1-b}} f_0 \left((m^{1-b} - (1 - b)CI(t))^{\frac{1}{1-b}} \right), \quad (\text{B9})$$



710 considering the relevant regime with a positive argument in the function f_0 and to constraints with the saturation memory function as specified in the main text (this leads to specific conditions and thus piecewise definitions of the solution). In the main text only the case $b = 1/3$ is studied, corresponding to spherical hydrometeors. The detailed treatment of the characteristic curves at the boundary condition is also discussed in the main text.

Note here that for a given function $S(t)$, the procedure results into the well-known method of characteristics. However, in our case the right hand side of the differential equation for the saturation ratio directly depends on the distribution function f itself via the mass concentration, i.e. via the (weighted) integral over f . Generally, it is not clear that in such cases the method of characteristics works. However, general problems of this form are treated by Keimer and Pflug (2017) and others in the field of non-local balance laws. We apply the method of characteristics even for this general case in the spirit of these investigations.

Appendix C: Lognormal distribution

720 We define the lognormal distribution

$$f(m) := \frac{1}{\sqrt{2\pi} \log(\sigma_m)} \exp\left(-\frac{1}{2} \left(\frac{\log\left(\frac{m}{m_m}\right)}{\log(\sigma_m)}\right)^2\right) \frac{1}{m} \quad (C1)$$

with a geometric mean mass m_m and geometric mass standard deviation σ_m , respectively. As usual, we normalize the distribution by the total number concentration of particles n . The general moments of the distribution can be calculated as

$$\mu_k = n m_m^k \exp\left(\frac{1}{2} (k \log(\sigma_m))^2\right). \quad (C2)$$

725 Following the suggestions by Spichtinger and Gierens (2009), we can define a shape parameter

$$r_0 := \frac{\mu_0 \mu_2}{\mu_1^2} = \exp\left((\log(\sigma_m))^2\right), \quad (C3)$$

which can be used for describing the general moments in a nicer way, i.e.,

$$\mu_k = n m_m^k r_0^{\frac{k^2}{2}} = n \bar{m}^k r_0^{\frac{k(k-1)}{2}}, \quad (C4)$$

using the mean mass $\bar{m} = \frac{\mu_1}{\mu_0} = \frac{q}{n}$. For a closure of a two moment scheme one can fix the type of the distribution to lognormal and set r_0 to a constant value (e.g. $r_0 = 3$, following Spichtinger and Gierens, 2009).



Code availability. The code for evaluation is available at Zenodo (Bergner et al., 2026, <https://doi.org/10.5281/zenodo.23181154>).

Data availability. In this study, the data is produced by the numerical models; it can be reproduced using the code, which is available at Zenodo: <https://doi.org/10.5281/zenodo.23181154>

735 *Author contributions.* ATA and PS conceptualized the study. ATA developed the different models and carried out the numerical calculations with the PBE. HB formulated the shape parameter fitting and conducted the comparison of the PBE and the different evaporation schemes. All authors discussed the results and wrote together the manuscript.

Competing interests. The authors state that there are no competing interests.

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