

# Particle size distributions in atmospheric clouds

By Roberto Paoli & Karim Shariff †

In this note, we derive a transport equation for a spatially integrated distribution function of particles size that is suitable for sparse particle systems, such as in atmospheric clouds. This is done by integrating a Boltzmann equation for a (local) distribution function over an arbitrary but finite volume. A methodology for evolving the moments of the integrated distribution is presented. These moments can be either tracked for a finite number of discrete populations (“clusters”) or treated as continuum variables.

---

## 1. Introduction

Particles are present in atmospheric clouds in several forms such as liquid droplets, non-volatile aerosols or ice crystals. Their microphysical properties control many processes such as the production of rain in stratocumulus clouds and radiation through cirrus clouds. These properties depend on the way particles interact with the surrounding air, through fluid-dynamic and thermodynamic processes. As these processes usually take place at small spatial scales, the interaction of particles with atmospheric turbulence is an important, though complex, problem in cloud physics (Shaw 2003). From a computational point of view, two major factors contribute to this complexity. First is the very high turbulence Reynolds number and the large range of spatial scales (Vaillancourt & Yau 2000; Shaw 2003): for convective clouds, the ratio of energy-containing to dissipative length scales is  $\mathcal{O}(10^5)$ , while the Reynolds number of the largest eddies is  $\mathcal{O}(10^6$  to  $10^7)$ . The second factor is that the mean distance  $\lambda$  between particles is of the order of the Kolmogorov scale  $\eta$ . Thus, if one contemplated direct numerical simulation (DNS) where all spatial scales are resolved, then one would have to track individual particles. Since DNS resolution is not affordable for these flows, Eulerian formulations for the liquid/solid phase are widely used in the simulation of clouds. These formulations fall into two main classes. The first is a “two fluid model” where particles are modeled as a continuum having a local mass density per unit volume. This approach carries no information about the distribution of the particle size. In the second approach, some physical properties (e.g. the mean radius) of some “ensemble” of particles are explicitly computed at each physical location  $\mathbf{x}$ . The concept of particle size distribution at a point at this stage of our discussion is ambiguous but will be clarified later. A standard procedure used in two-phase flow models (e.g. Williams 1965; Cotton & Anthes 1989; Crowe *et al.* 1998) to describe an ensemble of particles is to define a distribution function  $f$ , in a manner analogous to the kinetic theory of gases. In kinetic theory, a distribution function  $f(\mathbf{x}, \mathbf{v}; t)$  is defined where  $f(\mathbf{x}, \mathbf{v}; t) \delta\mathbf{x} \delta\mathbf{v}$  represents the number of molecules that at time  $t$  are between  $\mathbf{x}$  and  $\mathbf{x} + \delta\mathbf{x}$  and whose velocity is between  $\mathbf{v}$  and  $\mathbf{v} + \delta\mathbf{v}$ . It is assumed that, as  $(\delta\mathbf{x}, \delta\mathbf{v}) \rightarrow 0$ , the phase volume still contains a sufficiently large population of molecules that statistics can be used. This is usually true in gasdynamics because the mean free path of molecules is much smaller than the continuum scale one

† NASA Ames Research Center, Moffett Field, CA 94035

cares about. The extension of this approach to particles other than molecules is formally straightforward (see for example the book by Williams 1965), as long as the continuum description remains valid. In the case of atmospheric clouds however,  $\lambda \simeq \eta$  so that only a few particles rather than a population are present in a volume  $V = \mathcal{O}(\eta^3)$ . The object of this note is first to derive a transport equation for an integrated distribution function  $\mathcal{F}_0$ , describing an ensemble of particles inside an arbitrary but finite volume  $V_0$ . Then, this approach is specialized to atmospheric clouds. Finally, a methodology is proposed to solve for the moments of the distribution function  $\mathcal{F}_0$ .

## 2. Distribution function in atmospheric clouds

Let us consider a population of particles in a cloud from an ensemble of realizations. At any time  $t$ , each particle  $p$  occupies the position  $\mathbf{x}_p(t)$  in physical space, moves with velocity  $\mathbf{u}_p(t)$  and changes its radius  $r_p(t)$ . This population can be represented in a phase space, defined by the generalized coordinates  $\mathbf{q}(t)$  and evolving via the generalized velocities  $\mathbf{U}(\mathbf{q}(t))$ :

$$\mathbf{q}(t) = \begin{bmatrix} r(t) \\ \mathbf{x}(t) \\ \mathbf{u}(t) \end{bmatrix}, \quad \mathbf{U}(\mathbf{q}(t)) = \frac{d\mathbf{q}}{dt} = \begin{bmatrix} \dot{r}(\mathbf{q}(t)) \\ \mathbf{u}(\mathbf{q}(t)) \\ \mathbf{F}(\mathbf{q}(t)) \end{bmatrix} \quad (2.1)$$

where  $\mathbf{x}$  and  $\mathbf{u}$  are the spatial coordinates and velocities,  $\mathbf{F}$  is the functional law of the force per unit mass acting on the particle and  $\dot{r}$  is the functional law of the growth rate of its radius. At any time  $t$ , each particle of the population occupies a point  $\mathbf{q}(t)$  in this space. Instead of tracking each particle we wish to follow the evolution of a distribution function  $f(\mathbf{q}(t); t)$  of the population. This is defined in such a way that

$$f(\mathbf{q}(t); t) \delta Q(t) \quad (2.2)$$

is the number of particles that at time  $t$  are inside a cube of volume  $\delta Q(t)$  in phase space, located between the coordinates  $\mathbf{q}(t)$  and  $\mathbf{q}(t) + \delta\mathbf{q}(t)$ . After a time  $dt$ , this volume has a value  $\delta Q(t + dt)$  and the diagonally opposite corners of the cube are mapped to  $\mathbf{q}(t + dt)$ , and  $\mathbf{q}(t + dt) + \delta\mathbf{q}(t + dt)$ , respectively. At the same time, each particle can change its  $\mathbf{q}$  and the particle number can vary because of evaporation or coagulation. To derive a transport equation for  $f$  we need to relate these quantities. For the sake of clarity, we will drop the explicit dependence on  $t$  in all variables, and define  $t' \equiv t + dt$ , and  $\mathbf{q}' \equiv \mathbf{q}(t + dt)$ . Then we have

$$\mathbf{q}' = \mathbf{q} + \mathbf{U}(\mathbf{q}) dt \quad (2.3)$$

$$\mathbf{q}' + \delta\mathbf{q}' = \mathbf{q} + \delta\mathbf{q} + \mathbf{U}(\mathbf{q} + \delta\mathbf{q}) dt \quad (2.4)$$

The last term in (2.4) can be expanded as  $\mathbf{U}(\mathbf{q} + \delta\mathbf{q}) = \mathbf{U}(\mathbf{q}) + (\nabla_{\mathbf{q}}\mathbf{U})\delta\mathbf{q}$  where  $\nabla_{\mathbf{q}}\mathbf{U}$  is the gradient of  $\mathbf{U}$  in phase space. Substituting (2.3) into (2.4) one gets

$$\delta\mathbf{q}' = (\mathbf{I} + dt \nabla_{\mathbf{q}}\mathbf{U}) \delta\mathbf{q} \quad (2.5)$$

The change of phase space volume  $\delta Q' - \delta Q$  is related to the divergence of the generalized velocity  $\mathbf{U}$  by

$$\frac{\delta Q' - \delta Q}{\delta Q} = (\nabla_{\mathbf{q}} \cdot \mathbf{U}) dt + \mathcal{O}(dt^2) \quad (2.6)$$

Let  $K(\mathbf{q}; t)$  describe the general rate of gain or loss of particle number due to coagulation or evaporation,

$$f(\mathbf{q}'; t') \delta Q' = f(\mathbf{q}; t) \delta Q + K(\mathbf{q}; t) dt \delta Q \quad (2.7)$$

Substituting (2.3), (2.4) and (2.6) into (2.7) and expanding gives:

$$\left[ f + \left( \frac{\partial f}{\partial t} + \nabla_{\mathbf{q}} f \cdot \mathbf{U} \right) dt + \mathcal{O}(dt^2) \right] \left[ 1 + \nabla_{\mathbf{q}} \cdot \mathbf{U} dt + \mathcal{O}(dt^2) \right] \delta Q = f \delta Q + K dt \delta Q \quad (2.8)$$

where all quantities are evaluated at  $t$ . Taking the limit  $dt \rightarrow 0$  and neglecting higher order infinitesimals gives a Boltzmann equation

$$\frac{\partial f}{\partial t} + \nabla_{\mathbf{q}} \cdot (f \mathbf{U}) = K \quad (2.9)$$

Finally, inserting the different components of  $\mathbf{q}$  and  $\mathbf{U}$  by means of (2.1) gives the more usual form employed in two-phase flow literature (e.g. Williams 1965):

$$\frac{\partial f}{\partial t} + \nabla_{\mathbf{x}} \cdot (f \mathbf{u}) + \nabla_{\mathbf{u}} \cdot (f \mathbf{F}) + \frac{\partial(f \dot{r})}{\partial r} = K \quad (2.10)$$

The term  $\mathbf{F}$  in (2.10) is the aerodynamic drag induced by the flow on particles,  $\mathbf{F} \approx (\mathbf{u}_p - \mathbf{u}_f)/\tau_p$  (see Crowe *et al.* 1998) where  $\mathbf{u}_f$  is the fluid velocity and  $\tau_p = 4\rho_p r_p^2/18\mu$  is a relaxation time. If the size of the particle  $r_p$  is small,  $\tau_p$  is also small and the particle velocity immediately adjusts to the flow velocity. In the following, we restrict our analysis to this case, so there is no dependence on  $\mathbf{F}$  in (2.10):

$$\frac{\partial f}{\partial t} + \nabla_{\mathbf{x}} \cdot (f \mathbf{u}) + \frac{\partial(f \dot{r})}{\partial r} = K \quad (2.11)$$

### 3. Integrated distribution

We now derive an integrated version of (2.11). Consider a point  $\mathbf{x}_0$  in physical space and an arbitrary (but finite) volume  $V_0(\mathbf{x}_0(t))$  around it. Then, a space integrated distribution function  $\mathcal{F}_0(r, \mathbf{x}_0; t)$  can be defined as

$$\mathcal{F}_0(r, \mathbf{x}_0; t) = \int_{V_0} f(r, \mathbf{x}; t) dV_0(\mathbf{x}) \quad (3.1)$$

so that  $\mathcal{F}_0(r, \mathbf{x}_0; t) \delta r$  represents the number of particles that at time  $t$  are inside a finite volume  $V_0$  around  $\mathbf{x}_0$  and whose radius is in between  $r$  and  $r + \delta r$  (the dimensions of this function are  $[\mathcal{F}_0] = L^{-1}$  whereas  $[f] = L^{-1} \times L^{-3}$ ). The mean  $\psi_0$  within the volume  $V_0$  of any quantity  $\psi(r, \mathbf{x}; t)$  associated with each particle is

$$\psi_0(r, \mathbf{x}_0; t) = \frac{1}{\mathcal{F}_0(r, \mathbf{x}_0; t)} \int_{V_0} f(r, \mathbf{x}; t) \psi(r, \mathbf{x}; t) dV_0(\mathbf{x}) \quad (3.2)$$

Let us introduce a local coordinate  $\mathbf{y}$  around  $\mathbf{x}_0$ ,  $\mathbf{x} = \mathbf{x}(\mathbf{x}_0, \mathbf{y}) = \mathbf{x}_0 + \mathbf{y}$ , so that

$$\nabla_{\mathbf{x}} \cdot (\bullet) = \nabla_{\mathbf{x}_0} \cdot (\bullet) + \nabla_{\mathbf{y}} \cdot (\bullet). \quad (3.3)$$

Using (3.2) for  $\mathbf{u}$  and  $\dot{r}$ , their integral values over  $V_0$  become

$$\mathbf{u}_0(r, \mathbf{x}_0; t) = \frac{1}{\mathcal{F}_0(r, \mathbf{x}_0; t)} \int_{V_0} f(r, \mathbf{x}_0 + \mathbf{y}; t) \mathbf{u}(r, \mathbf{x}_0 + \mathbf{y}; t) dV_0(\mathbf{y}) \quad (3.4)$$

$$\dot{r}_0(r, \mathbf{x}_0; t) = \frac{1}{\mathcal{F}_0(r, \mathbf{x}_0; t)} \int_{V_0} f(r, \mathbf{x}_0 + \mathbf{y}; t) \dot{r}(r, \mathbf{x}_0 + \mathbf{y}; t) dV_0(\mathbf{y}) \quad (3.5)$$

Using (3.3) to express the divergence term  $\nabla_{\mathbf{x}} \cdot (f \mathbf{u}) = \nabla_{\mathbf{x}_0} \cdot (f \mathbf{u}) + \nabla_{\mathbf{y}} \cdot (f \mathbf{u})$ , a transport equation for  $\mathcal{F}_0$  can be derived by integrating (2.11) inside volume  $V_0$ :

$$\int_{V_0} \frac{\partial f}{\partial t} dV_0 + \nabla_{\mathbf{x}_0} \cdot \left( \int_{V_0} f \mathbf{u} dV_0 \right) + \int_{V_0} \nabla_{\mathbf{y}} \cdot (f \mathbf{u}) dV_0 + \frac{\partial}{\partial r} \left( \int_{V_0} f \dot{r} dV_0 \right) = \int_{V_0} K dV_0 \quad (3.6)$$

As in general  $\mathbf{x}_0$  and  $V_0$  vary in time, we need to switch volume integration and time derivative in the first term of the above equation. Using Leibnitz rule and (3.1), one has:

$$\frac{\partial \mathcal{F}_0}{\partial t} \equiv \frac{\partial}{\partial t} \int_{V_0} f dV_0 = \int_{V_0} \frac{\partial f}{\partial t} dV_0 + \oint_{S_0} f \dot{\mathbf{S}}_f \cdot \mathbf{n}_0 dS_0 \quad (3.7)$$

where  $S_0$  is the surface around  $V_0$  and  $\dot{\mathbf{S}}_f$  is the velocity of  $S_0$  with respect to the fixed reference frame. Using (3.7), Gauss theorem and the definitions (3.4) and (3.5) in (3.6), and introducing the relative velocity with respect to  $S_0$ ,  $\mathbf{w} = \mathbf{u} - \dot{\mathbf{S}}_f$ , finally gives

$$\frac{\partial \mathcal{F}_0}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathcal{F}_0 \mathbf{u}_0) + \frac{\partial (\mathcal{F}_0 \dot{r}_0)}{\partial r} + \oint_{S_0} f \mathbf{w} \cdot \mathbf{n}_0 dS_0 = K_0 \quad (3.8)$$

which is a Boltzmann equation for the integral distribution function  $\mathcal{F}_0$ . Equation (3.8) formally differs from (2.11) because of the surface integral in the left-hand side. This contains the (unknown) local distribution function  $f$ , which must be modeled in some way. Note, however, that if  $V_0$  is a material volume, then  $\mathbf{u} = \dot{\mathbf{S}}_f$  on  $S_0$ , and the surface flux goes to zero.

### Ergodic hypothesis

In the previous derivation we had to introduce an ensemble of realizations in order to derive a local Boltzmann equation which we then integrated over a finite volume. We now make the hypothesis that  $V_0$  is large enough to contain a population of particles that we can by-pass the ensemble. In other words, we hypothesize that the integrated Boltzmann equation (3.8) is valid for a single realization if  $V_0$  is large enough. Typically, the grid size in cloud codes is  $\Delta \geq 1 m$  while  $\lambda \simeq \eta \approx 10^{-3} m$ , so each grid cell contains at least  $10^9$  particles.

### 3.1. Ensemble averages

The total number of particles  $N_0$  in spatial volume  $V_0$  can be obtained by integrating  $\mathcal{F}_0$  over all possible radii,

$$N_0(\mathbf{x}_0; t) = \int_0^\infty \mathcal{F}_0(r, \mathbf{x}_0; t) dr. \quad (3.9)$$

Using (3.9), the ensemble average  $\langle \psi_0 \rangle$  of any variable  $\psi$  is obtained by integrating (3.2) over  $r$ :

$$\langle \psi_0 \rangle (\mathbf{x}_0; t) = \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty \psi_0(r, \mathbf{x}_0; t) \mathcal{F}_0(r, \mathbf{x}_0; t) dr \quad (3.10)$$

In particular, the ensemble velocity and radius growth rate are

$$\begin{aligned} \langle \mathbf{u}_0 \rangle (\mathbf{x}_0; t) &= \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty \mathbf{u}_0(r, \mathbf{x}_0; t) \mathcal{F}_0(r, \mathbf{x}_0; t) dr, \\ \langle \dot{r}_0 \rangle (\mathbf{x}_0; t) &= \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty \dot{r}_0(r, \mathbf{x}_0; t) \mathcal{F}_0(r, \mathbf{x}_0; t) dr \end{aligned} \quad (3.11)$$

For further analysis, it will be useful to introduce the mean radius,  $\langle r_0 \rangle$ , and the variance,  $\langle \Delta r_0^2 \rangle$ , of the population. Using (3.2), these are given by

$$\langle r_0 \rangle (\mathbf{x}_0; t) = \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty r \mathcal{F}_0(r, \mathbf{x}_0; t) dr \quad (3.12)$$

$$\langle \Delta r_0^2 \rangle (\mathbf{x}_0; t) = \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty [r - \langle r_0 \rangle (\mathbf{u}_0; t)]^2 \mathcal{F}_0(r, \mathbf{x}_0; t) dr \quad (3.13)$$

Note that  $\mathbf{u}(r, \mathbf{y}; t)$  and  $\langle \mathbf{u}_0 \rangle (\mathbf{x}_0; t)$  (same for  $\dot{r}$  and  $\langle \dot{r}_0 \rangle$ ) have a different physical meaning:  $\mathbf{u}(\mathbf{y})$  represents the velocity of a particle in a neighborhood of  $\mathbf{y}$ . On the other hand,  $\mathbf{u}_0(\mathbf{x}_0)$  represents a statistical average within a population of particles and is a continuum velocity field, associated to any point,  $\mathbf{x}_0$ , of the physical domain. Indeed, one could, in principle, obtain  $\langle \mathbf{u}_0 \rangle$  and  $\langle \dot{r}_0 \rangle$  as

$$\langle \mathbf{u}_0 \rangle = \frac{1}{N_0} \sum_{p=1}^{N_0} \mathbf{u}_p, \quad \langle \dot{r}_0 \rangle = \frac{1}{N_0} \sum_{p=1}^{N_0} \dot{r}_p \quad (3.14)$$

where  $\mathbf{u}_p$  and  $\dot{r}_p$  are the velocity and the radius growth rate of particle  $p$  inside  $V_0$ . In the limit of  $N_0 \rightarrow \infty$ , (3.11) and (3.14) are equivalent but we only have access to  $\mathcal{F}_0$  because the details concerning  $\mathbf{u}_p$  and  $\dot{r}_p$  of each physical particle are unknown.

The next step is to relate the continuum fields  $\langle \mathbf{u}_0 \rangle$  and  $\langle \dot{r}_0 \rangle$  to the corresponding flow variables. As we do not consider here any force acting on particles (Sec. 2), they are simply convected by the fluid. Therefore, there is no reason why two particles of the same population and different radius should have different velocities, i.e.  $\mathbf{u}_0$  is statistically uncorrelated with  $r$ ,

$$\mathbf{u}_0(r, \mathbf{x}_0; t) \equiv \langle \mathbf{u}_0 \rangle (\mathbf{x}_0; t) = \mathbf{u}_f(\mathbf{x}_0; t) \quad (3.15)$$

where  $\mathbf{u}_f(\mathbf{x}_0; t)$  is the fluid velocity at  $\mathbf{x}_0$ . The same arguments, cannot be applied to  $\dot{r}_0$ , i.e.  $\dot{r}_0 \neq \langle \dot{r} \rangle_0$ , because each particle of the population may have a different growth rate due to different "reactions" to turbulent fluctuations in the flow-field, as discussed next.

#### 4. Particle growth by condensation

The growth of the radius of a single particle in a medium at rest can be simply derived by considering a diffusion equation for water vapor on a particle surface (Pruppacher & Klett (1997) p. 502) and is given by

$$\frac{dr}{dt} = \frac{D(\rho_v - \rho_v^s(T))}{\Gamma \rho_w r} = \frac{D S}{\Gamma \rho_w r} \quad (4.1)$$

where  $D$  is the diffusion coefficient of water vapor in air and  $\Gamma$  is the psychrometric correction associated with the latent heat of condensation; and  $\rho_w$  is the density of either water or ice. The vapor densities  $\rho_v$  and  $\rho_v^s(T)$  are evaluated, respectively, at some "ambient" condition far from the particle and at the surface of the particle (which coincides with the saturation value because vapor there is in thermodynamic equilibrium with water/ice). Thus, in such a single-particle picture, the radius growth rate is only controlled by the supersaturation  $S = \rho_v - \rho_v^s$ . As first pointed out by Srivastava (1989) (see also Khvorostyanov & Curry 1999), this description cannot be extended straightforwardly to a population of particles. In fact, even in the absence of turbulence and uniform  $S$  initially, the available vapor in a cloud is not equally distributed among all particles

because of their random spatial distribution, so that the effective supersaturation available at a droplet surface (“microsaturation”), can differ significantly from the overall ensemble averaged supersaturation (“macrosaturation”) (Srivastava 1989). In addition, in a turbulent cloud, each particle “reacts” in a different way to turbulent fluctuations in the flow-field: for example, if a supersaturation fluctuation arises, it will be absorbed by each particle through a complex diffusional process of vapor involving all elements of the population (Srivastava 1989). Several approaches have been developed in the atmospheric science literature (Pruppacher & Klett 1997) to try to solve this complex problem. One of these, the so-called “stochastic condensation” approach performs Reynolds averaging on the equation for condensational growth, resulting in covariances that can be thought as “Reynolds stresses” (Shaw 2003). In particular, we follow Khvorostyanov & Curry (1999) (see also Pruppacher & Klett (1997) p.505) who use kinetic theory to relate the micro- and macro-saturation in a cloud. Their arguments are as follows. For the moment consider the situation where the particle radius is so small to be comparable with the mean free path of vapor molecules. In this case, one should account for the Brownian motion of molecules, that is the diffusion associated to the (random) molecular impact on particles surface. As shown by Pruppacher & Klett (1997), this can be done in (4.1) by introducing a modified diffusion coefficient  $D^*(r)$  which depends linearly on the radius  $r$  (Pruppacher & Klett (1997) p.506). It can be argued (Crowe *et al.* 1998; Khvorostyanov & Curry 1999) that the effects of turbulent fluctuations of vapor density or in supersaturation is similar to Brownian motion, whereas the molecular impact on particles is substituted by their interaction with turbulent eddies (note that this picture can also be extended to account for equivalent Brownian dispersion of particles, induced by fluctuating fluid forces rather than density fluctuations) (Crowe *et al.* 1998). The “micro” supersaturation  $S_p$  available to particle  $p$  of an ensemble is

$$S_p = \frac{\langle S_0 \rangle}{\langle r_0 \rangle} r_p \quad (4.2)$$

where  $\langle S_0 \rangle(\mathbf{x}_0; t)$  is the ensemble supersaturation available to the population within volume  $V_0$ . It represents the supersaturation that would be at  $\mathbf{x}_0$  if there were no particles, then it can be thought as the fluid supersaturation at  $\mathbf{x}_0$ ,  $\langle S_0 \rangle(\mathbf{x}_0; t) = S_f(\mathbf{x}_0; t)$ . Using (4.2) and the previous formalism ( $r_p \rightarrow r$ ;  $S_p \rightarrow S(r, \mathbf{y})$ ;  $\dot{r}_p \rightarrow \dot{r}(r, \mathbf{y})$ ), one gets to

$$\frac{S(r, \mathbf{y})}{r} = \frac{\langle S_0 \rangle}{\langle r_0 \rangle} = \frac{S_f}{\langle r_0 \rangle} \quad (4.3)$$

$$\dot{r}(r, \mathbf{y}) = \frac{DS(r, \mathbf{y})}{\Gamma \rho_w r} = \frac{DS_f}{\Gamma \rho_w \langle r_0 \rangle} \quad (4.4)$$

Substituting (4.3) and (4.4) into (3.11) finally gives

$$\langle \dot{r}_0 \rangle = \frac{1}{N_0(\mathbf{x}_0; t)} \int_0^\infty \mathcal{F}_0 \dot{r}_0 dr = \int_0^\infty \int_{V_0} f \frac{DS(r, \mathbf{y})}{\Gamma \rho_w r} dV_0 dr \equiv \frac{D S_f}{\Gamma \rho_w \langle r_0 \rangle} \quad (4.5)$$

## 5. Method of moments

Even neglecting the surface term, (3.8) is a p.d.e. in four-dimensional space  $(r, \mathbf{x}_0; t)$  that can only be solved numerically. Some atmospheric cloud codes solve a transport equation for a distribution function by discretizing the particle size  $r$  in a finite number of bins, at each grid location (although it is not explicitly mentioned, they are conceptually discretizing (3.8) with neglected surface terms).

In this section, we present a simulation strategy based on the method of moments proposed in Paoli *et al.* (2002). The moments  $m_k$  of the distribution  $\mathcal{F}_0$  are defined by

$$m_k(\mathbf{x}_0; t) = \int_0^\infty r^k \mathcal{F}_0(r, \mathbf{x}_0; t) dr \quad (5.1)$$

Multiplying (3.8) by  $r^k$  gives

$$\frac{\partial}{\partial t} \left( \int_0^\infty r^k \mathcal{F}_0 dr \right) + \nabla_{\mathbf{x}_0} \cdot \left( \int_0^\infty r^k \mathcal{F}_0 \mathbf{u}_0 dr \right) = -\mathcal{F}_0 \dot{r}_0 r^k \Big|_0^\infty + k \int_0^\infty r^{k-1} \mathcal{F}_0 \dot{r}_0 dr \quad (5.2)$$

Using (3.15), (4.5) and (5.1) and assuming that  $\mathcal{F}_0 \rightarrow 0$  sufficiently fast as  $r \rightarrow \infty$ , (5.2) becomes

$$\frac{\partial m_k}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathbf{u}_f m_k) = \frac{D S_f}{\Gamma \rho_w \langle r_0 \rangle} k m_{k-1} \quad (5.3)$$

Under all assumptions made, (5.3) describes the evolution of the moments of the integral distribution function  $\mathcal{F}_0$ . An attractive property, deriving from the microsaturation model (4.2), is that the evolution of the  $k^{\text{th}}$  moment only depends on the previous order moment which allows one to close the system (5.3) without any further assumptions and without presuming the shape of  $\mathcal{F}_0$ . Using (3.9)–(3.13) and (5.1), the zero and the first two moments are easily found and are related to the ensemble average radius and variance,

$$m_0 = N_0, \quad m_1 = N_0 \langle r_0 \rangle, \quad m_2 = N_0 \left[ \langle \Delta r_0^2 \rangle + \langle r_0 \rangle^2 \right]. \quad (5.4)$$

The corresponding evolution equations are (we put  $\alpha \equiv D/\Gamma \rho_w$ )

$$\frac{\partial N_0}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathbf{u}_f N_0) = 0 \quad (5.5)$$

$$\frac{\partial m_1}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathbf{u}_f m_1) = \alpha S_f \frac{N_0^2}{m_1} \quad (5.6)$$

$$\frac{\partial m_2}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathbf{u}_f m_2) = 2 \alpha S_f N_0 \quad (5.7)$$

These equations are coupled to the continuum fluid phase through  $\mathbf{u}_f$  and  $S_f$ . In particular, an increase in particle radius by condensation implies vapor depletion  $\dot{\rho}_v$ , with

$$\dot{\rho}_v = - \int_0^\infty 4\pi \rho_w r^2 \dot{r}_0 \mathcal{F}_0 dr = - \frac{4\pi \rho_w D}{\Gamma \rho_w} \frac{S_f}{\langle r_0 \rangle} \int_0^\infty r^2 \mathcal{F}_0 dr = -4\pi \rho_w \alpha S_f \frac{N_0 m_2}{m_1} \quad (5.8)$$

The usual convection-diffusion-reaction equation for a scalar  $Y_v$  in conservative form ( $\rho_v \equiv Y_v \rho_f$  where  $\rho_f$  is the total gas phase density) then becomes:

$$\frac{\partial \rho_v}{\partial t} + \nabla_{\mathbf{x}_0} \cdot (\mathbf{u}_f \rho_v) + \nabla_{\mathbf{x}_0} \cdot (\rho_f D \nabla_{\mathbf{x}_0} Y_v) = \dot{\rho}_v = -4\pi \rho_w \alpha S_f \frac{N_0 m_2}{m_1} \quad (5.9)$$

Under all approximations made, (5.5)–(5.7) and (5.9) (together with Navier-Stokes equations) describe the evolution of the first moments of the size distribution of a population of particles. These moments can be solved by using either an Eulerian or a Lagrangian description as discussed next.

### Eulerian description

In this case, one has to solve for  $m_k$  using (5.5)–(5.7) with the further condition that the volume  $V_0$  is constant in time (in discretized form it can be the volume of a grid cell). It is worth mentioning that  $\mathcal{F}_0$  and  $m_k$  are continuous functions of space (not grid averages!),

so their gradients contain all spatial fluctuations in a turbulent flow. In particular, if they are filtered in a LES approach, the correlations  $\overline{m_k \mathbf{u}_f}$  and  $\overline{m_k S_f}$  exist at subgrid scale level and must be modeled.

#### An approximate Lagrangian method

Let us divide the total number of particles in the cloud into  $N_c$  "clusters", each containing a fixed number,  $N_j$ , of particles (where  $j = 1, \dots, N_c$ ). The position of each cluster is assumed to evolve according to

$$\frac{d\mathbf{x}_0^j}{dt} = \mathbf{u}_f(\mathbf{x}_0^j) \quad (5.10)$$

where  $\mathbf{x}_0^j$  is the center of the volume  $V_0^j$  containing cluster  $j$  and  $\mathbf{u}_f(\mathbf{x}_0^j)$  is the fluid velocity at  $\mathbf{x}_0^j$ . Note that we are assuming that the cluster advects rigidly without deforming. Introducing the total derivative  $d()/dt = \partial()/\partial t + \nabla_{\mathbf{x}_0}() \mathbf{u}_f$  in (5.5)–(5.7), one can "track" the moments of each cluster  $j$  as (note that the zeroth moment equation,  $N_j = \text{const}$  is now trivial)

$$\frac{dm_1^j}{dt} = \alpha S_f \frac{N_j^2}{m_1^j} \quad (5.11)$$

$$\frac{dm_2^j}{dt} = 2 \alpha S_f N_j \quad (5.12)$$

where  $S_f \equiv S_f(\mathbf{x}_0^j)$  is the fluid supersaturation at  $\mathbf{x}_0^j$ . The advantage of the Lagrangian approach is that the surface term in (3.8), which was neglected to get to (5.5)–(5.7), is now zero because  $V_0^j$  is a material volume,  $\mathbf{u} = \dot{\mathbf{S}}_f^j$  on  $S_0^j$  for all  $j$ .

## 6. Conclusions

In this note we derived a transport equation for the radius distribution function of a population of particles in an atmospheric cloud. We used a simple stochastic condensation model for the radius growth (taken from the atmospheric science literature) to relate the microsaturation around each particle to the macrosaturation of the entire population. Finally, we described a procedure to solve for the moments of the distribution, and showed that this can be either used in Eulerian continuum formulations or Lagrangian tracking of "clusters" of particles. Future plans include testing of this method by comparison with DNS of homogeneous and isotropic turbulence and individual particle tracking; and application of the method to natural and contrail-generated cirrus.

## REFERENCES

- COTTON, W. R. & ANTHES, R. A. 1989 *Storm and Cloud Dynamics*. Academic Press.
- CROWE, C., SOMMERFELD, M. & TSUJI, Y. 1998 *Multiphase Flows with Droplets and Particles*. CRC Press.
- KHVOROSTYANOV, V. I. & CURRY, J. A. 1999 Toward the Theory of Stochastic Condensation in Clouds. Part I: A general kinetic equation *J. Atmos. Sci.*, **56**, 3985–3996.
- PAOLI, R., HÉLIE, J., POINSOT, T. & GHOSAL, S. 2002 Contrail formation in aircraft wakes using large eddy simulation *Proc. 2002 Summer Program* Center for Turbulence Research, Stanford University, Stanford, CA .

- PRUPPACHER, H. R. & KLETT, J. D. 1997 *Microphysics of Clouds and Precipitation*. Kluwer Academic Publishers, Dordrecht, The Netherlands.
- ROGERS, R. R. & YAU, M. K. 1989 *A short course in Cloud Physics*. Butterworth-Heinemann.
- SHAW, R. A. 2003 Particle-Turbulence Interactions in Atmospheric Clouds. *Annu. Rev. Fluid Mech.* **35**, 183-227.
- SRIVASTAVA, R. C. 1989 Growth of cloud Drops by Condensation: A criticism of currently accepted theory and a new approach *J. Atmos. Sci.*, **46**, 869-887.
- VAILLANCOURT, P. A. & YAU, M. K. 2000 Review of Particle-Turbulence Interactions and Consequences for Cloud Physics *Bull. Am. Meteorol. Soc.*, **81**, 285-298.
- WILLIAMS, F. A. 1965 *Combustion Theory* Addison-Wesley Publishing Company.