Droplet Growth by Turbulent Coagulation

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1. INTRODUCTION

Clouds are an important environment for multiphase chemical transformations. These processes are crucially dependent on the cloud microphysical properties and have far reaching consequences concerning the direct and indirect climate effect of aerosols (Kreidenweis et al., 2003).

Droplet composition is observed to depend on droplet size, so an adequate description of the heterogeneous reactions in clouds therefore relies on a detailed knowledge of the factors controlling the droplet size distribution. In this context, droplet - turbulence interactions may play a major role (Pinsky and Khain, 1997; Shaw, 2003).

As laboratory and numerical work have shown the velocity and spatial distributions of particles may be modified significantly in a turbulent flow field. The key mechanisms of particle-turbulence interactions are that: (1) particle inertia leads to relative velocities and less correlated velocity directions and hence to higher collision rates, (2) wind field shear produces collisions between particles even with the same inertia (Saffman and Turner, 1956), and (3) coagulation rates should be enhanced due to local concentration increases for particles with response times on the order of the Kolmorgorov scale (Maxey, 1987). Although there is a general agreement in current literature that turbulence enhances the collision frequency of cloud droplets, this process is not yet well understood and therefore ignored in most current cloud models.

In our study we investigate by numerical simulation the impact of turbulence on the coagulation of cloud droplets using a kernel derived from direct numerical simulations (DNS) (Zhou et al., 2001). The developed model will be the basis for further evaluation of in-cloud chemistry, in particular addressing the question of sulfate production.

2. METHOD

Since experimental data of the droplet-turbulence interactions is difficult to obtain, modeling studies are an important tool for investigation. Zhou et al. (2001) recently developed a model for the turbulent coagulation kernel on the basis of the solution of the Navier-Stokes equations using direct numerical simulations (DNS). Their parameterization covers all three mechanisms mentioned above.

Employing this kernel, we consider a Lagrangian box model of a cloud parcel simulating the evolution of the cloud droplet size distribution, thereby addressing two questions:

- Can turbulence enhance the coagulation process in a way that large droplets are formed fast enough to explain the observed rapid production of warm rain?
- Do our predictions reproduce the observed broadening of the size distribution?

2.1 The turbulent coagulation kernel

The ensemble average of the collision kernel $K_t(r,r')$ for two particles with the radii r and r' in a turbulent fluid can be expressed in a generalized form by (Sundaram and Collins, 1997):

$$K_t(r,r') = 2\pi(r+r')^2 \left\langle |w_r| \right\rangle g(r+r'), \qquad (1)$$

$\left\langle |w_r| \right\rangle$: Ensemble average of the relative velocity of the particles.
$g(r+r')$: Radial distribution function at contact.

The functions $g(r+r')$ and $\left\langle |w_r| \right\rangle$ are parameterized on the basis of the DNS simulations

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depending on the Stokes number of the droplets, the dissipation rate ε of the fluid and the r.m.s. velocity fluctuation *u'* of the fluid

2.2 Calculation of the size distribution

The stochastic collection equation describes the evolution of a colliding and coalescing cloud droplet size distribution (Pruppacher and Klett, 1997). In a first set of simulations, we solve this equation using the flux method by Bott (1998) and the turbulent coagulation kernel K_t according to equation (1). We use a Gamma function of the form $n(m,t) = L_w / m_g^2 \, exp(-m / m_g)$ as initial distribution, where L_w is the total cloud water content and m_g is the mean droplet mass which is related to the mean droplet radius by $m_g(r) = 4/3\pi\rho r_g^3$. For our simulations, L_w is set to 1 g m^{-3} and r_g to 10 μm.

In a second set of simulations we consider an air parcel with a prescribed aerosol distribution that undergoes a cooling process. The aerosol particles are activated and the processes of condensation/evaporation and coagulation shape the size distribution and feed back on the supersaturation of the system. The growth rate of the particles is calculated according to Majeed and Wexler (2001).

The dry aerosol mass is treated on a fixed grid, for the water mass we use a moving grid. The advantage of this hybrid approach for the treatment of condensation/evaporation of water is that it avoids unwanted numerical diffusion. It also ensures that the aerosol mass is regenerated after evaporation, which is important for the investigation of aerosol processing.

We use a log-normal distribution with a mean dry diameter of 0.3 μm, a total number of 1000 cm^{-3}, and a standard deviation of 1.65 as initial dry aerosol distribution. The temperature is 283 K at the start of the simulation and decreases over the course of the simulation with $\partial T / \partial t =$ 0.006 K s^{-1}.

3. RESULTS

Figure 1 shows the turbulent coagulation kernel K_t according to equation (1) for the base case (ε = 300 $cm^2 s^{-3}$ and *u'* = 3.5 m s^{-1}). The turbulent coagulation kernel K_t exhibits a distinct local maximum for the combination of droplet radii near 65 μm and 250 μm using the base case values for ε and *u'*. The exact position as well as the magnitude of this maximum depend on ε and *u'*. For droplets smaller than 100 μm and/or equally sized droplets the turbulent kernel is two magnitudes larger that the sedimentation kernel in calm air. We can therefore expect that the impact of turbulent coagulation is especially significant in the initial stage of the development of the cloud whereas the impact decreases for large cloud droplets or rain drops.

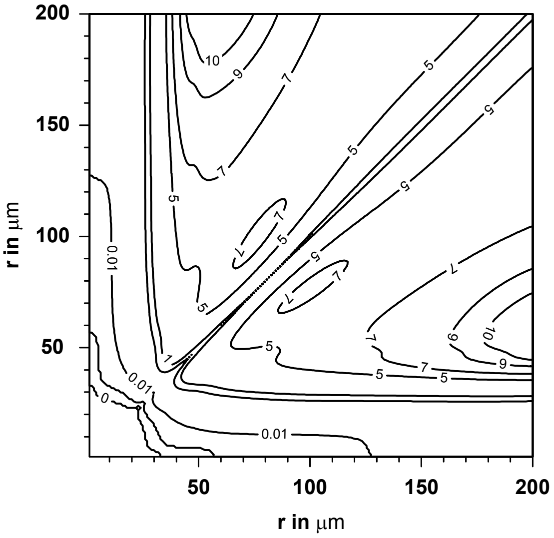

Fig. 1: K_t in $cm^3 s^{-1}$ for ε = 300 $cm^2 s^{-3}$ and *u'* = 3.5 m s^{-1} (base case).

Figure 2 shows results of the first set of simulations where only coagulation is considered and condensation is not included. Figure 2(a) and Figure 2(b) display the development of the mass distribution *g*(ln *r*) with time for the sedimentation kernel and for the turbulent kernel for the base case. For the sedimentation kernel a second mode appears only after about 30 minutes. At t = 30 min., most of the mass (97%) is still distributed over the droplet size range smaller than 100 μm, confirming the well known fact that sedimentation alone cannot explain the fast formation of large droplets.

For the turbulent kernel, the second mode forms already after 10 minutes indicating that the turbulent coagulation kernel of Zhou et al. (2001) accelerates the formation of large drops. Figure 2(c) shows the effect of the combined sedimentation and turbulent kernel. For this case, the second mode becomes visible after only 5 minutes, and after 30 minutes 96% of the mass is transferred to sizes larger than 100 μm.

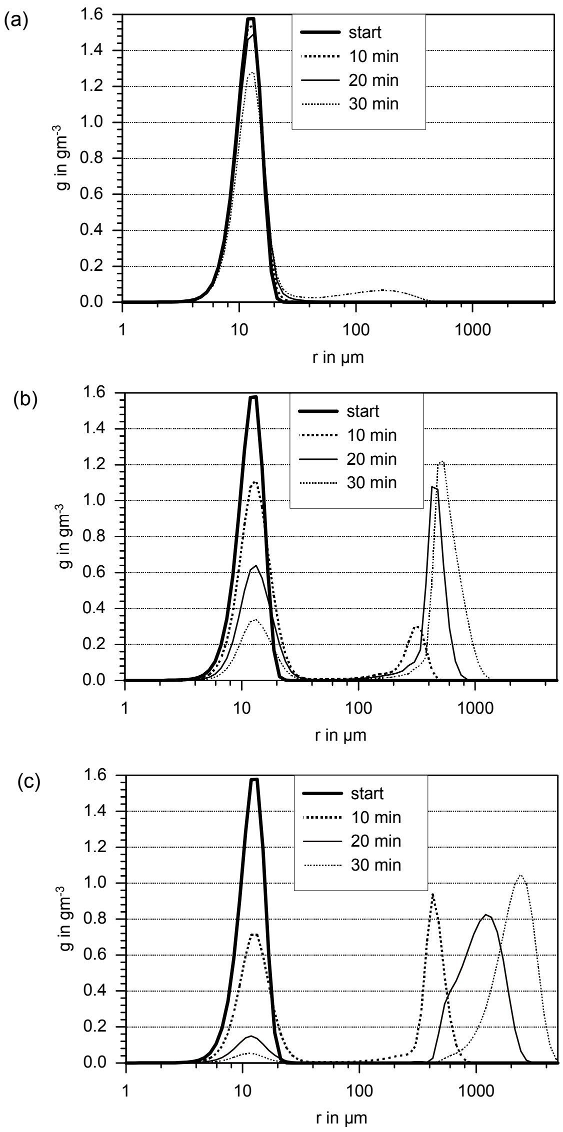

Fig. 2: (a) Temporal evolution of the mass size distribution with sedimentation kernel according to Hall (1980). (b) Same as Fig. 2(a), but for the turbulent kernel, base case. (c) Same as Fig. 2(a), but for the combined turbulent and sedimentation kernel.

The results so far show that turbulent coagulation significantly enhances the droplet growth, primarily due to the accumulation effect. To improve our approximation of real atmospheric conditions, we now also include the processes of condensation and evaporation as outlined in Section 2.

Figure 3 shows results of this second set of simulations where an aerosol distribution in an air parcel is considered that undergoes cooling. In Figure 3(a) the growth of the particles is shown when coagulation is not included. The well known narrowing of the distribution is obvious. Figure 3(b), on the other hand, includes turbulent coagulation and clearly shows a fast growth accompagnied by a broadening of the size distribution.

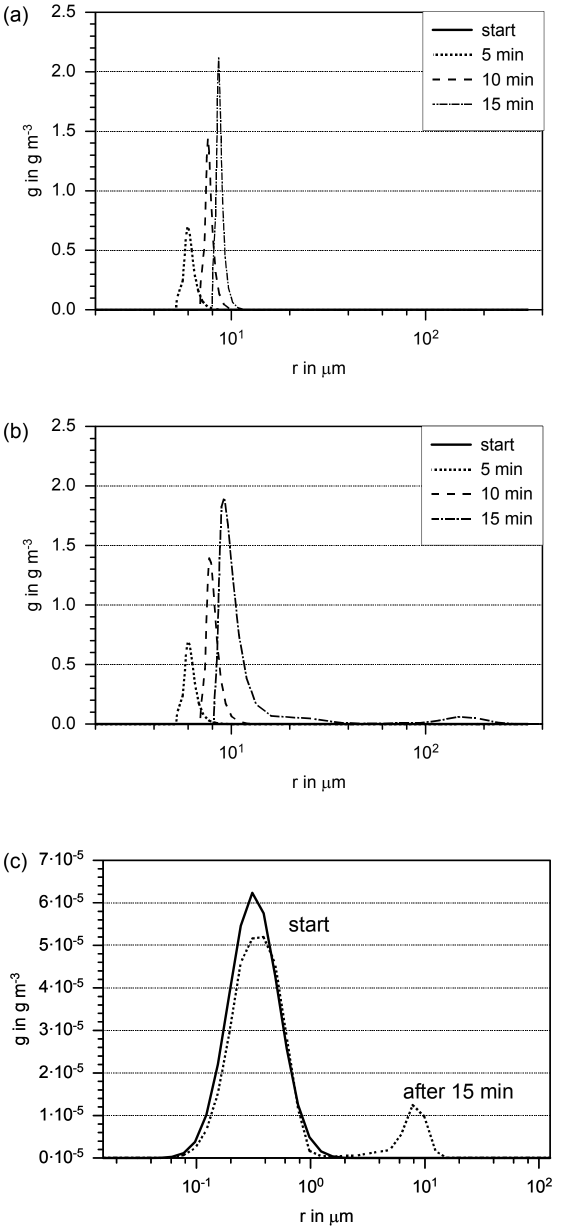

Fig. 3: (a) Temporal evolution of the mass size distribution of water, condensation only. (b) same as Fig, 3(a) but for condensation and turbulent coagulation. (c) same as Fig. 3(b) but for dry aerosol mass.

With the formation of larger droplets also aerosol mass is transferred to larger sizes. This is demonstrated by Figure 3(c) and illustrates that turbulent coagulation of cloud droplets plays a role in aerosol processing.

4. CONCLUSIONS

In this paper we investigate the impact of turbulence on the development of cloud droplet spectra. In a two-step approach we show first that – compared to the effect of sedimentation in calm air only – even moderate turbulence can enhance the formation of large droplets significantly. The largest impact of turbulence is expected for similar sized particles and/or for particles in the size range smaller than 100 μm. Here, the collision kernel is enhanced by several orders of magnitude if turbulence is included, which accelerates the growth of droplets dramatically.

By treating both coagulation and condensation we show that turbulent coagulation is also consistent with the observed broadening on the right side of the size distribution.

The presented model framework will be further extended to investigate in-cloud chemical processes and aerosol processing.

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