Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

Stochastic coalescence in Lagrangian cloud microphysics

Piotr Dziekan and Hanna Pawlowska
Institute of Geophysics, Faculty of Physics, University of Warsaw, Poland
Correspondence to: P. Dziekan (pdziekan@igf.fuw.edu.pl)

Abstract. Stochasticity in collisional growth of cloud droplets is studied in a box model using the super-droplet method (SDM). The SDM is compared with direct numerical simulations and the master equation. We use the SDM to study fluctuations in autoconversion time and the sol-gel transition. We determine how many computational droplets are necessary to correctly model expected number and standard deviation of autoconversion time. Also, growth rate of lucky droplets is determined and compared with a theoretical prediction. Size of the coalescence cell is found to strongly affect system behavior. In small cells, correlations in droplet sizes and droplet depletion affect evolution of the system. In large cells, unrealistic collisions between rain drops, caused by the assumption that the cell is well-mixed, become important. Maximal size of a volume that is turbulently well-mixed with respect to coalescence is estimated at $V_{mix} = 1.5 \cdot 10^{-2}$ cm^3. It is argued that larger cells can be considered approximately well-mixed, but only through comparison with fine-grid simulations. In addition, validity of the Smoluchowski equation is tested. Discrepancy between the SDM and the Smoluchowski equation is observed if droplets are initially relatively small. This implies that cloud models that use the Smoluchowski equation might produce rain too soon.

1 Introduction

Coalescence of hydrometeors is commonly modeled using the Smoluchowski equation (Smoluchowski, 1916), often also called the stochastic coalescence equation. It is a mean-field equation that can be derived from the more fundamental stochastic description by neglecting correlations in the number of droplets of different sizes (Gillespie, 1972; Bayewitz et al., 1974). These correlations are especially important in small volumes and neglecting them can lead to unphysical behavior. For example, when a single drop contains majority of water in a coalescence cell (gelation), the Smoluchowski equation does not conserve mass for some coalescence kernels (Leyvraz, 2003).

Another limitation of the Smoluchowski equation is that it describes evolution only of the expected number of droplets of given size. It does not contain information about fluctuations around this number, which are suspected to be crucial for precipitation onset (Telford, 1955; Scott, 1967; Marcus, 1968). Rate of collisions between droplets depends on their sizes. Small droplets rarely collide with each other, because they are repelled by disturbance flow induced by their settling. Once a droplet reaches a threshold size, it becomes more efficient at collecting smaller droplets. The mean time for a droplet to reach the threshold size is long, but some lucky droplets could reach it much sooner through a series of unlikely collisions. Then they grow quickly, resulting in a sooner onset of precipitation. This effect cannot be described using the Smoluchowski equation. The Smoluchowski equation can be written for the discrete number of droplets of given size, but often droplet concentration is

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

used instead. This adds an additional assumption that the coalescence volume is large, somewhat in agreement with neglection of fluctuations and correlations in droplet numbers (Gillespie, 1972).

A number of methods alternative to the Smoluchowski equation exist. They are capable of solving stochastic coalescence, but have some shortcomings that make their use in large-scale cloud simulations impossible. The most accurate one is the direct numerical simulation (DNS). In it, trajectories of droplets are modeled explicitly and collisions occur when they come in contact. The downside of DNS is that it is computationally extremely demanding. Running large ensemble of simulations from which statistics could be obtained would take prohibitively long time. An alternative approach is to use a master equation (Gillespie, 1972). It describes temporal evolution of probability of observing a given number of particles of a given size. Collisions are allowed between all particles in some coalescence volume and are assumed to be Markovian, i.e. they only depend on the instantaneous state of the system and not on its history. This can only be justified if the volume is well-mixed, i.e. if droplets are randomly redistributed within the volume between collisions. It is worth to note that DNS does not require such assumptions, so it reproduces correlations between positions and sizes of droplets. The master equation was analytically solved only for a constant coalescence kernel (Bayewitz et al., 1974). A more general form of the Bayewitz equation is given in Wang et al. (2006), but cannot be solved for any realistic coalescence kernel. Solving master equation numerically also proves extremely difficult due to huge phase space to be considered. Recently, Alfonso (2015) developed a method to solve the master equation numerically, but was only able to apply the method to a system of up to 40 droplets (Alfonso and Raga, 2016). Alternatively, the stochastic simulation algorithm (SSA) (Gillespie, 1975; Seeβelberg et al., 1996) can be used to model single trajectory obeying the master equation, but obtaining large enough statistics would require very long computations.

Recently, several Lagrangian schemes have been developed to model cloud microphysics in Large Eddy Simulations (Andrejczuk et al., 2008; Shima et al., 2009; Riechelmann et al., 2012). Their common point is that they explicitly model microphysical processes on a small population of computational particles, each representing large number of real particles. In this paper, we use box model simulations to investigate if the super-droplet method (Shima et al., 2009) is capable of exactly representing the stochastic nature of coalescence.

In Sec. 3 we compare super-droplet method results with master equation results for a system undergoing the sol-gel transition. Next, we validate SDM against direct numerical simulations of conversion of cloud droplets to rain drops (Sec. 4). We also determine how many computational particles are needed in SDM to obtain the correct mean behavior and correct fluctuations. In Sec. 5 we look for the minimal system size to which Smoluchowski equation can be applied. We use SDM in Sec. 6 to quantify how quickly the luckiest cloud droplets become rain drops and we compare the results with theoretical predictions. Finally, in Sec. 7, size of a well-mixed air parcel is estimated.

2 The super-droplet method

Consider N_{SD} computational particles, called super-droplets (SDs), in a well-mixed volume. Each SD is characterized by two parameters: radius r and multiplicity ξ. Multiplicity is the number of real droplets that this SD represents. Coalescence is calculated in a stochastic manner. Consider two randomly selected droplets i and j. Probability that they will collide during

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

timestep Δt is $P_{ij} = K(r_i, r_j)\Delta t/V$, where K is the coalescence kernel and V is the volume of the box. Two assumptions are made that affect amplitude of fluctuations in number of collisions. The first assumption is that SDs collide in an "all-or-nothing" manner. If a collision happens, all real droplets represented by the SD with lower multiplicity collide. The second simplification is that instead of considering all $N_{SD}(N_{SD}-1)/2$ collision pairs, only $[N_{SD}/2]$ non-overlapping pairs are randomly selected. The notation $[x]$ stand for the largest integer equal to, or smaller than x. To keep the expected number of collisions equal to the real one, coalescence probabilities are scaled up be the factor $(N_{SD}(N_{SD}-1)/2)/[N_{SD}/2]$. Intuitively, one would expect that these assumptions lead to much larger fluctuations than in the real system, because number of collision trials is artificially reduced.

We will use two types of simulations. In "one-to-one" simulations, all super-droplets have multiplicity $\xi = 1$. They are initialized by randomly drawing radii from the assumed initial distribution. Droplet collisions cause one of the SDs to be discarded. Timestep length is chosen so that none of the collision pairs has coalescence probability greater than one. This approach is similar to the Direct Simulation Monte Carlo method used in diluted gas dynamics (Bird, 1994). The second type of simulation, in which number of super-droplets is constant, is closer to the original idea of Shima et al. (2009). We will refer to it as "constant SD" simulations. In that type of simulations, the number of super-droplets is prescribed, but they have different multiplicities. To avoid large differences in initial droplet distribution between realizations, SD sizes are not completely randomly drawn from the assumed distribution as in the "one-to-one" simulations. Instead, the assumed distribution is divided into N_{SD} bins and size of a single SD is randomly selected within each bin. The first step of initialization is finding the largest and smallest initial droplet radius, r_{max} and r_{min}. They are defined by the relation $n(\ln(r_e))\Delta\ln(r)V = 1$, where r_e is either r_{max} or r_{min}, $n(\ln(r))$ is the initial droplet sizes distribution and $\Delta\ln(r) = (\ln(r_{max}) - \ln(r_{min}))/N_{SD}$. Then, within each bin of size $\Delta\ln(r)$, radius of one SD is randomly selected and its multiplicity is given by $n(\ln(r))\Delta\ln(r)V$. This procedure does not represent well the tails of the distribution, especially for large N_{SD}. Since the large tail is important for coalescence, we draw additional $\int_{\ln(r_{max})}^{\infty} n(\ln(r))\,\mathrm{d}\ln(r)$ super-droplets with $\xi = 1$ from the distribution for radii greater than r_{max}. In this type of simulations, coalescence probabilities can exceed unity. If they do, they represent multiple collisions between the pair of SDs. If multiplicity of a SD drops to zero, it is used to split the SD with largest ξ in the system into two. Super-droplets are discarded after collision only if all other SDs have $\xi = 1$.

We use an implementation of SDM from the libcloudph++ library (Arabas et al., 2015). It is an open-source project available at https://github.com/igfuw/libcloudphxx.

3 The sol-gel transition

In a system of aggregating particles, the sol-gel transition (gelation) occurs when most of the total mass is located in a single agglomerate (Leyvraz, 2003). For some forms of coalescence kernel, the Smoluchowski equation is known not to conserve mass after the transition. Alfonso and Raga (2016) presented exact solutions of the master equation for a small cloud volume undergoing the sol-gel transition. We perform simulations for the same setup as in Alfonso and Raga (2016) to test if SDM can reproduce these exact results. Consider a 1 cm^3 volume containing 20 droplets with radius of 17 µm and 10 droplets of

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

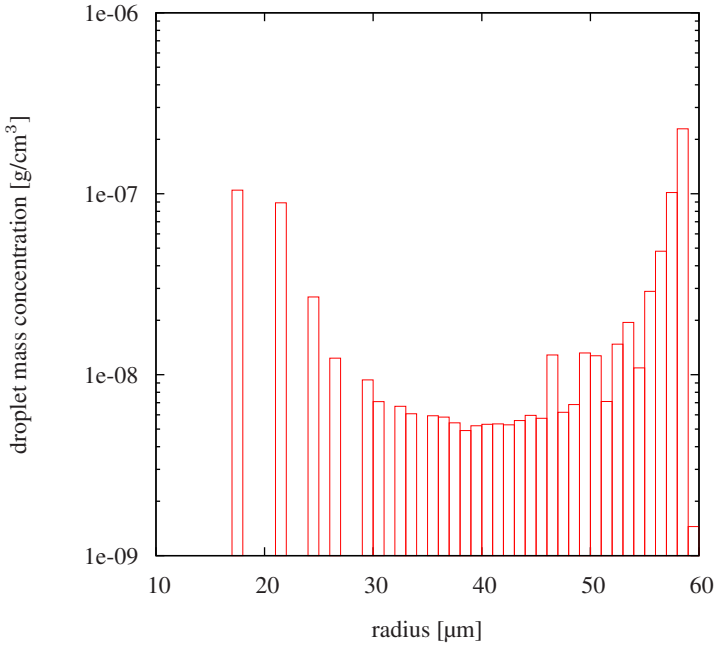

Figure 1. Mass of droplets per size bin averaged over 10^5 simulations. Bins are 1 μm wide. Shown for comparison with Fig. 8 in Alfonso and Raga (2016).

radius 21.4 μm. Gravitational collision kernel is used with collision efficiencies from Hall (1980). Droplet terminal velocities are calculated using the formula from Beard (1976).

In the "one-to-one" simulations, every real droplet is represented by a single computational droplet. That way the "all-or-nothing" simplification is removed, but the number of collision pairs is reduced. Figure 1 shows the mass distribution between droplet sizes after $t = 2500$ s averaged over an ensemble of $\Omega = 10^5$ realizations. It compares well with results presented in Fig. 8 in Alfonso and Raga (2016). This implies that the "one-to-one" SDM simulation gives correct average result of coalescence, accounting for correlations between number of droplets of each size. To check if it also gives correct fluctuations in the number of collisions, relative standard deviation of mass of the largest droplet $\sigma(m_{max})/\langle m_{max}\rangle$ is plotted in Fig. 2. The same plot obtained using master equation is shown in Fig. 7 of Alfonso and Raga (2016). Again, they compare very well, signifying that fluctuations are unaffected by the reduction in the number of collision pairs considered.

In a "one-to-one" simulation, probability $P_o dt$ that two randomly selected droplets coalesce in a short time step dt is $P_o dt = P_{pair}\eta P_{col} dt$, where P_{pair} is the probability of these two droplets belonging to the same randomly selected pair, $\eta = \frac{N_{SD}(N_{SD}-1)}{2}/[\frac{N_S D}{2}]$ is the scaling-up of probability (Shima et al., 2009) and $P_{col}dt$ is the probability of coalescence if all pairs were considered. To calculate P_{pair}, we first consider even values of N_{SD}. Consider a random permutation of droplet indices. Probability that the first droplet from the pair is at an odd position in the permutation and the second is at the next position to the right is $\frac{1}{2}\frac{1}{N_{SD}-1}$. Probability that the first is at an even position and the second is to the left of it is the same.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

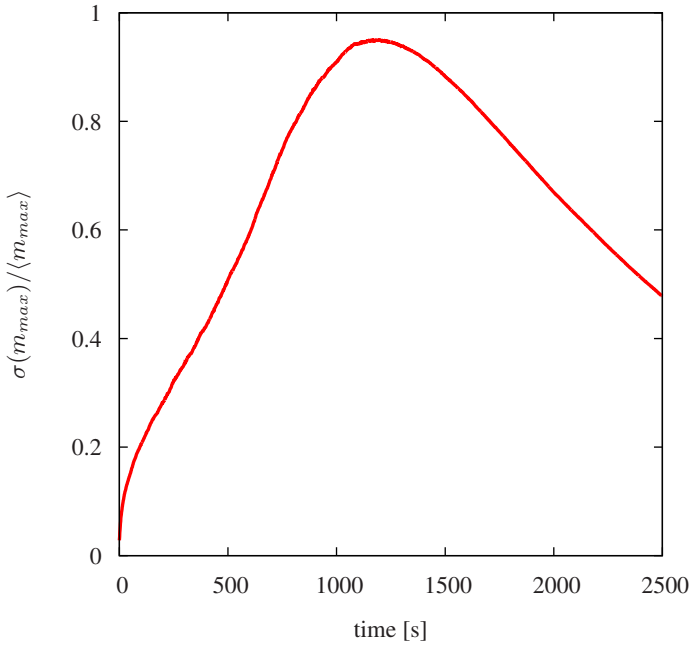

Figure 2. Relative standard deviation of mass of the largest droplet from an ensemble of 10^5 simulations. Shown for comparison with Fig. 7 in Alfonso and Raga (2016).

Summing these two we get $P_{pair}^{even}(N_{SD}) = 1/(N_{SD}-1)$. If N_{SD} is odd, the probability is $P_{pair}^{odd} = P_{pair}^{even}(N_{SD}-1)\frac{N_{SD}-2}{N_{SD}}$. We can write an expression for both odd and even cases $P_{pair} = 1/(N_{SD}-1+2*(N_{SD}/2-[N_{SD}/2]))$. It is readily obtained that $P_o = P_{col}$, i.e. that the probability of collision between any pair of real droplets is conserved in the "one-to-one" simulations.

The super-droplet method is computationally more efficient than solving the master equation directly, or using the SSA. It also puts no constraints on the initial distribution of droplets. Therefore we can use SDM to predict gelation times for larger systems and more realistic initial conditions. We use an initial droplet distribution that is exponential in mass $n(m) = \frac{n_0}{\overline{m}} exp(-m/\overline{m})$, where $n(m)dm$ is the number of droplets in mass range $(m, m+dm)$ in unit volume, $n_0 = 142$ cm^{-3} and $\overline{m}$ is the mass of a droplets with radius $\overline{r} = 15$ μm. This is the same distribution as in Onishi et al. (2015). The total initial number of droplets in the system is $N_0 = n_0 V$. Results of the "one-to-one" simulations for N_0 up to 10^6 are shown in Fig. 3. For $N_0 \geq 10^2$, the relative standard deviation of mass of the largest droplet, which quantifies amplitude of fluctuations, decreases with increasing system size. This can be understood if we look at a larger cell as an ensemble of ten smaller cells. Comparing between independent realizations, variability in the size of the single, largest droplet will be smaller if this droplet is selected from ten cells in each realization than if it was selected from only a single cell per realization. Interestingly, for $N_0 = 10^5$ an inflection point appears around $t = 500$ s. It is not seen in smaller cells. This indicates that some new source of variability is introduced. We believe that it is associated with collisions between large rain drops. We will come back to this in Sec. 5.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

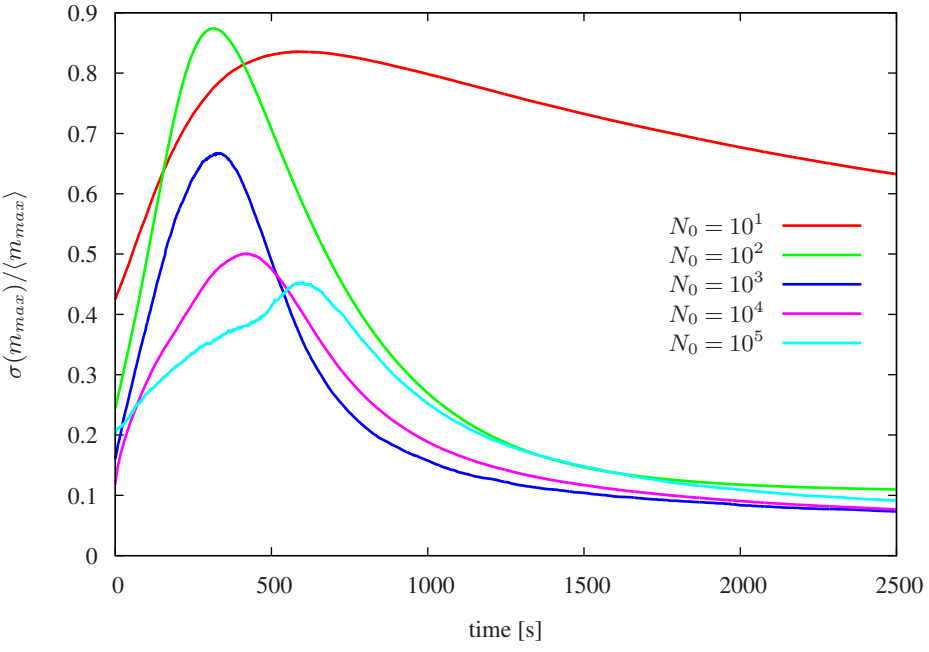

Figure 3. Relative standard deviation of mass of the largest droplet for different system sizes. Obtained from ensembles of 10^4 simulations.

The sol-gel transition time coincides with the time at which $\sigma(m_{max})/\langle m_{max}\rangle$ reaches maximum (Leyvraz, 2003; Alfonso and Raga, 2016). Intuitively, we would expect the time for most of the mass to accumulate in a single agglomerate to increase with increasing system size. This turns out to be true for systems with $N_0 > 10^3$. For system sizes $10^2 < N_0 < 10^3$ gelation time is approximately the same, around 300 s. Behavior of an extremely small system with only 10 droplets is much different. Maximum relative fluctuations are smaller and gelation time is longer than in a ten times larger system. Also, the maximum of $\sigma(m_{max})/\langle m_{max}\rangle$ is not very distinct. This is a manifestation of strong correlations in number of droplets of a given size. For example, if particles collide to form only two droplets of similar size, these two droplets may not collide for a very long time. Hence we observe large fluctuations even at $t = 2500$ s.

4 Fluctuations in conversion to rain drops

Fluctuations in time of conversion of cloud droplets to rain drops were studied using direct numerical simulations by Onishi et al. (2015). Following their notation, by $t_{10\%}$ we denote time after which 10% of mass of cloud droplets is turned into droplets with $r > 40$ µm. Droplets of this size should then quickly grow through coalescence. The time $t_{10\%}$ is used as a measure of efficiency of rain production. We will compare results of "one-to-one" simulations with DNS and try to determine how many super-droplets are needed in the "constant SD" simulations to accurately represent coalescence. The same initial droplet distribution and coalescence kernel is used as in Sec. 3.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

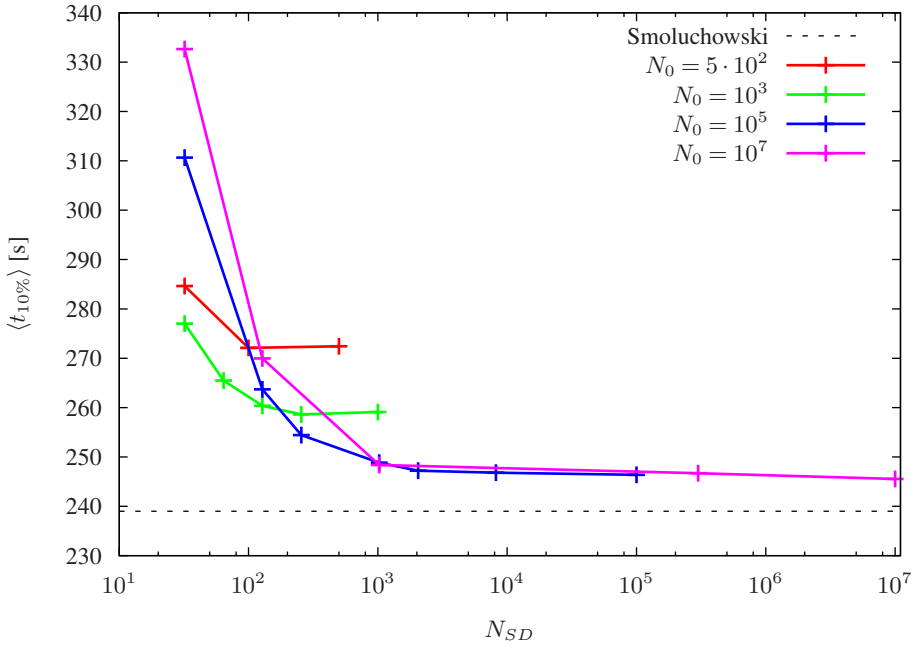

Figure 4. Mean $t_{10\%}$ from ensemble sizes $\Omega \geq 10^3$. Errorbars are smaller than plotted points. Rightmost point for each series is from "one-to-one" simulations. Other points are from simulations with various values of N_{SD}. The horizontal line is the value obtained from the Smoluchowski equation.

In Fig. 4, values of mean $\langle t_{10\%} \rangle$ for different initial number of droplets are plotted against the number of SDs. Results of both the "one-to-one" (rightmost points in each series) and "constant SD" (rest of the points) simulations are presented. Horizontal line shows the result of solving Smoluchowski equation using the flux method from Bott (1998). The "one-to-one" results converge with increasing system volume (i.e. increasing N_0). It is not clear if they would converge to Smoluchowski result, or to some higher value. The error caused by using SDs with $\xi > 1$ (in "constant SD" simulations) weakly depends on the system size. Using 10^3 SDs gives $\langle t_{10\%} \rangle$ within 1% of the "one-to-one" value. Using 10^2 SDs causes about 10% delay in rain formation. It is worth noting that on modern computers, large eddy simulations (LES) with 10^2 SDs per cell are feasible, but those with $N_{SD} = 10^3$ would be very demanding.

To analyze the amplification of fluctuations in the "constant SD" method, we plot the relative standard deviation of $t_{10\%}$ in Fig. 5. For reference, results from DNS from Onishi et al. (2015) are shown. Results from our "one-to-one" simulations are in good agreement with them. Small discrepancies are probably caused by the fact that in Onishi et al. (2015) different coalescence kernels were used for different N_0. Results of "one-to-one" simulations were fitted with a function $\alpha\sqrt{1/N_0}$ with $\alpha = 6$. Figure 5 also presents fluctuations in "constant SD" simulations for various N_{SD}. This type of simulations gives correct amplitude of fluctuations only for relatively low values of the ratio N_0/N_{SD}. For constant N_{SD}, as N_0 increases, amplitude of fluctuations correctly decreases. Then, above some critical value of the N_0/N_{SD} ratio, fluctuations stop to decrease and

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

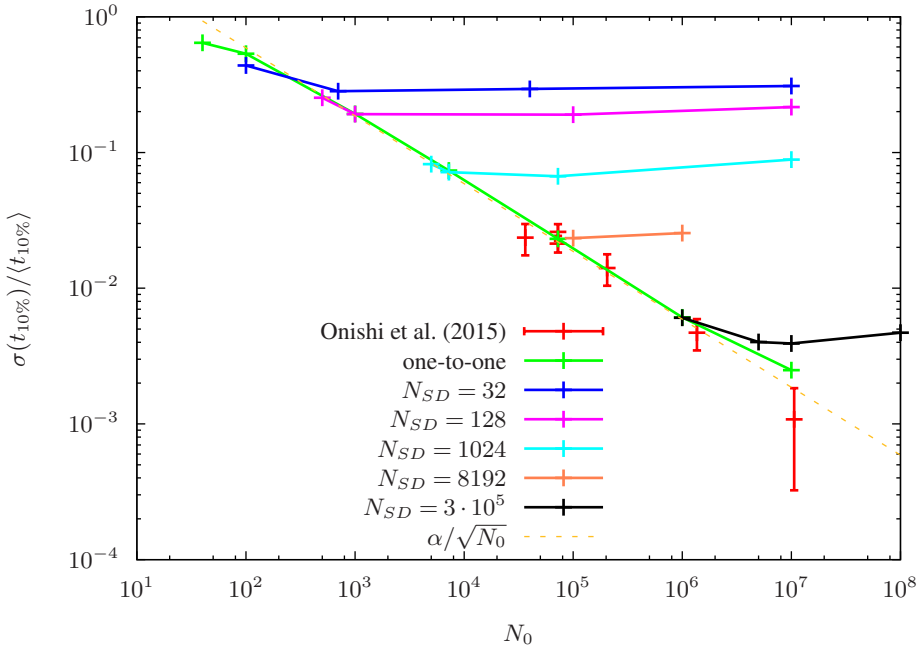

Figure 5. Relative standard deviation of $t_{10\%}$ against system size. Our results are based on samples of size of at least 10^3. Where not shown, errorbars are smaller than plotted points. Curve fitting gave $\alpha = 6$.

remain constant independent of the system size. This is a result of introducing unrealistic correlations between droplet sizes, a consequence of low number of simulational particles (Bayewitz et al., 1974). We show the limiting, minimal value of relative standard deviation of $t_{10\%}$ in Fig. 6. It decreases as $\beta\sqrt{1/N_{SD}}$, with $\beta = 2$. By comparing it with $\alpha = 6$, we conclude that in order to obtain correct fluctuations in $t_{10\%}$, number of SDs has to be $N_{SD} \geq \frac{1}{9}N_0$. Using so many SDs is not feasible in LES simulations, but is possible in smaller scale simulations. Also, knowing α and β we can estimate the magnitude of fluctuation amplification in the SDM.

5 Validity of the Smoluchowski equation

The Smoluchowski equation presents a mean-field description of the evolution of the size spectrum. It is exact only in the thermodynamic limit ($V \to \infty$). We will try to determine minimal system size for which Smoluchowski equation can be used without introducing major errors. To do so, we analyze evolution of θ, the ratio of rain water ($r \geq 40$ µm) content to the total water content. Onishi et al. (2015) denote this value by τ. We do not adopt this notation to avoid confusion with the characteristic time.

We compare results of "one-to-one" simulations with solutions of the Smoluchowski equation in two cases - with fast and with slow rain development. In both cases collision efficiencies for large droplets were taken from Hall (1980) and for small

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

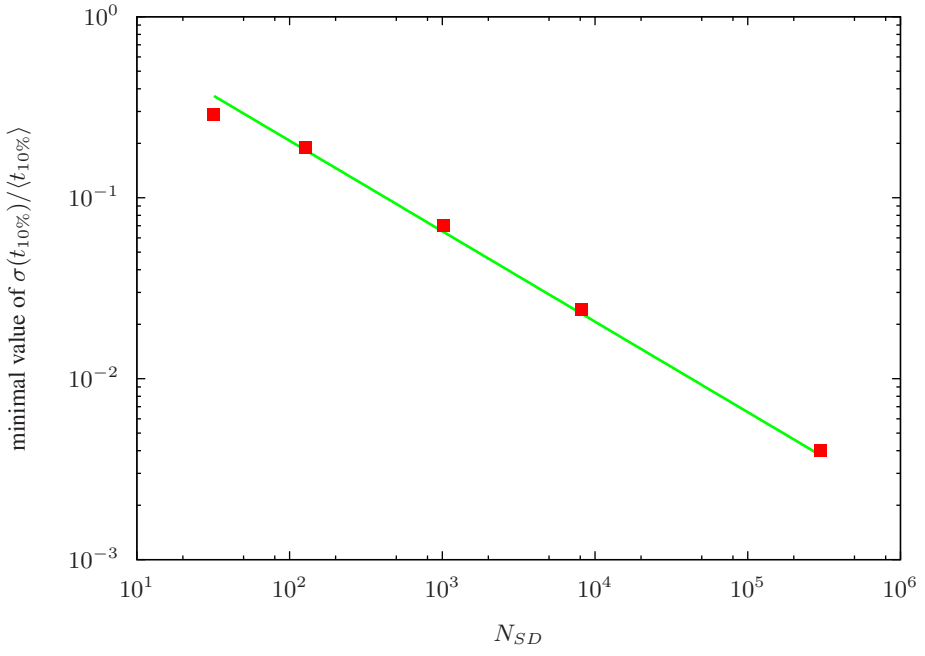

Figure 6. Minimal, limiting value of the relative standard deviation of $t_{10\%}$ for a given number of super-droplets (squares). It is calculated as an average of the points to the right of the $\alpha/\sqrt{N_0}$ curve in Fig. 5. Line is the fitted function $\beta/\sqrt{N_{SD}}$ with $\beta = 2$.

droplets from Davis (1972). In the first case, we use the same initial distribution as in Secs. 3 and 4, which results in rapid rain development. As seen in Fig. 7, the Smoluchowski equation gives correct mean rain development for systems with $N_0 \geq 10^4$. In smaller systems, rain develops slower than predicted by the Smoluchowski equation. Agreement of stochastic coalescence in large systems with the Smoluchowski equation for a similar initial distribution was shown in Seeβelberg et al. (1996). Onishi et al. (2015) present figures similar to Fig. 7, but obtained from DNS runs for $N_0 = 7.24 \cdot 10^4$ (Figs. 1(a) and 1(b) therein). They also show good agreement between model results and the Smoluchowski equation, at least up to $t = 330$ s.

The second case is well below the size gap, i.e. the range of radii for which both collisional and condensational growths are slow. We use $\overline{r} = 9.3$ µm and $n_0 = 297$ cm^{-3} as in Wang et al. (2006). In addition, we cut the distribution to 0 at $r = 20$ µm. That way we get rid of the occasional very large droplets present at $t = 0$ in some realizations. Then, rain development takes much longer and fluctuations can play a bigger role. Results are presented in Fig. 8. Surprisingly, the "one-to-one" results do not converge to the Smoluchowski results with increasing system size. We see convergence towards some other, higher value in the time it takes for rain to form. Up to $\langle\theta\rangle = 0.2$ results for $N_0 = 10^5$ are the same as for $N_0 = 10^6$, but much different from the Smoluchowski results. The latter might be affected by numerical diffusion because the simulation runs for quite long before any rain is formed. For $\langle\theta\rangle \geq 0.2$, there are discrepancies between $N_0 = 10^5$ and $N_0 = 10^6$ results. The rate of growth of $\langle\theta\rangle$ decreases earlier with increasing system size. In consequence, after $t = 5000$ s we get lower rain fraction for $N_0 = 10^5$ than for $N_0 = 10^4$, and even lower for $N_0 = 10^6$.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

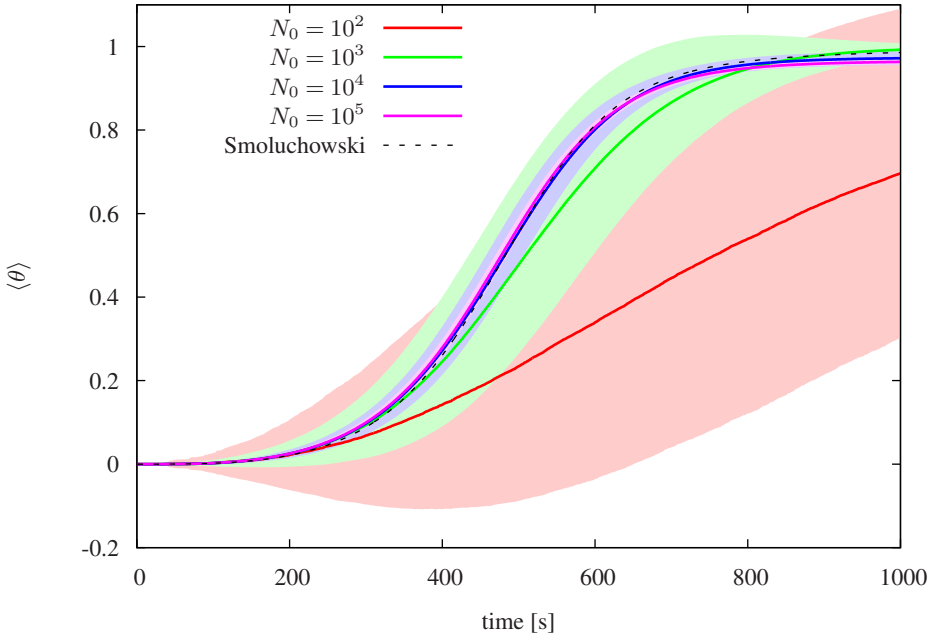

Figure 7. Rain content ratio θ for different system sizes averaged over ensembles of $\Omega = 10^4$ simulations. Shaded regions show one standard deviation interval.

The decrease of rain growth rate coincides with a decrease in concentration of rain drops n_r, as shown in Fig. 9. Number of rain drops decreases due to collisions between droplets from this category. A single droplet that results out of such collision is less efficient at scavenging cloud droplets than two pre-collision droplets. As a result, growth rate of θ decreases. In coalescence cells with $N_0 \leq 10^4$, we do not observe the decrease in number of rain drops within 5000 s, probably because sizes of rain drops are similar. For larger cells, more rain drops with a broader distribution are formed. In consequence, they collide more often which decreases their number and the rate of collection of cloud droplets. It is likely that the same effect is responsible for the additional inflection point around $t = 500$ s in the plot of relative standard deviation of the largest droplet mass for $N_0 = 10^5$ (cf. Fig. 3). This could also lead to the deviation from the $\sim 1/\sqrt{N_0}$ scaling seen in Fig. 5. Fluctuations in cells with $N_0 = 10^7$ are greater than predicted using this scaling.

Judging from Fig. 9, we conclude that Smoluchowski equation consistently overestimates the number of rain drops during the initial phase, that is when $\langle n_r \rangle$ increases. Smoluchowski equation gives up to 50% higher values of $\langle n_r \rangle$. We also observe that although the amount of rain water depends strongly on the cell size, the number of rain drops does not. In larger cells rain drops acquire larger sizes through collisions with rain drops, but rate of production of rain drops is not affected by using larger cell size. In box model simulations, the Smoluchowski equation produces too much rain if initial distribution is well below size gap and droplets slowly grow through coalescence. It is difficult to tell if using the Smoluchowski equation in cloud models

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

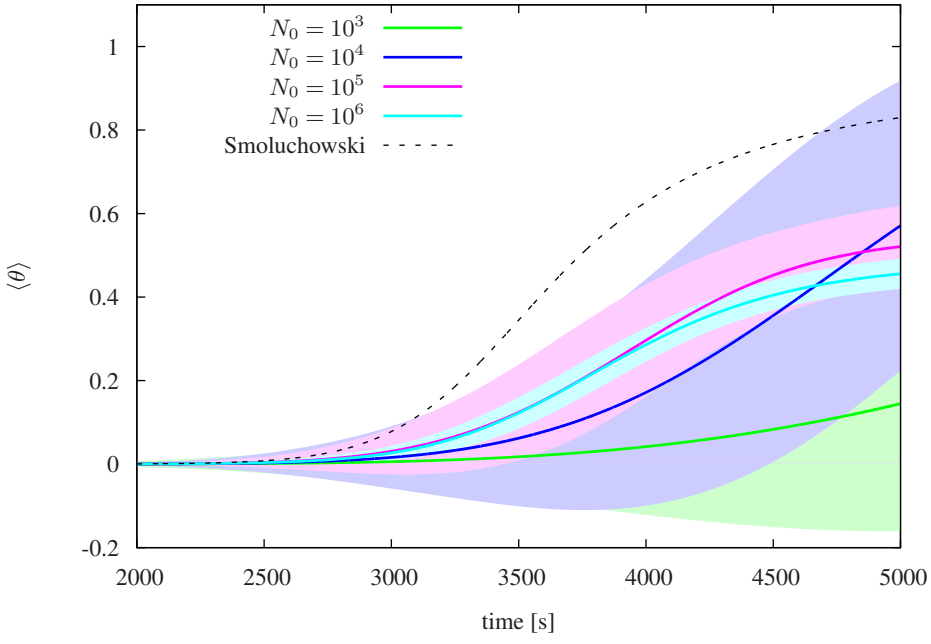

Figure 8. As in Fig. 7, but for an initial distribution with $\overline{r} = 9.3$ µm, $n_0 = 297$ cm^{-3} and a cutoff at $r = 20$ µm.

Table 1. Average, standard deviation and sample size of time (in seconds) for the lucky realizations to produce single rain droplet.

	$\gamma = 10^{-4}$			$\gamma = 10^{-3}$			$\gamma = 10^{-2}$			$\gamma = 10^{-1}$			$\gamma = 1$		
N_0	$\langle t_{40}^\gamma \rangle$	$\sigma(t_{40}^\gamma)$	$\gamma\Omega$	$\langle t_{40}^\gamma \rangle$	$\sigma(t_{40}^\gamma)$	$\gamma\Omega$	$\langle t_{40}^\gamma \rangle$	$\sigma(t_{40}^\gamma)$	$\gamma\Omega$	$\langle t_{40}^\gamma \rangle$	$\sigma(t_{40}^\gamma)$	$\gamma\Omega$	$\langle t_{40}^\gamma \rangle$	$\sigma(t_{40}^\gamma)$	$\gamma\Omega$
10^2	2052	212	10	2930	356	10	4053	517	10^2	6365	1158	10^3	14777	6099	10^3
10^3	1366	120	10^2	1762	170	10^3	2400	267	10^4	3440	505	10^5	6500	1700	10^6
10^4	1089	173	3	1336	103	10	1717	176	10^2	2354	276	10^3	3912	764	10^4
10^5	946	33	2	1090	60	20	1334	85	200	1721	169	2000	2552	415	10^4
10^6							1038	165	2	1301	176	20	1831	277	10^2

overestimates the amount of rain. Possibly, condensational growth helps droplets cross the gap, leading to initial condition that is closer to the one in the first case ($\overline{r} = 15$ µm). For such initial condition, Smoluchowski equation gives correct results.

6 Lucky droplets

There is a well-established idea that some droplets undergo series of unlikely collisions and grow much faster than an average droplet (Telford, 1955; Scott, 1967; Marcus, 1968; Robertson, 1974; Mason, 2010). These few lucky droplets are argued to be responsible for droplet spectra broadening and rain formation quicker than predicted by the Smoluchowski equation.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

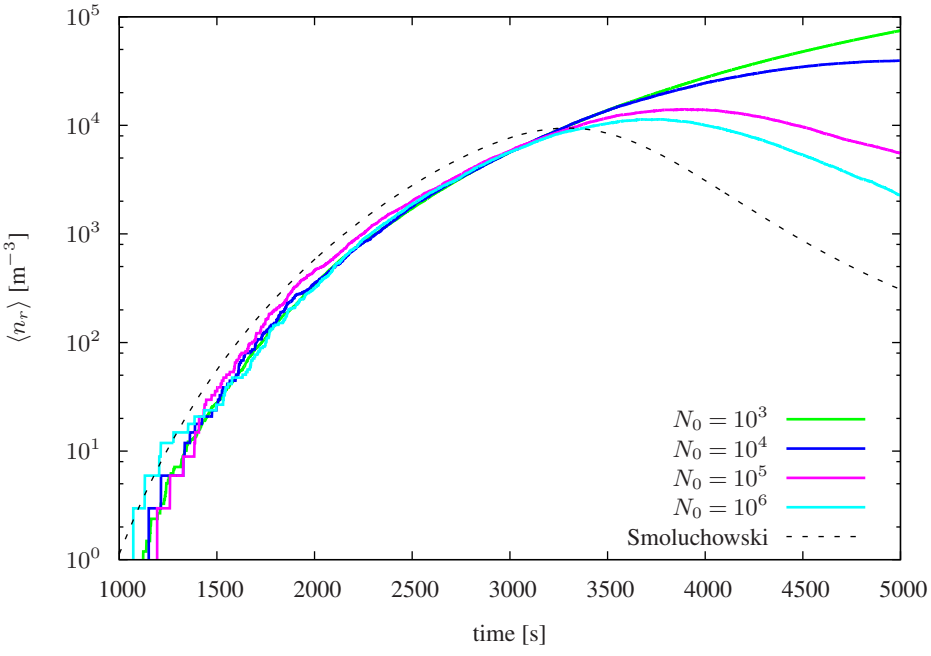

Figure 9. Mean concentration of rain drops for the same initial distribution as in Fig. 8 from ensembles of at least $\Omega = 10^4$ simulations.

Luck is supposed to be especially important during crossing of the size gap, when collisions happen rarely (Robertson, 1974; Kostinski and Shaw, 2005). A single droplet that would cross the size gap through lucky collisions could then initiate a cascade of collisions. We use the same initial distribution as in the second case in Sec. 5. The mean radius is $\overline{r} = 9.3$ µm, well below the size gap. Theoretical estimation of the "luck factor" was presented in Kostinski and Shaw (2005). We use "one-to-one" simulations to test predictions from that paper.

We are interested in time t_{40} it takes for the largest droplet in the system to grow to $r = 40$ µm. From an ensemble of Ω realizations, we select sub-ensembles of luckiest realizations, i.e. those with smallest t_{40}. We consider sub-ensembles of size $\gamma\Omega$ with $log_{10}(\gamma) = -4, -3, -2, -1, 0$. In each sub-ensemble, we calculate mean $\langle t_{40}^{\gamma} \rangle$ and standard deviation $\sigma(t_{40}^{\gamma})$. The results for different cell sizes are shown in Tab. 1. There is a large variability in $\langle t_{40}^{\gamma} \rangle$ with cell size. This is caused by the fact that t_{40} depends only on a single largest droplet. Larger cells contain more droplets, so probability of producing single large droplet increases with cell size. We notice that $\langle t_{40}^{\gamma} \rangle$ is approximately the same along the diagonals of Tab. 1. For example, cell containing 10^6 droplets on average will produce first rain droplet in 30 minutes. If we divided it into 10 cells with 10^5 droplets each, the luckiest one would also produce a droplet in 30 minutes on average. This shows that using large coalescence cells does not affect formation of first rain drops. The differences discussed in previous Sections emerge later, when there are already some rain drops that can collide with each other. Moving to very small cells, we no longer observe same $\langle t_{40}^{\gamma} \rangle$ along the diagonals. Ten cells with $N_0 = 10^2$ produce rain drops slower than a single cell with $N_0 = 10^3$. This is due to depletion

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

of water droplets in small cells. The largest droplet a cell with $N_0 = 10^2$ can produce has $r \approx 43$ µm, close to the 40 µm rain threshold.

Kostinski and Shaw (2005) estimate that the luckiest 10^{-3} fraction of droplets should cross the size gap around six times faster than average, while the luckiest 10^{-5} around nine times faster. We compare these values with our simulations for $N_0 = 10^3$. We choose this cell size, because it is the smallest one for which water depletion does not affect t_{40}. As far as t_{40} is concerned, larger cells behave exactly like an ensemble of cells of this size. We find $\langle t_{40}^{10^{-3}} \rangle / \langle t_{40}^{1} \rangle \approx 3.7$ and $\langle t_{40}^{10^{-5}} \rangle / \langle t_{40}^{1} \rangle \approx 6$. The value of $\langle t_{40}^{10^{-5}} \rangle$ was estimated at 1090 s based on values along the diagonal for larger γ and larger N_0. These ratios are lower than given in Kostinski and Shaw (2005), showing that their theoretical analysis overestimates the "luckiness" in droplet growth. Nevertheless, we agree with their conclusion that fluctuations play an important role in rain formation. Thanks to lucky collisions in some realizations (or, alternatively, in some parts of the cloud), mean concentration of rain drops after 30 minutes is about 200 m^{-3}. On the other hand, using the Smoluchowski equation leads to higher rain drop concentration than can be produced by lucky collisions (cf. Fig. 9). Significant role of fluctuations can also be seen in Fig. 8. Relative standard deviation of θ is high in small cells ($N_0 \leq 10^4$). This implies that small parts of the cloud could produce significant amount of rain much faster than average.

7 Coalescence cell size

In the previous sections we have seen that size of the coalescence cell has a profound impact on the system evolution. Possibly, many of these differences would disappear once turbulent droplet motion and sedimentation are modeled. In this Section we try to determine the size of a cell that could be used in such modeling. All methods in which each droplet within a cell can collide with any other droplet within the same cell rely on the assumption that the cell is well-mixed. This includes the master equation, SSA, SDM as well as the Smoluchowski equation. The assumption that a cell is well-mixed is valid if $\tau_{mix} \ll \tau_{coal}$, where τ_{coal} and τ_{mix} are the characteristic times for coalescence and cell homogenization, respectively (Lehmann et al., 2009; Gillespie et al., 2014). By well-mixed we mean that droplets should be distributed homogeneously within the cell before every collision. Droplet coalescence generates inhomogeneities, i.e. correlations between droplet positions and sizes. Consider two droplets growing independently within a cell. After gaining large sizes, they collide and generate even larger droplet. In reality they could not both obtain large sizes before the collision, because they would deplete liquid water from each other's surrounding.

Rigorously, characteristic time for coalescence is the mean time between coalescence events, as in diffusion-limited chemical systems (Gillespie et al., 2014). To estimate its magnitude, consider a single large collector droplet falling through a field of smaller droplets. Using geometric coalescence kernel with efficiency E, the mean time between collisions is $\tau_{coal} = (E\pi(r_l + r_s)^2 v_r n_s)^{-1}$, where r_l and r_s are radii of large and small droplets, v_r is the relative velocity and n_s is the concentration of small droplets. For $r_l = 100$ µm, $r_s = 10$ µm, $v_r = 70$ cm/s, $E = 1$ and $n_s = 100$ cm^{-3} we get $\tau_{coal} \approx 0.4$ s.

Droplets in the cell can be mixed through turbulence. Turbulence acts similarly to diffusion and its characteristic time for mixing is $\tau_{mix}^t = (V^{(2/3)}/\varepsilon)^{(1/3)}$, where V is cell volume and ε is turbulent energy dissipation rate (Lehmann et al., 2009).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

Turbulent energy dissipation rate in clouds is in the range from 10 $\mathrm{cm}^2/\mathrm{s}^3$ for stratocumulus clouds to 10^3 $\mathrm{cm}^2/\mathrm{s}^3$ for cumulonimbus clouds (Malinowski et al., 2013; Grabowski and Wang, 2013). Let us assume that $\tau^t_{mix} \ll \tau_{coal}$ is satisfied if $\tau^t_{mix} = 0.1\tau_{coal}$. Even in the most turbulent clouds, this means that the coalescence cell has to be very small $V \approx 1.5{\cdot}10^{-2}\mathrm{cm}^3$. On average, this volume would contain around one droplet, depending on concentration of droplets. The Smoluchowski equation cannot be used for such small populations. Using super-droplet microphysics would be very cumbersome, because extremely short time steps would be required to decouple motion from collisions. To use larger cells, we need to choose some less strict value of characteristic time of coalescence. For instance, Shima et al. (2009) assume $\tau_{coal} = 100$ s without much explanation. Some larger cell size, that would be approximately well-mixed, could be found phenomenologically through exact simulations including droplet motion. One example of such reference simulations are DNS runs from Onishi et al. (2015) discussed in Sec. 5. They prove that in the case with $\overline{r} = 15$ µm, Smoluchowski equation gives correct results. This suggests that cells with $N_0 \geq 10^4$ can be used in this case.

Another process that can mix droplets is sedimentation. It is difficult to assess its timescale, because it strongly depends on droplet sizes. Droplets of similar sizes are not mixed by sedimentation, but it is efficient at mixing rain drops with cloud droplets. We can expect that it would prevent depletion of cloud droplets in the surrounding of a rain droplet that was observed for smallest cells in Secs. 3 and 6. Sedimentation acts only in one direction, so it could only allow us to use cells larger only in the vertical direction.

8 Conclusions

The super-droplet method can exactly represent stochastic coalescence. It was compared with the master equation approach (see Sec. 3) and with direct numerical simulations (see Sec. 4). Precision of the SDM is controlled by the number of super-droplets used. Fluctuations in the autoconversion time are represented well if $N_{SD} \geq N_0/9$. Using smaller N_{SD} increases standard deviation of autoconversion time by a factor $\frac{1}{3}\sqrt{N_0/N_{SD}}$ (cf. Sec. 4). It is computationally less expensive to correctly reproduce mean autoconversion time. Using $N_{SD} = 10^3$ gives mean results within a 1% margin, while using $N_{SD} = 10^2$ - within 10%.

The SDM was used to study stochastic coalescence for two initial droplet size distributions - with small ($\overline{r} = 9.3$ µm) and with large ($\overline{r} = 15$ µm) droplets. They result in slow and fast rain formation, respectively. Dependence of the system behavior on coalescence cell size was observed, especially in the small droplets case. Cell size not only affects fluctuations in observables, but also their expected value. If the coalescence cell is small ($N_0 < 10^3$), sizes of droplets are strongly correlated and depletion of cloud water plays an important role. In reality, these two effects are probably not manifested, because collector drop sedimentation acts against them. If the coalescence cell is relatively large ($N_0 > 10^4$), rain drops that in reality would form far from each other and would need time to get close, can collide immediately. This is because the coalescence cell is assumed to be well-mixed, which is usually not true. We estimate a well-mixed (with respect to coalescence) volume in the most turbulent clouds to be only $1.5 \cdot 10^{-2}$ cm^3.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

Unrealistic collisions between rain drops, caused by the assumption that coalescence cell is well-mixed, do not affect results if droplets are initially large. Then, collisions of cloud and rain drops and between cloud droplets are frequent, so relatively rare collisions between rain drops are not important. The mean behavior of the system converges to the Smoluchowski equation results with increasing system size. Good agreement with it is found for systems with $N_0 \geq 10^4$. The picture is different if droplets are initially small. Conversion of cloud droplets into rain drops is slow, so decrease in rain drop concentration due to these unrealistic collisions is relatively more important. Coalescence of rain drops decreases the rate of collection of cloud droplets, because a single larger drop has smaller collisional cross section than two smaller drops with the same total volume. In consequence, mean behavior of the system no longer converges with increasing cell size. Up to $\langle\theta\rangle = 0.2$, results for $N_0 = 10^5$ are the same as for $N_0 = 10^6$. Then, rate of rain growth decreases sooner in the larger coalescence cell. Another aspect of the slow-coalescence scenario is that in it, some lucky droplets can grow much faster than average droplets. We found that a single luckiest droplet out of a thousand grows 3.7 times faster than average and the luckiest out of a hundred thousand - 6 times faster. These values are smaller than predicted by Kostinski and Shaw (2005), but large enough to be important for quick formation of rain.

The size of a well-mixed volume, i.e. a volume within which droplets are randomly rearranged through turbulence between coalescence events, is of the order of the volume occupied by a single droplet. Larger cells can be assumed to be only approximately well-mixed. For example, in the fast-coalescence case, DNS modeling gives the same results as the Smoluchowski equation (Onishi et al., 2015). Box model simulations using well-mixed volume with $N_0 = 10^5$ droplets also gives the same results. Therefore it can be assumed that such volume is approximately well-mixed in the case of fast coalescence. On the other hand, in the slow-coalescence case, box model simulations do no converge to the Smoluchowski result. This implies that models that use the Smoluchowski equation might produce rain too soon. The real behavior of the system could be determined through DNS modeling or SDM simulations with droplet motion.

Cells used in LES are typically ten orders of magnitude larger than a well-mixed volume. They do not necessarily have to be well-mixed. It is sufficient if they are homogeneous, i.e. they are an ensemble of identical, approximately well-mixed sub-cells. Some statistical moments for such ensembles were presented in this work. In general, it is not clear what could be the size of these sub-cells and if the Smoluchowski equation is valid for them. We have shown that for initially small droplets, the Smoluchowski equation gives wrong results, but is correct for initially large droplets. One could hope that condensational growth leads to initial conditions close to the ones for which the Smoluchowski equation is valid, but justifying it would require further research.

9 Code availability

Simulation code is available at https://github.com/pdziekan/coal_fluctu. The libcloudph++ library is available at https://github.com/igfuw/lib

Acknowledgements. This study was financed from Poland's National Science Center "POLONEZ 1" grant 2015/19/P/ST10/02596 (this project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Curie grant agreement No. 665778) and Poland's National Science Center "HARMONIA 3" grant 2012/06/M/ST10/00434. Numerical simulations were carried out at the Cyfronet AGH computer center, accessed through the PLGrid project. We are grateful to Wojciech W. Grabowski for fruitful discussions.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-314, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 19 April 2017

Atmospheric Chemistry and Physics Discussions

References

Alfonso, L. (2015). An algorithm for the numerical solution of the multivariate master equation for stochastic coalescence. *Atmospheric Chemistry and Physics*, 15(21):12315–12326.

Alfonso, L. and Raga, G. B. (2016). The impact of fluctuations and correlations in droplet growth by collision-coalescence revisited. part i: Numerical calculation of post-gel droplet size distribution. *Atmospheric Chemistry and Physics Discussions*, 2016:1–33.

Andrejczuk, M., Reisner, J., Henson, B., Dubey, M., and Jeffery, C. (2008). The potential impacts of pollution on a nondrizzling stratus deck: Does aerosol number matter more than type? *Journal of Geophysical Research: Atmospheres*, 113(D19).

Arabas, S., Jaruga, A., Pawlowska, H., and Grabowski, W. (2015). libcloudph++ 1.0: a single-moment bulk, double-moment bulk, and particle-based warm-rain microphysics library in c++. *Geoscientific Model Development*, 8(6):1677–1707.

Bayewitz, M. H., Yerushalmi, J., Katz, S., and Shinnar, R. (1974). The extent of correlations in a stochastic coalescence process. *Journal of the Atmospheric Sciences*, 31(6):1604–1614.

Beard, K. (1976). Terminal velocity and shape of cloud and precipitation drops aloft. *Journal of the Atmospheric Sciences*, 33(5):851–864.

Bird, G. (1994). *Molecular gas dynamics and the direct simulation monte carlo of gas flows*. Clarendon, Oxford.

Bott, A. (1998). A flux method for the numerical solution of the stochastic collection equation. *Journal of the atmospheric sciences*, 55(13):2284–2293.

Davis, M. H. (1972). Collisions of small cloud droplets: Gas kinetic effects. *Journal of the Atmospheric Sciences*, 29(5):911–915.

Gillespie, D. T. (1972). The stochastic coalescence model for cloud droplet growth. *Journal of the Atmospheric Sciences*, 29(8):1496–1510.

Gillespie, D. T. (1975). An exact method for numerically simulating the stochastic coalescence process in a cloud. *Journal of the Atmospheric Sciences*, 32(10):1977–1989.

Gillespie, D. T., Petzold, L. R., and Seitaridou, E. (2014). Validity conditions for stochastic chemical kinetics in diffusion-limited systems. *The Journal of chemical physics*, 140(5):02B604_1.

Grabowski, W. W. and Wang, L.-P. (2013). Growth of cloud droplets in a turbulent environment. *Annual Review of Fluid Mechanics*, 45:293–324.

Hall, W. D. (1980). A detailed microphysical model within a two-dimensional dynamic framework: Model description and preliminary results. *Journal of the Atmospheric Sciences*, 37(11):2486–2507.

Kostinski, A. B. and Shaw, R. A. (2005). Fluctuations and luck in droplet growth by coalescence. *Bulletin of the American Meteorological Society*, 86(2):235–244.

Lehmann, K., Siebert, H., and Shaw, R. A. (2009). Homogeneous and inhomogeneous mixing in cumulus clouds: Dependence on local turbulence structure. *Journal of the Atmospheric Sciences*, 66(12):3641–3659.

Leyvraz, F. (2003). Scaling theory and exactly solved models in the kinetics of irreversible aggregation. *Physics Reports*, 383(2):95–212.

Malinowski, S., Gerber, H., Plante, J.-L., Kopec, M., Kumala, W., Nurowska, K., Chuang, P., Khelif, D., Haman, K., et al. (2013). Physics of stratocumulus top (post): turbulent mixing across capping inversion. *Atmospheric Chemistry and Physics*, 13(24):12171–12186.

Marcus, A. H. (1968). Stochastic coalescence. *Technometrics*, 10(1):133–143.

Mason, B. J. (2010). *Physics of clouds*. Clarendon Press.

Onishi, R., Matsuda, K., and Takahashi, K. (2015). Lagrangian tracking simulation of droplet growth in turbulence–turbulence enhancement of autoconversion rate. *Journal of the Atmospheric Sciences*, 72(7):2591–2607.

Riechelmann, T., Noh, Y., and Raasch, S. (2012). A new method for large-eddy simulations of clouds with lagrangian droplets including the effects of turbulent collision. *New Journal of Physics*, 14(6):065008.

Robertson, D. (1974). Monte carlo simulations of drop growth by accretion. *Journal of the Atmospheric Sciences*, 31(5):1344–1350.

Scott, W. T. (1967). Poisson statistics in distributions of coalescing droplets. *Journal of the Atmospheric Sciences*, 24(2):221–225.

Seeβelberg, M., Trautmann, T., and Thorn, M. (1996). Stochastic simulations as a benchmark for mathematical methods solving the coalescence equation. *Atmospheric Research*, 40(1):33–48.

Shima, S.-i., Kusano, K., Kawano, A., Sugiyama, T., and Kawahara, S. (2009). The super-droplet method for the numerical simulation of clouds and precipitation: A particle-based and probabilistic microphysics model coupled with a non-hydrostatic model. *Quarterly Journal of the Royal Meteorological Society*, 135(642):1307–1320.

Smoluchowski, M. V. (1916). Drei Vortrage uber Diffusion, Brownsche Bewegung und Koagulation von Kolloidteilchen. *Zeitschrift fur Physik*, 17:557–585.

Telford, J. (1955). A new aspect of coalescence theory. *Journal of Meteorology*, 12(5):436–444.

Wang, L.-P., Xue, Y., Ayala, O., and Grabowski, W. W. (2006). Effects of stochastic coalescence and air turbulence on the size distribution of cloud droplets. *Atmospheric Research*, 82(1):416–432.