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Interactive Comment

Interactive comment on "The validity of the kinetic collection equation revisited – Part 3: Sol-gel transition under turbulent conditions" *by* L. Alfonso et al.

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Reply to anonymous referee #1

First, we would like to thank the anonymous referee for his/her comments that will improve the quality of our paper. Our revised version will include several of his/her suggestions.

Anonymous referee # 1

1) Referee comment: "Sol-gel transition" is only a new name, and it is unclear what aspect of this concept is novel to the rain formation problem. Contact with work done

in two other fields, physical chemistry (gelation) and astrophysics (planet formation) is worthy of notice but does not suffice. Can the scientific novelty be expressed in a precise way?"

Reply: As pointed out correctly the referee, the problem of the influence of stochastic fluctuations in the formation of precipitation in warm cloud has been addressed with different approaches by a large number of authors. On the other hand, it is widely accepted now that turbulence enhances the rate of particle collisions.

However, no methods exist as proposed for estimating the time of formation of the drop which triggers the coalescence process for realistic collection kernels (modified by turbulence) initial conditions typical of warm clouds and using the pure stochastic Monte Carlo method developed by Gillespie (1975). That is the novel aspect of our approach.

Additionally, the problem of sol-gel transition has been treated in detail by specialists in astrophysics and physical chemistry, with interesting developments in recent years. The application of these concepts (runaway droplet, sol-gel transition) to the problem of precipitation formation in warm clouds is another contribution of our work.

Despite the referee's opinion, the interpretation of the formation of precipitation by collection in warm clouds in terms of a sol-gel transition is not familiar to most specialists in our area. The application of these concepts allows for a rich duality with other areas of knowledge.

To fully clarify this point in the revised version of the paper we will discuss this point in more detail.

2. Referee comment: "The relevance of fluctuations compared to mean particle growth and runaway formation has been discussed extensively, for example by Langmuir (1948), Telford (1955), Robertson (1974), Gillespie (1975), Young (1975), Kostinski Shaw (2005), Wang et al.(2006), and others (including Bayewitz et al. 1974, already

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cited in the paper). Several of these authors treat the sudden conversion of cloud water to rain water and its dependence on stochastic fluctuations. Again, what is new here is not at all clear."

Reply: There are important differences between our work and the works mentioned by the referee. Although all the works mentioned address in some way the influence of stochastic fluctuations in the process of precipitation formation, approaches and scope of each are very different.

In general, the average spectrum obtained from the kinetic collection equation (KCE) and the ensemble averages spectrum obtained over different realizations of the stochastic collection process, are different. The solution to the KCE and the expected values calculated from the Monte Carlo method are equal only if the covariances are omitted from the probabilistic model. Although the term stochastic has been associated with that equation for historical reasons, it is clearly deterministic and has no stochastic correlations or fluctuations included. This is the quasi-stochastic approach (Gillespie, 1975a) which ignores fluctuations in the drop mass spectrum.

Most of the works cited by the referee use the quasi-stochastic approach (Telford, 1955; Robertson, 1974; Young, 1974; Kostinski and Shaw, 2005) and are stochastically rather incomplete. The pure stochastic approach was only used in the papers by Gillespie (1975), Wang et al. (2006) and Bayewitz et al. (1974) with their deduction of a master equation. However, in the works of Gillespie (1975) and Bayewitz (1974) a constant collection kernel was used to derive the results.

Gillespie (1975a) shows that three growth models are possible, depending upon the physical interpretation given to the quantity A(m,m')N(m')dt, where A(m,m′) is the collection kernel, N(m') is the droplet concentration and dt is an infinitesimal interval of time: the continuous model, the quasi-stochastic and the pure stochastic.

Below I will detail the differences between the works outlined by the referee:

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a) In Telford (1955) the quasi-stochastic approach was adopted (See Gillespie, 1975a). According to this approach, the magnitude: A(m,m')N(m')dt = fraction of the drops of mass m which will collect a droplet in time dt, which differs from the interpretation of the pure stochastic model. In the quasi-stochastic model, the state of the drops are described by the function N(m,t), which gives the number of drops having mass m at time t. This approach ignores fluctuations in the drop mass spectrum. On the other hand, reliable information on the collision efficiencies were not available, and Telford assume E=1 in his calculations.

b) In Robertson's (1974) basically the same approach as Telford's (1955) was used. In that case a Monte Carlo procedure was employed in order to calculate the collection process. As in Telford's paper, they consider an idealized cloud of constant volume and assumed that only drop-droplet interactions are permitted. His method for picking the time between coalescence events is not stochastically complete.

c) The pure stochastic model with the first exact stochastic simulation algorithm was presented by Gillespie (1975). In the pure stochastic model the magnitude: A(m,m')N(m')dt = the probability that any drop of mass m will collect a drop a droplet in time dt. This requires focusing not on the average drop volume spectrum which appears in the kinetic collection equation (KCE) but rather on the coalescence probability density function. The algorithm was more accurate than all the previously proposed Monte Carlo procedures and take proper account of correlations that are ignored in the derivation of the KCE, a problem that was addressed by Bayewitz et al. (1974) in more detail. However, in Bayewitz et al. (1974) and Gillespie (1975) a constant collection kernel was assumed for the analysis.

d) Kostinski and Shaw (2005) published a simplified version of Telford's analysis to illustrate the role of stochastic fluctuations in accelerating the growth of a few lucky droplets. However, as Telford's (1995) approach, their description is stochastically incomplete since coalescence events occur randomly its rates vary in space, time and with realizations. Consequently, the modeling of the size distribution must consider the

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stochastic nature of the collection process.

e) In Wang et al.(2006), the pure stochastic approach was adopted (Gillespie, 1975a). Monte Carlo simulations were performed for a turbulence collection kernel with the Monte Carlo method of Gillespie. The master equation (for the probabilities) and the full stochastic collection equation were derived for the hydrodynamic kernel. In Bayewitz et al. (1974) this equation was derived but only for the special case of a constant collection kernel.

f) In our work, we follow the pure stochastic model (Gillespie, 1975a). As remarked in the pure stochastic model the magnitude A(m,m')N(m')dt = the probability that any drop of mass m will collect a drop a droplet in time dt. For this case, a fully stochastic collision-coalescence calculation was performed using a modification of the Monte Carlo method of Gillespie (Laurenzi et al., 2002) which inherently incorporates all stochastic correlations presented in the collection process. This computational procedure is rigorously based on the probability, instead of the kinetic collection equation. Additionally, the time of formation of the lucky droplet that becomes the embryo for raindrops is estimated from Monte Carlo simulations for turbulence conditions and realistic initial conditions.

In the revised version of the paper we will include a detailed comparison with previous works in order to clarified this point.

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Laurenzi, I.J., Bartels, S.L., and Diamond, S.L.: A general algorithm for exact simulation of multicomponent aggregation, J. Comput. Phys., 177, 418, 2002.

3.Referee comment: " It is okay if the novelty is in the results of the simulations themselves, but that is not clear either. A dependence of gelation time on the coalescence rate (the collision kernels) is hardly surprising. Apparently the primary result of the simulations is that an increase in the collision kernel leads to earlier formation of a runaway droplet.Whether the increase is due to turbulence or some other effect is not demonstrated. Furthermore, the conclusion that “ratio _ never reaches its maximum, confirming that the sol-gel transition does not take place under these conditions” is probably incorrect. The accompanying figure seems to indicate that it will take place, but at longer times than were simulated (consistent with the lower kernels)."

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Reply: In the present paper, a bimodal droplet distribution was considered: 150 droplets of 10 μm in radius and another 150 droplets of 12.6 μm in radius, corresponding to liquid water content (LWC) of 1.9 gm-3. As shown by simulations, in this case, the collision efficiencies are too small to allow development of drizzle drops in times less than an hour.

In summary, we could not obtain a phase transition for time intervals relevant to the problem under discussion by using the collection kernel without turbulence effects.

As the referee correctly points, an increase in the collection rate leads to the formation of a runaway droplet. However this can be accomplished in two ways. By increasing the collection kernel (considering collection efficiencies modified by turbulence effects), or by modifying the initial distribution of droplets but assuming unrealistic initial conditions quite different from those in real clouds.

4. Referee Comment: "The paper emphasizes the importance of stochastic element, but the turbulence contribution is introduced through a deterministic collision kernel. This appears inconsistent, or at very least incomplete".

Reply: There is no contradiction since in the pure stochastic approach the collection kernel is interpreted in terms of a probability:

a(i,j)=K(i,j)NixNj/V=Pr{ Probability that two unlike particles i and j with populations (number of particles) Ni and Nj will collide within the imminent time interval} (1)

The time of the next collision and the indexes of the colliding droplets are then calculated randomly from probability density functions constructed from (1). This method was originally developed by Gillespie (1975) inherently incorporates all stochastic correlations presented in the collection process. In the revised version a complete description of the method will be included in the appendix.

As in any Monte Carlo method the expression for the distribution functions are "deterministic". The random nature of the process arises from the random number generation

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using these “deterministic” distribution functions.

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