México City, September 25, 2019

Reply to Reviewer:

First, we would like to thank the anonymous referee for his/her comments that surely will improve the quality of our paper.

1) **Referee Comment:**

My primary contention is that the authors don't show why the said hypothesis (authors call it sol-gel hypothesis) will lead to a particular frequency distribution of the size of the biggest droplets or the shown size distribution is unique to only this hypothesis. Second, the connection between observed frequency distribution in fog data and simulations is tenious.

Reply to reviewer:

In Botet and Płoszajczak (2005), the Kolmogorov-Smirnov distribution was found as the exact largest cluster distribution for critical mean-field percolation. In the same paper, by means of detailed numerical simulations of the Smoluchowski equation with a product kernel, the authors conjectured that the Kolmogorov-Smirnov distribution is the largest particle distribution at sol-gel transition time. This results are for infinite systems.
To the best of my knowledge, there is no similar theoretical result for the largest cluster distribution at critical point in finite systems. For finite systems, in a previous paper (Botet, 2011), it was demonstrated (by means of detailed numerical simulations for multiplicative coagulation), that the distribution (1) is a good approximation to the empirical largest particle distribution for all values of θ.

$$f(x,\theta,\mu_1,\beta,\mu_2,\sigma) = \theta Gumbel(x,\mu_1,\beta) + (1-\theta) Gauss(x,\mu_2,\sigma) \quad (1)$$

Our approach is based on numerical simulations, and analysis of observations in order to check whether or not the largest droplet radius will follow a mixture of Gumbel and gaussian distributions. One aspect that strengthens our hypothesis is that we used formal statistical tests in order to check whether or not the observed distribution follow or not the mixture of distributions, with good results. These points are analyzed in more detailed below.

Results for finite systems:

It was hypothesized that the largest droplet mass (for finite systems) is distributed following an admixture of distributions (Gumbel and Gaussian), because in an early stage of cloud formation (disordered or statistical phase), fluctuations and correlations are negligible, there only a few collision events, and droplets are randomly distributed. According to extreme value theory, in a system without correlations, the random variable $s_{max} = \max_{i=1,\ldots,m} s_i$, where s_i denotes the size of the droplet number i, and there are m droplets in the system), is distributed following an asymptotic $\Phi_{max}(s_{max})$, which must be either Gumbel, Frechet or Weibull distribution (Gumbel, 1958). Examples of distributions belonging to the domain of attraction of the Gumbel distribution are the exponential and the Gaussian distribution, a fact that explain the ubiquity of the Gumbel distribution in extreme value theory applications.

For an uncorrelated system, no other distributions can appear for the extremal variable. At later times, away from the pseudocritical region (which is the finite system equivalent of a sol-gel transition time) the Central Limit theorem applies, and the corresponding probability distribution must be Gaussian (provided correlations be negligible). The equation (1) will work for the uncorrelated case (early stage of collision-coalescence process), but with small (close to 0) or large (close to 1) values of θ. In the presence of correlations, in the pseudocritical region (which is the equivalent of a phase transition for a finite system) the values of θ will be in the vicinity of 0.5, and $f(x,\theta,\mu_1,\beta,\mu_2,\sigma)$ takes a non-trivial form, which is well fitted by Eq. (1).

In our paper, the Kolmogorov-Smirnov goodness of fit test were performed in order to check whether or not the distribution (1) fits our data. The admixture of distributions was always a better model than the Gumbel and Gaussian distributions for all sample sizes, with large p-values. So, in the majority of the cases, the null hypothesis (the sample comes from a mixture of distributions) could not be rejected.

Gruyer et al. (2013), in a very different context, found that the distribution of the largest fragments in nuclear multifragmentation reactions (simultaneous break-up of an excited nuclear system in several fragments) have o nontrivial form which is well fitted by the function (1). The context is different, but the physical interpretation is similar. At higher and lower energies, fluctuations are negligible, and the largest fragment is distributed Gumbel or Gaussian. At intermediate energies (with larger correlation), there is an admixture an admixture of the two asymptotic distributions.

Results for infinite systems:

In theory of critical phenomena, little is known about the distribution of the largest fragment at the critical point. However, there is a remarkable result in mean-field percolation theory (Botet and Płoszajczak, 2005), showing (from theoretical backgrounds), that the largest cluster distribution at the critical point is the Kolmogorov-Smirnov (K-S) distribution. The K-S distributionis the distribution of the maximum value of the deviation between the experimental realization of a random process and its theoretical cumulative distribution and it has the cumulative distribution:

$$K_1(z)=\sum_{k=-\infty}^{\infty}(-1)^k\, e^{-k^2\pi^2 z/6} \qquad (2)$$

Or the equivalent expression:

$$K_1(z)=\sqrt{\frac{6}{\pi z}}\sum_{k=-\infty}^{\infty} e^{-3(2k+1)^2/(2z)} \qquad (3)$$

In Botet and Płoszajczak, 2005 it was also conjectured, by means of numerical simulations, that the largest particle distribution at sol-gel transition time for the Smoluchowski model (Eq. 4) with a product kernel $K(i,j)=Cx_i x_j$ is also the Kolmogorov-Smirnov distribution (1,2).

$$\frac{\partial N(i,t)}{\partial t}=\frac{1}{2}\sum_{j=1}^{i-1} K(i-j,j)N(i-j)N(j)-N(i)\sum_{j=1}^{\infty} K(i,j)N(j) \qquad (4)$$

We must remember that, for equation (4), there is a sol-gel transition at $T_{gel}=\left[CM_2(t_0)\right]^{-1}$,

where $M_2(t)=\sum_{i=1}^{N_d} x_i^2 N(i,t)$ is the second moment of droplet mass spectrum.

2) Referee Comment:

Third, sufficient details are not provided to ascertain whether the frequency distribution as the sum of two distributions with a particular value of weighting factor is better than say if only one of the distribution (that is weighting factor either zero or one) was fitted.

Reply to referee:

The results with the Kolmogorov-Smirnov goodness of fit test are shown in Table 1 of the paper. As can be observed in Table 1, for 100 randomly selected samples with different sample sizes, the admixture is always a better model than the Gumbel and Gaussian distributions for all sample sizes.

The fact that the Gumbel distribution fits so poorly the data is because most of the samples are in the pseudo-critical region. Effectively, Figure 8 of the paper shows that for 90% of the samples the ratio $\eta=\frac{w_{Gaussian}-w_{Gumbel}}{w_{Gaussian}+w_{Gumbel}}$ lies in the interval [-0.9, 0.9], clearly indicating that 90% of the samples are on the pseudocritical domain.

Table 1. For each sample size, number of samples with the null hypothesis H_0 rejected at $\alpha=0.05$ for all the distributions.

Case	Total number of random samples	Sample size	Fitted Distributions	At α=0.05 Reject H_0 (Number of Samples)
1	100	100	Mixture	13
			Gumbel	92
			Gaussian	35
2	100	200	Mixture	27
			Gumbel	96
			Gaussian	58
3	100	300	Mixture	35
			Gumbel	98
			Gaussian	70

4	100	400	Mixture Gumbel Gaussian	40 100 77
5	100	500	Mixture Gumbel Gaussian	50 100 83

In the revised version we will make more emphasis on this problem.

References:

Botet, R.: Where are correlations hidden in the distribution of the largest fragment?. *PoS*, 007, 2011.

Botet, R. and Płoszajczak, M.: Exact order-parameter distribution for critical mean-field percolation and critical aggregation. *Physical Review Letters*, 9*5*(18), 185702, 2005.

Gruyer, D., Frankland, J. D., Botet, R., Płoszajczak, M., Bonnet, E., Chbihi, A., ... and Guinet, D.: Nuclear multifragmentation time scale and fluctuations of the largest fragment size. *Physical review letters*, *110*(17), 172701, 2013.