Analytical Results for Nontrivial Polydispersity Exponents in Aggregation Models

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We study a Smoluchowski equation describing a simple mean-field model of particles moving in $d$ dimensions and aggregating with conservation of ‘mass’ $s = R^d$ ($R$ is the particle radius). In the scaling regime the scaled mass distribution $P(s) \sim s^{-\tau}$, and $\tau$ can be computed by perturbative and non perturbative expansions. A possible application to two-dimensional decaying turbulence is briefly discussed.

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Aggregation phenomena are widespread in nature. They have such an impact on material sciences, chemistry, astrophysics, that a large amount of literature was devoted to them. In such dynamical processes, particles or objects as different in geometry and size as colloidal particles, galaxies, small molecules, droplets, polymers, can merge to form a new entity when they come into close contact or interpenetrate, through diffusion (Brownian coagulation), ballistic motion (ballistic agglomeration), droplet growth and coalescence or droplet deposition.

We are usually interested in the evolution of the statistical distribution of the ‘mass’ $s$, a quantity characteristic of each particle, that is conserved in the coalescence process: it can be either the real mass, the volume, the area, the electric charge, or any other parameter depending on the underlying physics.

Great advance was achieved when it was proposed and observed both in real experiments and in numerical simulations that the distribution $N(s,t)$ exhibits scale invariance at large time:

$$ N(s,t) \sim S(t)^{-\beta} f \left( \frac{s}{S(t)} \right) $$

where the characteristic mass $S(t)$ diverges as $t^z$ when $t \to \infty$, ensuring the oblivion of initial conditions and physical cut-off or discreteness, as does the diverging correlation length of critical phenomena: universality arises in dynamics as well, with new universality classes.

The exponents $z$ and $\beta$ are easily deduced from conservation laws and physical arguments, but in many cases a polydispersity exponent $\tau$ defined by $f(x) \sim x^{-\tau}$ when $x \to 0$ is observed, whose value is nontrivial though universal. The prediction of $\tau$ is still a challenge.

The earliest tool for tackling the problem is still one of the most popular, that is the Smoluchowski equation. It is a master equation for the distribution $N(s,t)$:

$$ \frac{\partial N(s,t)}{\partial t} = \frac{1}{2} \int N(s_1,t)N(s-s_1,t)K(s_1,s-s_1)ds_1 - N(s,t) \int N(s_1,t)K(s,s_1)ds_1 $$

where the aggregation kernel $K(x,y)$ is symmetric and characteristic of the physics of the aggregation process on a more or less coarse-grained level. Such kinetic equations are usually derived within a mean-field approximation, but in certain cases it is possible to go beyond mean-field limitations investigated by van Dongen and Ernst who classified the kernels according to their homogeneity and asymptotic behavior:

$$ K(bx,by) = b^\lambda K(x,y) $$

For a given physical system, the homogeneity $\lambda$ is easily determined using scaling arguments. We consider only nongelling systems with $\lambda \leq 1$.[14] For $\mu > 0$, the exponent $\tau$ is trivial and found to be $\tau = 1 + \lambda$, whereas for $\mu = 0$, $\tau$ depends on the whole solution $f$ of the scaling equation derived from Eq. (2) (see Eq. (3) below). $\mu < 0$ does not lead to any power law behavior but rather to a bell-shaped scaling function $f$. In the following, we shall focus on the $\mu = 0$ case for which the exponent $\tau$ has been so far only determined numerically by direct simulation of Smoluchowski equation (not an easy task)[15], by time series[16], and of course by direct simulation of the physical system described by the considered Smoluchowski equation. Considering the ubiquity and the importance of the $\mu = 0$ case leading to nontrivial polydispersity exponents, analytical results would be certainly welcome.

The purpose of this Letter is to show, working with a physically relevant simple kernel, that some information about $\tau$ can be extracted from the kernel itself using exact bounds, estimates and expansions around exactly solvable kernels. We compare our results to numerical studies in the literature and briefly discuss a possible original application to two-dimensional decaying turbulence.
Consider hyperspherical particles in a $d$-dimensional box, of polydisperse radii $R$ with distribution $F(R,t)$, evolving the following way: at time $t$ we choose the positions of their centers with uniform probability in $d$-space. Then each pair of overlapping spheres of radii $R_1$ and $R_2$ merges to form a new sphere of radius,

$$R = (R_1^D + R_2^D)\frac{1}{2}$$

(5)

where $D$ is a parameter with $D \geq d$. $D$ can be the actual dimension of the spheres, as for instance in the case of $D = 3$ spheres deposited on a $d = 2$ plane [3]. Once each coalescence has been resolved, we have reached $t + 1$.

The conserved variable is $s = R^D$ and the corresponding kernel for the equation ($\beta$) is $K(x,y) = (x^\frac{D}{2} + y^\frac{D}{2})^d$.

This kernel has been introduced in many contexts from molecular coagulation [12] to cosmology [15, 17] for specific values of $d$ and $D$, and is one of the most studied in the literature [4, 14, 18] although very few analytical results are known. This kernel has $\lambda = \frac{D}{2}$ and $\mu = 0$. From the conservation law we get $\left(\frac{D}{2}\right) = 2$ and if we plug the scaling form into (2) we get $z = D/(D-d)$ for $d < D$ and:

$$sf'(s) + 2f(s) = f(s)f_0^+ f(s_1)(s_1^\frac{D}{2} + s_1^\frac{D}{2})^d ds_1$$

$$- \frac{1}{2} \int_0^s f(s_1)f(s-s_1)(s_1^\frac{D}{2} + (s-s_1)^\frac{D}{2})^d ds_1$$

(6)

If $\tau \geq 1$ each term of the RHS of Eq. (6) is separately divergent and they should be properly grouped [3, 13].

In $d = 0$ or $D = \infty$, Eq. (6) reduces to the constant kernel equation with exact solution $f_0(x) = 2e^{-s}$ and $f_\infty(s) = 2^{1-d}e^{-s}$. In the case $d = 1, D = 1$ an exact analytic solution is also known for the time dependent equation, with $\tau = 3/2$ [3].

Now, for given $d$ and $D$, and plugging the exact small s behavior $f(s) \sim s^{-\tau}$ into Eq. (6), one first gets that $\tau < 1 + \lambda = 1 + d/D$. Then, matching the behavior of both sides of Eq. (1) [4, 14, 13], one finds:

$$\tau = 2 - \int_0^\infty f(x) x^{\frac{D}{2}} dx$$

(7)

If $\alpha > \tau - 1$ we obtain by multiplying Eq. (1) by $x^\alpha$ and integrating [4, 14, 13]:

$$2(1-\alpha) \int x^\alpha f(x) dx = \int f(x)f(y)(x^\frac{D}{2} + y^\frac{D}{2})^d [x^\alpha + y^\alpha - (x+y)^\alpha] dxdy$$

(8)

By studying the large s behavior of Eq. (1), one can show that $f(s)$ decays as $c_\infty s^{-\frac{D}{2}}e^{-s}$ with $c_\infty = f_0^{1/2}(x^\frac{D}{2} + (1-x)^\frac{D}{2})^d x^{-\frac{D}{2}} (1-x)^{-\frac{D}{2}} dx$.

Exact bounds and estimates - We first show that $\tau \geq 1$ if $d \geq 1$. Suppose $\tau < 1$ and consider Eq. (1) with $\alpha = 0$. For $d \geq 1$, we have $(x^\frac{D}{2} + y^\frac{D}{2})^d \geq x^\frac{D}{2} + y^\frac{D}{2}$, which leads to $\int f(x) dx \leq \int f(x) dx \int f(x) x^\frac{D}{2} dx$, hence $1 \geq 2 - \tau$, which is contradictory. Notice that Eq. (8) with $\alpha = 2$ for $d = 1$ and $D = 1$ leads to $\int x^2 f(x) dx = 2(\int x^2 f(x) dx) (\int f(x) dx)$, and we recover the exact result $\tau = 2 - \int f(x) dx = 3/2$ in a very simple way.

We now introduce an extremely simple method of getting lower and upper bounds for $\tau$. We rely on Eq. (8) valid for $\alpha > \tau - 1$. Combining Eq. (1) and (8), we get:

$$\tau = 2 - (1-\alpha) \frac{\int g(x,y) dxdy}{\int g(x,y) A(x,y) dxdy}$$

(9)

where $A(u) = (1 + u^\alpha - (1 + u)^\alpha)/(1 + u^\frac{D}{2})$, $u = (x^\frac{D}{2} + y^\frac{D}{2})^d/(x^\alpha + y^\alpha)$ satisfies $A(u) = A(1/u)$ and $g(x,y) = (x^\alpha y^\alpha + x^\frac{D}{2} y^\alpha) f(x)f(y)$. The ratio in Eq. (9) can then be interpreted as the inverse of a kind of average of $A(x,y)$ with the weight $g(x,y)$. For a given $\alpha \leq d/D$, we determine numerically the lower and upper bounds $m_\alpha$ and $M_\alpha$ of the function $A(u)$. Using Eq. (9), this gives $2 - (1 - \alpha)/m_\alpha \leq \tau \leq 2 - (1 - \alpha)/M_\alpha$. We then choose the best values of $\alpha \leq d/D$ compatible with $\alpha > \tau - 1$ leading to the tightest bounds. This allows us to greatly improve the exact bounds given in [4, 15] for $d = 1$ and to obtain new such bounds for $d > 1$. For instance for the physically interesting cases (see below) ($d = 1, D = 2$), ($d = 1, D = 4$) and ($d = 2, D = 4$) we respectively found $1.084 \leq \tau \leq 1.147$, $1 \leq \tau \leq 1.075$ (compared to $1 \leq \tau \leq 1.28$ and $1 \leq \tau \leq 1.109$ in [15]) and $1.25 \leq \tau \leq 1.5$.

It is also possible to obtain good estimates by evaluating the ‘average’ in Eq. (9) using a reasonable trial weight function $g(x,y)$ instead of the unknown exact one. A parameter free choice is obtained by replacing in the above expression of $g(x,y)$ the exact $f(x)$ by $x^{-\tau} \exp(-x)$ which has the correct leading asymptotic for small $x$ (by definition of $\tau$) and the expected exponential large $x$ decay [13, 14]. This form is known to be a good approximation of the actual $f(x)$ obtained in simulations [15], and is even obtained in exactly solvable models [14]. The simplest method is to determine $\tau$ self-consistently from Eq. (1), for instance with $\alpha = d/D$, but the result depends on the chosen $\alpha$ and may even violate exact bounds. A much better and hardly more intricate method is to choose a sample of values of $\alpha$, and minimize an error function measuring the violation of the corresponding Eq. (1) [13]. This method can be systematically improved by allowing $n$ free ‘fitting’ parameters (including $\tau$ itself) in the trial weight $g(x,y)$. Using the simplest $\chi^2$ form for the error function with a trial function $f(x) = (x^{-\tau} + x^{-\frac{D}{2}}) e^{-x}$ (to take into account the exact decay at large $x$), we obtain [13]: $\tau \approx 1.111$, $\tau \approx 1.016$ and $\tau \approx 1.431$ in the three cases considered above. The case $d = 1$, $D > 1$ has been numerically investigated by means of time series in [10]. The authors of [10] found $\tau \approx 1.11 \sim 1.12$ for $D = 2$ (using data inading the text of [10] and to $\tau \approx 1.06$ for $D = 4$ (as roughly extracted from Fig. 3 in [14]), in fair agreement with our
bounds and estimates. We shall find later that our estimate for \( d = 2 \) and \( d = 4 \) is in good agreement with a non-perturbative calculation for \( \tau \) and with a perturbative estimate. Our variational method requires very few CPU time and is straightforwardly implemented compared to methods used in \([13,14]\).

**Perturbative expansions for \( d < 1 \) -** Now we use the exactly solvable limits \( d = 0 \) and \( D = \infty \) as a basis for a perturbative expansion.

First, we consider the limit \( d \to 0 \). We expand \( f \) in series of \( d \): \( f(x) = f_0(x) + df_1(x) + O(d^2) \). A systematic way of expanding \( \tau \) would be to write down a linear (self-consistent) differential equation for \( f_1 \) to solve it and plug the result into Eq. (6). This method is used in \([13]\) to compute the next order \( O(d^2) \). However, as far as the first order is concerned we can get it without solving for \( f_1 \). By developing the integral expression of \( \tau \), Eq. (8), we get:

\[
\tau = 2 - \int f(x) x^2 dx = -d/D \int f_0(x) x dx - d \int f_1 + O(d^2).
\]

Now we develop both sides of Eq. (8) with \( a = 0 \) to get an equation for \( \int f_1 \): \( \int f_1 = \frac{1}{2} \int f_0(x)f_0(y) (x^2 + y^2) dx dy - (\int f_0) (\int f_1) \), hence \( \int f_1 = -\int e^{-x-y} (x^2 + y^2) dx dy \). After a simple calculation we get:

\[
\tau = 2d \int_0^1 \ln \left( 1 + \left( \frac{1-u}{u} \right)^{\frac{1}{2d}} \right) du + O(d^2). \tag{10}
\]

Let us mention that we can generalize this result to any homogeneous kernel of the form: \((g(x,y))^d\), leading to:

\[
\tau = 2d \int_0^1 \ln g(1, \frac{1-u}{u}) du + O(d^2).
\]

For \( D = 1 \), we get \( \tau = 2d + O(d^2) \), in good agreement with direct numerical integration of Smoluchowski’s equation performed by Krivitsky \([15]\), who obtained \( \tau \approx 0.2 \) for \( d = 0.1 \) and \( \tau \approx 0.38 \) for \( d = 0.2 \). This result for \( D = 1 \) also coincides up to order \( O(d) \) with the best inequalities for \( \tau \) that we obtained above (as already observed in \([13]\)), but not for other values of \( D \) \([13]\).

The order \( O(d^2) \) requires the computation of \( f_1 \) which satisfies a solvable linear second order equation. This cumbersome calculation will be presented in \([13]\). However, in the special case \( D = 1 \) it is possible to obtain explicitly the \( O(d^2) \) term by expanding Eq. (8) for \( a = d/D \). We obtain:

\[
\tau = 2d + \frac{\pi^2}{2} \cdot d(1-d) + O(d^2). \tag{11}
\]

For \( d = 0.2 \), we get \( \tau \approx 0.372 \) compared to the already mentioned \( \tau \approx 0.38 \), whereas for \( d = 0.4 \), we get \( \tau \approx 0.686 \) compared to \( \tau \approx 0.7 \) found numerically in \([13]\).

Now, we perform an expansion in powers of \( 1/D \) for \( d \leq 1 \), expanding \( f(x) = f_\infty(x) + \frac{1}{D} f_1(x) + \frac{1}{D^2} f_2(x) + O(1/D^3) \). We use exactly the same method: we develop Eq. (8) with \( a = 0 \) in powers of \( 1/D \), and plug the result in Eq. (6), yielding a vanishing first order term and a nontrivial second order term:

\[
\tau = 2 - 2^{1-d} + \frac{\pi^2 2^{d-1}(1-d)}{12 D^2} + O\left( \frac{1}{D^3} \right). \tag{11}
\]

Once again we were able to obtain a highly nontrivial expansion of \( \tau \) without solving for \( f_1 \) and \( f_2 \) themselves, although this can also be achieved this way \([13]\). Note that in the limit of large \( D \) and small \( d \), Eq. (11) and (13) coincide up to order \( O(d/D^2) \).

**Perturbative estimate for \( d > 1 \) -** In the case \( d \geq 1 \), we have shown that \( \tau \geq 1 \) and since \( \tau = 1 + \varepsilon \), we see that \( \tau \to 1 \) for \( D \to \infty \) and finite \( d > 1 \). The previous perturbation is not valid because \( f_1 \) is non integrable. Nevertheless we can try to obtain an estimate of \( \tau \) in the following way: we make the ansatz \( f \sim f_\infty + c_s \varepsilon^{1+\sigma} \), when \( s \to 0 \). We plug it into Eq. (6) and Eq. (8) for \( a = d/D \), and after some algebra \([13]\) we see that for consistency \( \varepsilon \) must be of order \( 1/D \) and that \( c = (1 - 2^{1-d}) (d/D - \varepsilon) \), and eventually that \( \varepsilon = \kappa/D + O(1/D^2) \) where \( \kappa \) is the solution of the nonlinear equation:

\[
\frac{2}{1 + 2^{1-d}} = \int_0^1 (1 + u^{\varepsilon/D})^d du \tag{12}
\]

This equation always has a solution consistent with the exact bound \( 1 < \tau < 1 + d/D \). For instance in the case \( d = 2, D = 4 \) we obtain \( \tau \approx 1.462 \), which is highly consistent with the previous estimate \( \tau \approx 1.431 \). Though it is still of order \( 1/D \), the obtained perturbative estimate depends on the choice of \( a \). \( a = d/D \) seems however to be the most natural choice.

In \( d = 1 \), \( c \) vanishes and we do not learn much. Notice that we have shown that all terms of the \( d < 1 \) series for \( \tau \) in powers of \( 1/D \) vanish for \( d \to 1 \), as can be seen in Eq. (11) for the two leading ones. Thus, the correction to \( \tau \) is for large \( D \) may be **non perturbative** in \( d = 1 \), which would again rule out the estimate \( \tau = 1 + 1/2D \) of \([20]\), which also violates our rigorous inequalities \([13,14]\).

If we now take the \( d \to \infty \) limit in Eq. (12), we obtain:

\[
\tau \approx 1 + \lambda - 2^{-d} \lambda (\lambda = d/D), \text{ a non perturbative behavior in } d \text{ which is to be related to the results below.}
\]

**Large \( d \) and \( D \) -** We now present a non perturbative calculation in the limit of large \( d \) and \( D \), keeping the ratio \( \lambda = d/D \) fixed. In this limit, the kernel can be written:

\[
(x^2 + y^2)^d = 2^d (x y)^{\frac{d}{2}} (1 + O(d/D^2)) \tag{13}
\]

and surprisingly transforms into the celebrated ‘product’ kernel \([14,10,20]\). Assuming scaling (a still controversial subject \([14,13]\)), one can easily show that \( \tau = 1 + \lambda = 1 + d/D \) \([14]\) (see also Eq. (8) and (11) and the discussion below them, as it corresponds to \( \mu = \lambda/2 > 0 \)). We have shown that including higher order corrections in power of \( 1/D \) does not change the value of \( \tau \) such that the correction to \( \tau = 1 + \lambda \) is certainly non perturbative. In fact, we can estimate this correction by assuming that for finite \( d \) and \( D \), \( f(s) \sim c_\lambda s^{1+\lambda-\sigma} \) for \( s \to 0 \). Plugging this estimate in Eq. (7) with the limit kernel of Eq. (11), we first get \( s^d \approx 2^{-d} c_\lambda/(1 - \lambda) \). \( c_\lambda \) can be determined by matching the coefficients of the leading terms in Eq. (7).
using the kernel of Eq. (13). After a straightforward calculation, one gets \( c_\lambda \) in the \( d \to \infty \) limit:

\[
\tau = 1 + \lambda - 2^{1-d} I_\lambda^{-1}, \quad c_\lambda = 2(1-\lambda)I_\lambda^{-1}
\]

(14)

\[
I_\lambda = \int_0^1 [u(1-u)]^{-1-\lambda/2} [u^{\lambda} + (1-u)^{\lambda} - 1] \, du
\]

We thus find a non-perturbative (exponentially small) correction to \( \tau \) in the large \( d \) and large \( D \) limit, consistent with the result obtained above for \( d > 1 \) and large \( D \). Note that Eq. (14) is also consistent with the exact result that \( \tau \to 1 + D/D \) for finite \( d > 1 \), a result that we obtain by setting \( \lambda = 0 \) (as \( I_\lambda \) diverges). In the case \( d = 2 \) and \( D = 4 \) of interest below, we already gave the estimate \( \tau \approx 1.431 \), whereas Eq. (14) leads to \( \tau \approx 1.428 \).

Physical applications of these results to massive particle aggregation systems and the generalization to other physically relevant kernels (as the one applying to the systems described in [7,8]) will be presented elsewhere. In this Letter, we would like to present an original application outside of this field of massive particle aggregation, namely the dynamics of vortices in two-dimensional decaying turbulence.

Recently, a statistical numerical model has been introduced [21,22] which describes the dynamics and the merging of vortices with the assumption that the typical core vorticity \( \omega \) and the total energy \( E \sim \int u^2 \, d^d x \sim \sum_i \omega_i^2 R_i^d \) are conserved (\( R_i \) is the radius of the \( i \)-th vortex) throughout the merging processes. This model reproduces the main features observed in direct numerical simulations (see [21,22] for details). For instance, after noting that a distribution of vortex radii satisfying \( P(R) \sim R^{-\beta} \) is equivalent to a Gaussian energy spectrum \( E(k) \sim k^{-\beta-6} \) [22], the simulation of this model was able to reproduce the fact that starting from a Batchelor spectrum \( E(k) \sim k^{-3} \) (\( \beta = 3 \)), the system evolves systematically to a steeper spectrum \( E(k) \sim k^{-\gamma} \) with \( \gamma = 6 - \beta \) in the range \( \gamma \approx 3 \to 5 \) [22].

Now, one expects that the collision kernel between two vortices is somewhat intermediate between the ballistic hard-disk form \( \sigma \sim (R_1 + R_2) \) [14], and the totally uncorrelated form \( \sigma \sim (R_1 + R_2)^2 \) (where the probability of colliding is proportional to the probability that two randomly placed vortices overlap, see also below Eq. (5)) [13]. Thus, one can describe approximately the decay of vortices due to mergings by means of Eq. (5) with \( 1 \leq d \leq 2 \) and \( D = 4 \), as two colliding vortices merge into a new one with \( R = (R_1^d + R_2^d)^{1/d} \) in order to conserve energy and core vorticity. One thus expects a power law radius distribution \( P(R) \sim R^{-\beta} \), with \( \beta = D(\tau - 1) + 1 \) and \( \tau \) given by our model. We find values of \( \gamma \) ranging from \( \gamma \approx 3.3 \) for \( d = 2 \) (taking \( \tau \approx 1.43 \)) to \( \gamma \approx 4.94 \) (taking \( \tau \approx 1.016 \)) for \( d = 1 \), in good qualitative agreement with observed exponents. As also found in direct simulations, the actual exponent (and here the value of the effective correct \( d \)) could depend on the actual initial conditions (\( \omega \) area occupied by the vortices \( \sim \) enstrophy). Note that the limit Batchelor case \( \gamma = 3 \), is obtained when taking the naive strict upper bound \( \tau = 1 + D/D \) with \( d = 2 \) and \( D = 4 \).

In conclusion, we have introduced a systematic scheme to obtain exact bounds and good estimates for the polydispersity exponent in aggregation models. We have also implemented perturbative and non-perturbative expansions found to be in good agreement with already known numerical results when available. Finally, this kind of calculations generalizes to other interesting kernels, with possible physical applications in the field of droplet deposition [5,13] or even two-dimensional decaying turbulence as briefly mentioned in the present Letter.

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