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LETTER TO THE EDITOR

On the conservation of volume during particle coagulation

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Abstract. If $(\partial n/\partial t)_c$ is the coagulational rate of change of the distribution function $n(v)$ for a system of particles, it is shown that $\int_0^\infty v(\partial n/\partial t)_c \, dv$ is not necessarily zero. Rather, depending on the coagulation kernel and the behaviour of $n(v)$ as $v \to \infty$, it may be zero, non-zero and finite, or infinite. This has implications for the asymptotic behaviour of the solution of the steady state, time-independent coagulation equation. The results are applied to Brownian and gravitational coagulation.

Consider a system of coagulating particles described by a distribution function $n(v, t)$. Then the rate of change of n due to coagulation is given by the standard result

$$[\partial n(v)/\partial t]_c = \tfrac{1}{2}\int_0^v P(u, v-u)n(u)n(v-u)\,du - n(v)\int_0^\infty P(u, v)n(u)\,du \tag{1}$$

where $P(u, v)$ is the coagulation kernel. It is usually considered that

$$Q \equiv \int_0^\infty v(\partial n/\partial t)_c \, dv = 0 \tag{2}$$

(see, for example, Friedlander (1977) or Twomey (1977)), and the reasoning for this is twofold. Firstly, since each particle coagulation separately conserves volume, equation (2) must hold since the left-hand side represents the rate of change of volume for the whole system of particles. Secondly, equation (2) can be proved mathematically, using equation (1). We have

$$Q = \tfrac{1}{2}\int_0^\infty dv \int_0^v vP(u, v-u)n(u)n(v-u)\,du - \int_0^\infty dv \int_0^\infty vP(u, v)n(u)n(v)\,du. \tag{3}$$

We substitute $w = v - u$ in the first term on the right-hand side of equation (3), and this then becomes

$$\tfrac{1}{2}\int_0^\infty dw \int_0^\infty (u+w)P(u, w)n(u)n(w)\,du$$

which cancels out with the second term on the RHS of equation (3) on using $P(u, w) = P(w, u)$.

The main purpose of the present contribution is to point out that despite the above remarks equation (2) is not necessarily true and that, depending on the behaviour of $n(v)$ as $v \to \infty$, Q may be zero, non-zero but finite, or infinite. This has consequences for the asymptotic behaviour of the solution of the stationary state, time-independent coagulation equation as we shall show presently.

The basis of our contention is that the mathematical procedure of integrating the RHS of equation (1) from 0 to ∞ requires more careful consideration than was given in deriving equation (3) above. In fact,

$$Q=\lim_{V\to\infty}\left(\tfrac{1}{2}\int_0^V \mathrm{d}v\int_0^v vP(u,v-u)n(u)n(v-u)\,\mathrm{d}u-\int_0^V \mathrm{d}v\int_0^\infty vP(u,v)n(u)n(v)\,\mathrm{d}u\right). \tag{4}$$

On substituting $w=v-u$ in the first term on the RHS of equation (4) and using $P(u,w)=P(w,u)$, we readily obtain

$$\begin{aligned} Q&=\lim_{V\to\infty}\left(\int_0^V \mathrm{d}w\int_0^{V-w} wP(u,w)n(u)n(w)\,\mathrm{d}u-\int_0^V \mathrm{d}v\int_0^\infty vP(u,v)n(u)n(v)\,\mathrm{d}u\right)\\ &=-\lim_{V\to\infty}\left(\int_0^V \mathrm{d}v\int_{V-v}^\infty vP(u,v)n(u)n(v)\,\mathrm{d}u\right). \end{aligned} \tag{5}$$

To proceed further with evaluation of the limit in equation (5), we now invoke the fact that P is invariably a homogeneous function of u and v, so that

$$P(\lambda u,\lambda v)=\lambda^r P(u,v) \tag{6}$$

for some value of r. Further, we shall assume that for sufficiently large values of v

$$n(v)\propto v^{-s} \tag{7}$$

for some value of s, and that this effectively holds for $v>\beta$ where β is a large but finite quantity. We now divide the v integration interval in equation (5) (0 to V) into three sub-intervals, 0 to β, β to $V-\beta$ and $V-\beta$ to V. Then for $\beta<v<V-\beta$, both u and v in the integrand of (5) will be greater than β so that the asymptotic form (7) will apply to both $n(u)$ and $n(v)$. Further, it may be shown that as $V\to\infty$ the contribution to the integral (5) arising from $\beta<v<V-\beta$ is greater than the contributions arising from $0<v<\beta$ and $V-\beta<v<V$. Now we are currently interested in deciding whether the RHS of equation (5) is zero or non-zero, and in view of the above remarks this depends on whether

$$Z=\lim_{V\to\infty}\left(\int_\beta^{V-\beta} \mathrm{d}v\int_{V-v}^\infty vP(u,v)n(u)n(v)\,\mathrm{d}u\right)$$

is zero or non-zero. To decide this, we introduce new variables x and y defined by $x=u/V$ and $y=v/V$ and obtain

$$Z=\lim_{V\to\infty}\left(V^{3+r-2s}\int_{(\beta/V)}^{1-(\beta/V)} \mathrm{d}y\int_{1-y}^\infty yP(x,y)n(x)n(y)\,\mathrm{d}x\right) \tag{8}$$

making use of the above comments and equations (6) and (7). It follows immediately that when

$$Y=\int_0^1 \mathrm{d}y\int_{1-y}^\infty yP(x,y)n(x)n(y)\,\mathrm{d}x \tag{9}$$

is convergent, Q will be zero only if

$$s>\tfrac{1}{2}(r+3). \tag{10}$$

For $s<\frac{1}{2}(r+3)$, Q will be infinite, while if $s=\frac{1}{2}(r+3)$, it will be non-zero and finite.

When, however, Y is divergent Q will be infinite if $s \leqslant \frac{1}{2}(r+3)$, while for $s > \frac{1}{2}(r+3)$ further consideration is necessary as discussed below.

In order to progress further, it is now necessary to specify more precisely the form of $P(x, y)$ in order to discuss the convergence and divergence of Y. In all cases of interest the form of $P(x, y)$ is a product of factors, each of the form $|x^\alpha \pm y^\alpha|^\gamma$ where α and γ may be positive or negative. Thus for the two-dimensional region over which the double integral (9) is taken, Y may become infinite for the regions $\{x = \infty, 0 \leqslant y \leqslant 1\}$, $\{y = 0, 1 \leqslant x \leqslant \infty\}$, $\{x = 0, y = 1\}$. Since it is the numerically largest powers (positive or negative) of x and y which determine the corresponding convergence or divergence of Y, we may represent $P(x, y)$ by

$$P(x, y) \sim x^{(r/2)} y^{(r/2)} \max[(x/y)^q, (y/x)^q]$$

for the purpose of investigating this convergence. Here q ($\geqslant 0$) is the largest power of (x/y) which arises in the product of the above-mentioned factors. Each $P(x, y)$ is thus characterised for the convergence investigation by the pair of quantities r and q. We consider first the region $\{x = \infty, 0 \leqslant y \leqslant 1\}$, and find that the integral (9) will be convergent in the neighbourhood of this region if

$$s > (r/2) + q + 1. \tag{11}$$

Further, it transpires that the integral (9) will be convergent in the neighbourhood of both of the regions $\{y = 0, 1 \leqslant x \leqslant \infty\}$ and $\{x = 0, y = 1\}$ if

$$s < (r/2) - q + 2. \tag{12}$$

It follows immediately from equations (11) and (12) that a necessary condition for Y to be convergent is that

$$q < \tfrac{1}{2}.$$

Y may diverge due to inequality (11) not being satisfied, and in that case Z will be infinite if $s \leqslant \frac{1}{2}(r+3)$ and indeterminate if $s > \frac{1}{2}(r+3)$ (see equations (8) and (10)). If, however, Y diverges due only to inequality (12) not being satisfied, there exists the possibility that Z remains finite due to the variable V occurring in front of the integral in (8) as well as in its limits. It transpires that the condition (11) is then such as to make Z zero. The above results may all be summarised as follows. If $q < \frac{1}{2}$, Q is infinite for $s < \frac{1}{2}(r+3)$, zero for $s > \frac{1}{2}(r+3)$ and non-zero, finite for $s = \frac{1}{2}(r+3)$. If $q \geqslant \frac{1}{2}$, Q is infinite for $s \leqslant \frac{1}{2}(r+3)$, indeterminate for $\frac{1}{2}(r+3) < s \leqslant (r/2) + q + 1$ and zero for $s > (r/2) + q + 1$.

As an illustration of these results we consider the case of Brownian coagulation where

$$P(x, y) \propto (x^{1/3} + y^{1/3})(x^{-1/3} + y^{-1/3})$$

(Friedlander 1977). This corresponds to $r = 0$ and $q = \frac{1}{3}$, so that Q is finite and non-zero for $s = \frac{3}{2}$, being infinite for values smaller than this and zero for values greater than this. For gravitational coagulation

$$P(x, y) \propto (x^{1/3} + y^{1/3})^3 |x^{1/3} - y^{1/3}|$$

assuming a size-independent coagulation efficiency. This corresponds to $r = \frac{4}{3}$ and $q = \frac{2}{3}$ so that Q is infinite for $s \leqslant \frac{13}{6}$, indeterminate for $(\frac{13}{6}) < s \leqslant (\frac{7}{3})$ and zero for $s > \frac{7}{3}$. If we take into account a commonly accepted efficiency factor (Pruppacher and Klett

1978), this multiplies the above gravitational P by

$$\min[x^{2/3}, y^{2/3}]/[2(x^{1/3}+y^{1/3})^2] \tag{13}$$

which gives $r=\frac{4}{3}$ and $q=0$. Q is then finite and non-zero for $s=\frac{13}{6}$, being infinite for smaller values and zero for greater values.

Finally, we consider the error in the physical argument given earlier that Q must be zero since all the particle coagulations separately conserve volume. The essential point here is that Q as given by equation (2) need not represent the rate of change of volume for the *whole* system of particles, since the range of integration is infinite. For particle volumes in the interval 0 to V the rate of change of total material volume is

$$R(V)=\int_0^V v(\partial n/\partial t)_c \, dv,$$

and this will be negative since coagulations correspond to a net flux of material to particles of larger volume. As V increases without limit there is clearly no necessity for $R(V)$ to tend to zero since, because the particle volume is unbounded, there always exists the possibility of material flux to particles of larger volume.

The conclusions reached above have important consequences for the solution of the stationary state, time-independent coagulation equation. If $S(v)$ is the time-independent source term, this equation takes the form

$$(\partial n/\partial t)_c + S(v) = 0, \tag{14}$$

and therefore

$$Q + \int_0^\infty vS(v)\, dv = 0. \tag{15}$$

If it were considered that Q must be zero, one would then deduce from equation (15) that equation (14) could only have a solution if $\int_0^\infty vS(v)\,dv=0$, corresponding to no net flux of material into the particle system—this result is clearly incorrect. In fact, since it corresponds to the rate at which material is being fed into the particle system, $\int_0^\infty vS(v)\,dv$ is clearly non-zero and finite, and thus Q must be non-zero, but finite. This in turn means that, assuming the asymptotic form of the solution of equation (14) to behave as shown in (7), the value of s is subject to the constraints derived above for Q to be non-zero, but finite. Thus if $q<\frac{1}{2}$, $s=\frac{1}{2}(r+3)$, while if $q>\frac{1}{2}$, $\frac{1}{2}(r+3)<s\leq(r/2)+q+1$. Applying this to the cases considered earlier, we find that for Brownian coagulation $s=1.50$, for gravitational coagulation with constant efficiency factor $2.17<s<2.33$, and for gravitational coagulation with the efficiency factor (13), $s=2.17$.

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