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PARTICLE CAPTURE BY INTERCEPTION AND ITS RELEVANCE TO NUCLEAR REACTOR CONTAINMENT SPRAYS

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Abstract—A falling drop may capture aerosol particles by one of a number of mechanisms, depending largely on the particle size. Submicrometre particles, which are the hardest to capture, tend to follow the streamlines of the suspending fluid and collide with the drop only by virtue of their finite radii. Many earlier calculations of the capture efficiency by this mechanism have neglected the boundary layer around the drop: it is shown that this can lead to significant error. A new interception formula is derived, taking the boundary layer into account, in which the efficiency depends on the Reynolds number of the drop as well as the ratio of the particle and drop radii. This formula is compared with an empirical formula derived experimentally by other workers, and both are applied to the removal of airborne particles by a nuclear reactor containment spray.

Key Words: particles, aerosols, washout, containment spray systems, two-phase flow, boundary layers, trajectories, perturbation theory

1. INTRODUCTION

The capture of aerosol particles by falling drops is of interest in various fields of study; the present work arises in the context of nuclear power technology, but many of the original references appear in the meteorological literature, and the results should be of general interest in aerosol science. Water-cooled nuclear reactors are equipped with sprays to condense steam and remove fission products from the containment atmosphere in the event of an accident. The speed with which fission products are removed has an obvious bearing on the size of any possible release to the environment and a particular area of interest is whether a small fraction of the original contaminant may persist for long after the bulk of it has been removed. This paper is concerned with the persistence of aerosol particles in the size range, generally about 0.1–1 μm radius, where removal by drops is least efficient.

Since large-scale simulations (Hilliard *et al.* 1970), however comprehensive, can cover only a finite range of possible conditions in the containment, a theoretical analysis is also desirable, in which pessimistic simplifying assumptions may be made in preference to a highly involved treatment of mitigating factors. In this work, electrostatic and phoretic effects, which may enhance the removal rate in many circumstances, are ignored, and the analysis is restricted to capture by inertial impaction, interception and Brownian diffusion. Interception, a mechanism by which aerosol particles following the streamlines of the suspending fluid collide with the drop only by virtue of their finite radii, would then dominate the capture of particles in the size range of interest. It is shown that many earlier calculations were erroneously based on a formula which ignored the boundary layer around the falling drop. A new interception formula is derived here, taking the boundary layer into account, and compared with an empirical formula derived experimentally by other authors. Both formulae are applied to the removal of particles by a PWR containment spray.

2. THEORY OF PARTICLE CAPTURE

The essential quantity to obtain is E, the total capture efficiency, which is defined here as the number of particles that a falling drop collects as a fraction of those initially in its path, or more precisely, of those whose centres initially lie within the cylindrical volume swept out by the falling drop. In the absence of electrostatic effects, temperature gradients and condensing steam, E

depends on four dimensionless parameters:

$$E = E(\sigma, \mathrm{Re}, \mathrm{St}, \kappa) \quad [1]$$

where

$$\sigma = \frac{\nu}{D} \quad [2]$$

$$\mathrm{Re} = 2a\frac{U}{\nu} \quad [3]$$

$$\mathrm{St} = \frac{2Ub^2\rho_w C}{9a\nu\rho_a} \quad [4]$$

and

$$\kappa = \frac{b}{a} \quad [5]$$

ν is the kinematic viscosity of the containment atmosphere and ρ_a its density, a is the radius of the drop and U its velocity, b is the radius of the aerosol particle, D its Brownian diffusivity in the containment atmosphere, ρ_w is the density of its constituent matter and C the correction factor for finite mean free path. The quantities σ, Re and St are, respectively, the Schmidt, Reynolds and Stokes numbers.

The capture of very small particles, which is governed by diffusion, depends only on σ and Re. At the other extreme, comparatively large and heavy particles are captured by inertial impaction, depending almost entirely on St and Re; in the limit where both σ^{-1} and κ are negligibly small, impaction efficiencies vary from nil, when St is below a critical value $\mathrm{St_c}$, which is itself a function of Re, to unity in the limit of high St. The region of greatest interest in the present work is the intermediate one, where the total efficiency is low and the ratio κ may be vitally important. If both σ^{-1} and St are negligible, a particle may be carried by the fluid along a streamline which passes so close to the drop that a collision occurs on account of the particle's finite size. The efficiency of capture by this process, interception, must be a function of κ and Re. The total efficiency is frequently represented by the expression

$$E(\sigma, \mathrm{Re}, \mathrm{St}, \kappa) = E_{\mathrm{dif}}(\sigma, \mathrm{Re}) + E_{\mathrm{int}}(\kappa, \mathrm{Re}) + E_{\mathrm{imp}}(\mathrm{St}, \mathrm{Re}) \quad [6]$$

ignoring any inaccuracy introduced by this simple separation of the contributing effects.

For the earliest analyses, efficiency calculations, apart from those for diffusion, were available only in the limits of infinite Re (potential flow) and negligible Re (Stokes flow). Parsly (1971) recognized these limitations in the impaction regime, and more recently Grist (1982) chose pessimistically to use the low impaction efficiencies derived by assuming Stokes flow. However, the interception formula derived by Ranz & Wong (1952), using potential flow theory, has consistently been represented as the minimum efficiency that can occur. The significance of this assumption can be demonstrated by comparing the resulting formula with that which is obtained at the opposite extreme with Stokes flow (Fuchs 1964):

$$E_{\mathrm{int}}(\mathrm{Re} \to \infty, \text{potential}) = 3\,\kappa \quad [7]$$

$$E_{\mathrm{int}}(\mathrm{Re} \to 0, \text{Stokes}) = 1.5\,\kappa^2 \quad [8]$$

the value of κ being assumed to be small in both cases. Clearly, the use of [7] in the earlier work may turn out to have overestimated the washout rate when Re is finite.

Spray drops in the size range of interest here may be regarded effectively as rigid spheres falling at their terminal velocities with Reynolds numbers mainly in the range 10–1000 (Pruppacher & Klett 1978). The nature of the fluid flow in such circumstances is discussed in standard engineering text books, such as Kay & Nedderman (1974), as well as the more specialized references to be cited later. Broadly speaking, the effect of viscosity in the flow round the leading face of the sphere is confined to a laminar boundary layer whose thickness is of the order $\mathrm{Re}^{-1/2}a$, where a is the radius of the sphere. A recirculatory wake follows, but outside these two regions the flow resembles that of a perfect fluid. It is shown later in this paper that the presence of the laminar boundary layer around a falling drop deflects the streamlines away from its surface by about 30 μm at their point of closest approach throughout the range of drop radii of interest. The inadequacy of the Ranz & Wong formula [7] for the capture of micrometre-sized particles is thus immediately apparent, since it follows that these must be captured, not from the outer fluid, but from within the boundary layer.

3. FLUID FLOW AROUND A SPHERE

At Reynolds numbers below about 400, which account for most of the range of interest here, the flow around a spherical drop is steady and can, in principle, be derived by numerically solving the Navier–Stokes and continuity equations which govern the fluid motion (Pruppacher & Klett 1978). (At higher Re, instability sets in with the periodic detachment of eddies in the wake of the sphere.) With the further assumption that the aerosol particles do not perturb the fluid flow, but move in it as if they were point masses subject to their own inertia and to viscous drag forces, the particle motion can also be solved numerically. This programme has been carried out for a range of St, Re and κ by Beard & Grover (1974) with flow fields derived by Le Clair *et al.* (1970). Slinn (1974) attempted to fit these results with an analytic expression in the form of [6] but the validity of his interception term has been questioned (Underwood 1983). For the present work, an alternative interception formula has been derived from boundary layer theory. The physical basis of the derivation, which also leads to an approximate formula for the critical Stokes number, is described below, with the detailed mathematics relegated to the appendix.

If the spray drop is moving through a perfect, inviscid fluid at a steady speed, U, the radial and tangential components of fluid velocity relative to the centre of the sphere may be derived by potential flow theory:

$$u_r = -U\cos\theta\left(1-\frac{a^3}{r^3}\right) \qquad [9]$$

and

$$u_\theta = U\sin\theta\left(1+\frac{a^3}{2r^3}\right) \qquad [10]$$

where θ is the angle between the position vector, $\mathbf{r}$, relative to the centre of the drop, and the direction of travel (see figure 1a). These equations may be approximated for the fluid close to the surface, where

$$u_r \approx -3\left(\frac{U}{a}\right)\cos\theta\,(r-a) \qquad [11]$$

and

$$u_\theta \approx 1.5\left(\frac{U}{a}\right)a\sin\theta. \qquad [12]$$

In the region of the forward stagnation point,

$$u_y \approx -3\left(\frac{U}{a}\right)y \qquad [13]$$

and

$$u_x \approx 1.5\left(\frac{U}{a}\right)x \qquad [14]$$

x and y being distances from the stagnation point, measured parallel and perpendicular respectively to the surface. If the curvature is neglected, but (U/a) remains finite, [13] and [14] describe a situation known as three-dimensional stagnation flow.

With finite viscosity and no slippage at the surface, the full equations for the fluid motion can still be considerably simplified in the case of three-dimensional stagnation flow (see the appendix). The resulting solution can clearly be interpreted in terms of a boundary layer of constant thickness, outside which u_x remains unchanged, as in [14], but u_y is modified so that

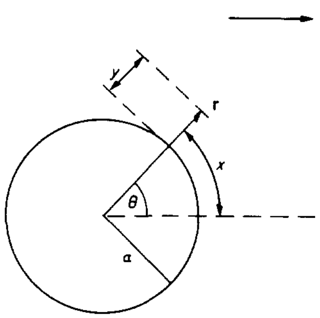

Figure 1a. Coordinate system for fluid flow around a falling drop.

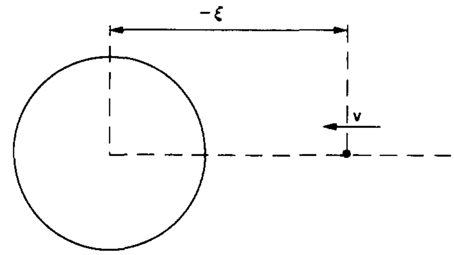

Figure 1b. Coordinate system for a particle on the central streamline.

$$u_y \to -3\left(\frac{U}{a}\right)(y - \delta_1) \qquad [15]$$

where

$$\delta_1 = 0.8045\left(\frac{a\nu}{3U}\right)^{1/2}. \qquad [16]$$

The quantity δ_1 is known as the displacement thickness, as it is the distance by which streamlines are displaced away from the surface by the effect of the boundary layer. Equations [15] and [16] also hold good as an approximation near the forward stagnation point of a finite drop at high Re, in which case [16] can be rewritten as

$$\delta_1 = 0.657\,\mathrm{Re}^{-1/2}a. \qquad [17]$$

This result is used in the appendix to calculate the critical Stokes number for impaction.

Further away from the stagnation point, the boundary layer equations are no longer exactly soluble, even in the limit of high Re, but good approximate solutions can be obtained by perturbation theory over the range of $\theta \lesssim 75°$. Beyond this, the perturbation expansion does not converge rapidly, and furthermore [12] ceases to be a reliable approximation to the outer fluid velocity, owing to the neglect of the wake behind the drop. However, it is shown in the appendix that streamlines which pass well inside the boundary layer, from which particles may be captured by interception, make their closest approach to the surface when $\theta \approx 70°$ (see figure 2), in contrast to the situation both in potential and Stokes flow, where the closest approach is at 90°.

This mathematical description of the boundary layer flow allows a new interception formula to be derived, which includes the dependence on Re. For small particles, where $\kappa a \ll \delta_1$, the formula is

$$E_{\mathrm{int}} = 1.00\,\mathrm{Re}^{1/2}\kappa^2. \qquad [18]$$

Since the displacement thickness, still of order $\mathrm{Re}^{-1/2}a$, must also be much smaller than the radius of the drop, the range of validity of [18] is

$$\kappa \ll \mathrm{Re}^{-1/2} \ll 1. \qquad [19]$$

By contrast, the formulae quoted in [7] and [8] are valid only for extreme values of Re.

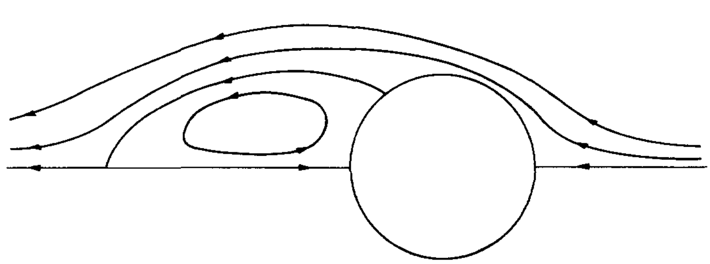

Figure 2. Flow pattern around a falling drop. Not to scale.

It is important to note that all three interception formulae rest on the assumption that the centre of the aerosol particle follows a streamline of the fluid through to the moment of collision, at which point the particle is captured. Significant departures from this theory in experimental results are discussed later.

4. WASHOUT RATES IN THE PWR CONTAINMENT

The implications of the new interception formula can now be tested by specific calculations for a PWR containment. The basic assumption is that spray drops, small enough to be regarded as undistorted spheres, fall through the containment atmosphere at their terminal velocity, and that fission products are present as an aerosol of aqueous particles which are very much smaller than the falling drops. One further possible mode of capture, through the entrainment of particles into the recirculatory wake as it develops behind the accelerating drop, is thus neglected. In a full analysis, a range of containment pressures, atmospheric compositions, temperatures and viscosities would be taken into account, but a single set of values is sufficient to demonstrate the main effects. The following values are therefore assumed:

$$\begin{aligned} \rho_a \text{ (density of atmosphere)} &= 1.2\,\mathrm{kg\,m^{-3}} \\ \rho_w \text{ (density of drops and particles)} &= 10^3\,\mathrm{kg\,m^{-3}} \\ \nu \text{ (kinematic viscosity of atmosphere)} &= 1.5 \times 10^{-5}\,\mathrm{m^2\,s^{-1}} \\ T \text{ (absolute temperature)} &= 300\,\mathrm{K}. \end{aligned} \qquad [20]$$

Spray drops with radii in the range 50–1000 μm are considered. In table 1, values of Re and the terminal velocity, U, have been calculated by standard methods (Fuchs 1964). The displacement thickness at 70° from the forward stagnation point, δ_1 (70°), is obtained from [A.29] of the appendix, the critical Stokes number, $\mathrm{St_c}$, from [A.43], and the corresponding radius, b_c, from [A.31] with the approximation that $C = 1$.

Capture by interception is only important for aerosol particles with radii $< b_c$, otherwise impaction is dominant. The values in table 1, therefore, show that intercepted particles must be much smaller than the displacement thickness and are thus captured from streamlines passing well within the boundary layer. In other words, the conditions specified for [18] to be correct are satisfied, except for the 50 μm drop, for which [8] is preferable. In no case should the Ranz & Wong formula [7] be applied.

By combining [18] with a standard formula for convective diffusion across a boundary layer (Underwood 1983) and substituting both into [6], the total efficiency in this region can be expressed as

$$E = 3.0\,\mathrm{Re}^{-1/2}\sigma^{-2/3} + 1.00\,\mathrm{Re}^{1/2}\kappa^2 \qquad [21]$$

where

$$\sigma = \frac{\nu}{D} \qquad [22]$$

$$D = \frac{CkT}{6\pi\rho_a \nu b} \qquad [23]$$

and k is Boltzmann's constant, $1.38 \times 10^{-23}\,\mathrm{J\,K^{-1}}$.

Table 1. Spray drop parameters for PWR containment

a (μm)	U (m s^{-1})	Re	δ_1(70°) (μm)	$\mathrm{St_c}$	b_c (μm)
50	0.25	1.7	35	0.84	3.7
100	0.69	9.2	30	0.50	2.4
200	1.6	42	28	0.34	1.9
300	2.4	97	28	0.28	1.7
700	5.2	480	29	0.22	1.5
1000	6.7	900	31	0.20	1.5

The minimum efficiency could be sought by exhaustive computation, but the problem is considerably simplified by observing that the terminal velocities of the spray drops happen to be approximately proportional to their radii over most of the region of interest; thus

$$\mathrm{Re}^{1/2} \approx 3.2 \times 10^4 a \qquad [24]$$

all lengths being measured in metres. Further substitution of numerical values from [20] into [21], with the approximation that $C = 1$, leads to the expression

$$E = a^{-1}(8.3 \times 10^{-13} b^{-2/3} + 3.2 \times 10^4 b^2). \qquad [25]$$

Simple calculus shows that E reaches a minimum of $2.0 \times 10^{-8} a^{-1}$ when $b = 4.0 \times 10^{-7}$ m. The efficiency is less than twice this minimum value throughout the range of particle radii from 0.1 to 1 μm. Minimum efficiencies for the range of collecting drop sizes shown in table 1 vary from 4×10^{-4} down to 2×10^{-5}.

The removal rate of aerosol particles from the containment can be calculated as follows:

$$\lambda(b) = \frac{\pi h}{V} \int a^2 E(a, b)\, \mathrm{d}N(a) \qquad [26]$$

where λ is the fractional removal rate, h is the height through which a drop falls, V is the volume of the containment and $N(a)$ is the number of spray drops emitted per second with radii $<a$; the corresponding volumetric rate of spraying is

$$F = \frac{4\pi}{3} \int a^3\, \mathrm{d}N(a). \qquad [27]$$

A relatively simple relationship between these quantities can be deduced by using [25] and assuming that the size distribution of spray drops is log-normal; the minimum removal rate in this case is

$$\lambda(0.4\,\mu\mathrm{m}) = 2 \times 10^{-8} \left(\frac{3hF}{4Va_{\mathrm{m}}^2}\right) \exp[2(\ln \sigma_{\mathrm{g}})^2] \qquad [28]$$

where a_{m} is the mass-median drop radius and σ_{g} the geometric standard deviation of the distribution. Reasonable numerical values for h, V and F (Grist 1982), a_{m} and σ_{g} (Hilliard *et al.* 1970) are

$$h = 36\ \mathrm{m}$$
$$V = 7.08 \times 10^4\ \mathrm{m}^3$$
$$F = 0.39\ \mathrm{m}^3\,\mathrm{s}^{-1}$$
$$a_{\mathrm{m}} = 6.05 \times 10^{-4}\ \mathrm{m}$$
$$\sigma_{\mathrm{g}} = 1.53 \qquad [29]$$

whence

$$\lambda(0.4\,\mu\mathrm{m}) = 1.2 \times 10^{-5}\ \mathrm{s}^{-1}. \qquad [30]$$

In other words, on the basis of these assumptions, the most persistent aerosol has a half-life of about 16 h in the sprayed containment.

For comparison, the calculation made above can be repeated with the Ranz & Wong formula in place of [18]. The result is a minimum capture efficiency of $1.7 \times 10^{-7} a^{-1}$ for aerosol particles of radius 2.3×10^{-8} m. The corresponding minimum removal rate in the sprayed containment is $1.0 \times 10^{-4}\ \mathrm{s}^{-1}$, a half-life of under 2 h.

Since [18] relies on a number of simplifying assumptions, which are discussed below, further comparison with an empirical formula is worthwhile. Röbig *et al.* (1978) were able to fit their experimental results as follows:

$$E_{\mathrm{int}}(\mathrm{empirical}) = 0.35\, \mathrm{Re}^{1/2} \kappa^{3/2} \qquad [31]$$

the numerical factor quoted here being modified on account of their different definition of Re. If this formula is used in place of [18], the minimum in the total efficiency is rather higher and its

Table 2. Position and size of minimum capture efficiency from theoretical and empirical formulae

	Theoretical		Empirical	
a (μm)	b_{min} (μm)	E_{min}	b_{min} (μm)	E_{min}
50	0.40	4.0×10^{-4}	0.23	6.4×10^{-4}
100	0.40	2.0×10^{-4}	0.20	3.6×10^{-4}
200	0.40	1.0×10^{-4}	0.17	2.0×10^{-4}
300	0.40	6.7×10^{-5}	0.16	1.4×10^{-4}
700	0.40	2.8×10^{-5}	0.13	6.9×10^{-5}
1000	0.40	2.0×10^{-5}	0.12	5.1×10^{-5}

position, which now depends on the drop radius, is shifted towards lower particle radii. For example, the minimum total efficiency for a drop of radius 100 μm is 3.6×10^{-4}, which occurs when the particle radius is 0.20 μm; more detailed figures are given in table 2. The removal rate in a sprayed containment can be calculated by methods similar to those used above, though rather more complicated on account of the less convenient mathematical form of E. The minimum rate predicted for otherwise identical conditions is

$$\lambda(0.14\,\mu\text{m, empirical}) = 2.5 \times 10^{-5}\,\text{s}^{-1} \qquad [32]$$

and the corresponding half-life is about 8 h.

5. DISCUSSION

In deriving [18] it has been necessary to neglect variations in the fluid velocity over distances comparable with (in fact, significantly greater than) the radius of the aerosol particles. Even with the great disparity in size between the particles and spray drops considered here, this assumption cannot hold good when the particle approaches the drop to within a few times the particle radius. Indeed, with ideally smooth surfaces and a perfectly continuous fluid, the particle and drop could never come into contact (Pruppacher & Klett 1978). The fact that they do so is attributable partly to interparticle forces and partly to the molecular nature of the fluid (the mean-free-path of molecules in the containment atmosphere is of the order of 0.1 μm). Calculated efficiencies in the impaction regime tend to be borne-out by experimental measurements, which confirm the theoretical prediction that the finite particle size is of only marginal effect here. This is clearly not the case in the interception regime, where the particle size is all-important. In the region of interest, the particle approaches the drop surface on a glancing trajectory in a fluid whose undisturbed velocity is proportional to the distance from the surface. The motion of the fluid must therefore be strongly influenced by the presence of the particle, which takes on a rolling motion so as to balance the viscous forces acting on its own surface. The capture efficiency, particularly of small particles by large drops, may also be affected by circulation within the drop, which modifies the boundary condition at the drop's surface. The neglect of such complexities implies that the interception formula of [18], though certainly preferable to the Ranz & Wong formula used hitherto, should not be considered in any way precise.

Experimental measurements by Röbig *et al.* (1978) confirm [18] only to the extent of its dependence on $\text{Re}^{1/2}$; otherwise the theoretically-derived formula appears significantly to underestimate the interceptive capture of the smaller particles. On the other hand, numerical computations by Beard (1974), which include the influence of terrestrial gravity on the aerosol particle but otherwise rest on the same assumptions as the present work, predict significantly higher efficiencies for the capture of submicrometre particles than were observed in these experiments (Underwood 1983). Although the empirical formula [31] can very reasonably be used as above in exploratory calculations, its lack of theoretical foundation suggests that it should be applied only with caution in any conditions which differ significantly from those of the experiments.

In practice, the capture of submicrometre particles is likely in many cases to be dominated by mechanisms excluded from consideration in this work. For example, the effect of Stefan flow and diffusiophoresis in the presence of condensing steam may well have been the controlling factor for the finest aerosol component in the Containment Systems Experiment (Hilliard *et al.* 1970), where the temperature of the vessel was maintained by steam injection; the lowest observed collection efficiency was 1.5×10^{-3} (Postma & Pasedag 1973), well above the theoretically predicted minimum. Although these phoretic effects can significantly reduce the persistence of submicrometre aerosols, a detailed analysis of evaporation, condensation and heat transfer processes in the containment, for a full range of accident conditions, would be needed in order to take them properly into account. Their importance in the meteorological context is discussed in the relevant literature (Pruppacher & Klett 1978).

6. CONCLUSIONS

Many earlier analyses of particle removal by falling drops have neglected the boundary layer around the drop in estimating the capture efficiency by interception. Equation [18], derived here so as to take account of the boundary layer, and the empirical formula of Röbig *et al.* (1978) both imply that this neglect may have led to an overestimate of the removal rate for the most persistent aerosol particles by factors ranging from 4 to 8, in the absence of electrostatic and phoretic effects. Further theoretical work at the microphysical level is required in order to understand fully the motion of an aerosol particle and the surrounding fluid in the interception regime; at present no physically based formula is totally consistent with the most relevant experimental data.

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APPENDIX

Mathematical Details

Flow around the spherical spray drop

The flow of air around the spray drop is governed by the continuity equation,

$$\nabla \cdot \mathbf{u} = 0 \tag{A.1}$$

and the Navier–Stokes equation,

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{\nabla P}{\rho_a} + \nu \nabla^2 \mathbf{u} \tag{A.2}$$

subject to the condition of no slip at the surface.

If the drop is falling with a steady velocity $\mathbf{U}$, the position coordinate and fluid velocity can be redefined in a reference frame in which the drop is at rest, so that the time-dependent term in [A.2] vanishes and the fluid velocity tends to $-\mathbf{U}$ at large distances. The potential flow solution, where $\nu \to 0$ and the fluid moves freely over the drop's surface, is given in [9] and [10] of the main text.

The boundary layer, which develops with finite viscosity, can be described in terms of two curvilinear coordinates, since the flow is axially symmetric about the trajectory of the drop's centre (see figure 1a):

$$x = a\theta \tag{A.3}$$

and

$$y = r - a. \tag{A.4}$$

In three-dimensional stagnation flow, where the curvature a^{-1} vanishes but (U/a) remains finite, an exact solution of [A.1] and [A.2] can be obtained in the following form, slightly adapted from Goldstein (1938):

$$u_x = 1.5\left(\frac{Ux}{a}\right)f'(\eta) \tag{A.5}$$

$$u_y = -3\left(\frac{Uc}{a}\right)f(\eta) \tag{A.6}$$

and

$$P = P_0 - \rho_a\left\{\frac{9}{8}\left(\frac{Ux}{a}\right)^2 + \frac{9}{2}\left(\frac{Uc}{a}\right)^2(f^2 + 2f')\right\} \tag{A.7}$$

where

$$\eta = c^{-1}y = \left(\frac{3U}{a\nu}\right)^{1/2} y \tag{A.8}$$

$$f''' + ff'' + \tfrac{1}{2}(1 - f'^2) = 0 \tag{A.9}$$

and the "primes" denote differentiation with respect to η. The boundary conditions

$$f(0) = f'(0) = 0 \tag{A.10}$$

and

$$f'(\eta \to \infty) = 1 \tag{A.11}$$

give the correct behaviour at the surface and lead to [14] and [15] of the main text in the outer fluid.

Equation [A.9] belongs to a class of boundary layer equations for which accurate numerical solutions are available (Evans 1968). The functions f and f' are plotted in figure A1 and for large values of η,

$$f(\eta \to \infty) = \eta - \delta_1^* = \eta - 0.8045. \tag{A.12}$$

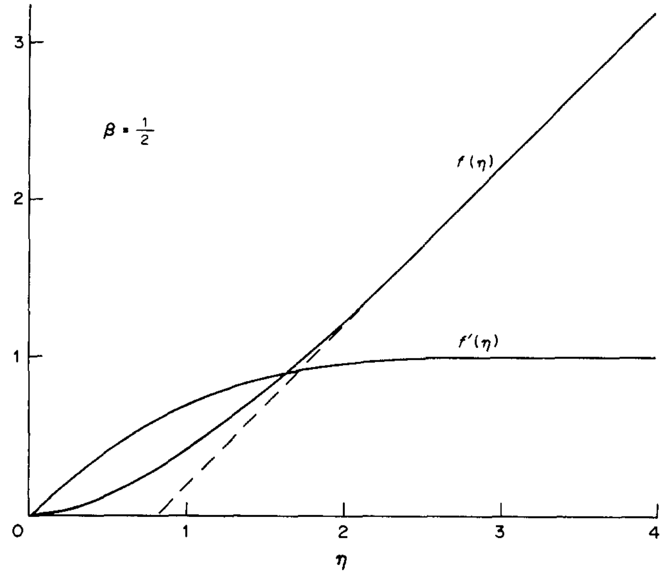

Figure A1. The dimensionless boundary layer functions for three-dimensional stagnation flow.

Equations [15] and [16] of the main text follow by substituting the original variables. When the finite radius is taken into account, an exact reduction of the full equation is no longer possible, but an approximate solution can be found for thin boundary layers by making a suitable coordinate transformation and then applying perturbation theory (Merk 1959). No attempt is made here to reproduce the full mathematical analysis, which is highly involved, but the various quantities that arise in the treatment by Evans (1968) are evaluated below for the special case of a sphere.

The curvilinear distance, x, from the stagnation point, the perpendicular distance, s, and the outer velocity, U_1, are respectively,

$$x = a\theta \qquad [A.13a]$$

$$s = a \sin\theta \qquad [A.13b]$$

and

$$U_1 = 1.5\, U \sin\theta \qquad [A.13c]$$

where the potential flow value from [12] of the main text has been taken as an approximation to U_1. The transformed coordinates used by Evans (1968) then become

$$\zeta = \int_0^x \left(\frac{U_1 s^2}{v}\right) \mathrm{d}x = \tfrac{1}{2} U \frac{a^3}{v} (1 - \cos\theta)^2 (2 + \cos\theta) \qquad [A.14]$$

and

$$\eta = \frac{U_1 s y}{v(2\zeta)^{1/2}} = \frac{y}{c} \qquad [A.15]$$

where

$$c = \frac{2\sqrt{2}}{3} \frac{a}{\mathrm{Re}^{1/2}} \frac{(2 + \cos\theta)^{1/2}}{(1 + \cos\theta)} \qquad [A.16]$$

and Re is defined as in [3]. The definition of η becomes identical to [A.8] at the stagnation point, where $\theta = 0$. The remaining parameters used in the analysis are

$$\beta = \frac{2\zeta}{U_1} \frac{\mathrm{d}U_1}{\mathrm{d}\zeta} = \frac{2\cos\theta(2 + \cos\theta)}{3(1 + \cos\theta)^2} \qquad [A.17]$$

and

$$\frac{d\beta}{d\chi} = 2\zeta\frac{d\beta}{d\zeta} = -\frac{8(1-\cos\theta)(2+\cos\theta)}{9(1+\cos\theta)^4}. \qquad [A.18]$$

It can be shown that

$$u_x = U_1 f'(\eta) \qquad [A.19]$$

where

$$f''' + ff'' + \beta(1+f'^2) = \frac{d\beta}{d\chi}\left(f'\frac{\partial f'}{\partial\beta} - f''\frac{\partial f}{\partial\beta}\right) \qquad [A.20]$$

and the "prime" notation now denotes partial differentiation with respect to η; the boundary conditions are identical to [A.10] and [A.11]. At the stagnation point, [A.20] becomes identical to [A.9].

If the terms on the r.h.s. of [A.20] are small, the equation can be treated by perturbation theory with the series expansion

$$f = f_1 + \frac{d\beta}{d\chi}f_2 + \cdots. \qquad [A.21]$$

Evans (1968) has computed the functions f_1 and f_2 and provides tables of the chief numerical parameters for a range of values of β.

With the boundary layer flow thus obtained by standard methods, an interception formula can now be derived by assuming that the centre of an aerosol particle precisely follows a streamline in the fluid; interception then occurs if the streamline passes closer to the surface than the radius of the particle. Because of the axial symmetry, the streamlines describe a set of tubular surfaces, each of which can be defined by a constant value of the stream function, Φ, which is equal to the total rate of fluid flow through the tube divided by 2π. The efficiency E_{int} was defined in section 2 so that a particle that is only just intercepted must therefore be travelling along a streamline for which

$$2\pi\Phi = E_{\text{int}}\pi a^2 U. \qquad [A.22]$$

The following expression for Φ in the boundary layer can be obtained from [A.15] and [A.19]:

$$\Phi = s\int_0^y u_x\,dy = sU_1 c f\left(\frac{y}{c}\right) \qquad [A.23]$$

which depends on θ through s, U_1, c and the functional form of f. At the point of closest approach,

$$\left.\frac{\partial y}{\partial\theta}\right|_\Phi = -\frac{\left.\frac{\partial\Phi}{\partial\theta}\right|_y}{\left.\frac{\partial\Phi}{\partial y}\right|_\theta} = 0 \qquad [A.24]$$

and E_{int} can therefore be found by maximizing the value of Φ in [A.23], where $y = b$, and substituting this value into [A.22].

If b is large enough to extend into the outer fluid,

$$\Phi \to 1.5Ua\sin^2\theta\left\{b - c\left(\delta_1^*(\beta) - \frac{d\beta}{d\chi}f_2(\eta\to\infty,\beta)\right)\right\} \qquad [A.25]$$

to second order in the perturbation series, and the Ranz & Wong interception formula [7] follows by neglecting the displacement term, i.e. assuming that $c \ll b$, in [A.25]. At the other extreme, where $b \ll c$, the function f can be approximated by the first non-zero term in a Taylor series,

$$f \approx \tfrac{1}{2}f''(0)\eta^2. \qquad [A.26]$$

Table A1. Boundary layer quantities for interception efficiency calculation

θ	β	$\frac{d\beta}{d\chi}$	f''_{10}	f''_{20}	$\frac{2\Phi}{b^2 U\,\mathrm{Re}^{1/2}}$
45	0.438	−0.083	0.883	−0.063	0.733
60	0.370	−0.219	0.831	−0.073	0.959
75	0.246	−0.593	0.729	−0.098	0.978
70	0.297	−0.422	0.772	−0.087	0.997

$\delta_1^*(0.297) = 0.913$; $f_2(\eta \rightarrow \infty, 0.297) = -0.137$.

By combining [A.22], [A.23] and [A.26] and substituting the original variables,

$$E_{\mathrm{int}} = \max\left\{\frac{9\,\mathrm{Re}^{1/2}\kappa^2}{4\sqrt{2}}\frac{\sin^2\theta(1+\cos\theta)}{(2+\cos\theta)^{1/2}}\left(f_1''(0) + \frac{d\beta}{d\chi}f_2''(0) + \cdots\right)\right\} + O(\mathrm{Re}\,\kappa^3) \qquad [A.27]$$

where κ is defined as in [5] of the main text. Table A1 shows the relevant quantities, evaluated for θ in the range 45°–75°, using quadratic interpolation between the values of $f_1''(0)$ and $f_2''(0)$ tabulated by Evans (1968), from which it appears that the maximum occurs when θ is about 70°. Here the second-order correction to Φ is slightly less than 5% of the first-order term, which suggests good convergence. After rounding to two decimal places, it follows that

$$E_{\mathrm{int}} = 1.00\,\mathrm{Re}^{1/2}\kappa^2[1 + \mathrm{O}(\mathrm{Re}^{1/2}\kappa)]. \qquad [A.28]$$

The displacement thickness at this angle is obtained via [A.25] by interpolating between tabulated values of $\delta_1^*(\beta)$ and $f_2(\eta \rightarrow \infty, \beta)$ and substituting for c, whence

$$\delta_1(\theta = 70°) = 0.92\,\mathrm{Re}^{-1/2}a. \qquad [A.29]$$

The critical Stokes number

Impaction calculations are generally based on the assumption that the drag on a small aerosol particle is directly proportional to its velocity relative to the surrounding fluid (Pruppacher & Klett 1978). If gravitational and interparticle forces are neglected, the equation of motion is

$$\mathrm{St}\frac{d^2\tilde{\mathbf{r}}}{d\tau^2} = \tilde{\mathbf{u}} - \frac{d\tilde{\mathbf{r}}}{d\tau} \qquad [A.30]$$

where

$$\tilde{\mathbf{r}} = \frac{r}{a}$$

$$\tilde{\mathbf{u}} = \frac{\mathbf{u}}{U}$$

$$\tau = \frac{Ut}{a}$$

and

$$\mathrm{St} = \frac{2Ub^2\rho_w C}{9av\rho_a}. \qquad [A.31]$$

St is the Stokes number, as in the main text. If the particle is approaching the drop along the central streamline, [A.30] can be rewritten in scalar form,

$$\mathrm{St}\,vv' + v = -\tilde{u}_r \qquad [A.32]$$

where v is the dimensionless velocity of the particle and the "prime" denotes differentiation with respect to ξ, the dimensionless distance from the centre of the drop to the particle, measured in the direction of the relative flow of the fluid; thus v is always positive and ξ negative for the approaching particle (see figure 1b). In potential flow, therefore,

$$\mathrm{St}\,vv' + v = 1 + \xi^{-3}. \qquad [A.33]$$

On this simple model, pure impaction will not occur unless a particle following the central streamline has a finite residual velocity on arrival at the surface, where $\xi = -1$; otherwise an identical particle, infinitesimally displaced from the central streamline, would be carried past the collecting drop by the fluid and could only be captured by virtue of its finite radius. If there is no residual velocity, let

$$v = (1 + \xi^{-3})\phi. \qquad [A.34]$$

It follows that

$$\mathrm{St}\,\phi[(1 + \xi^{-3})\phi' - 3\xi^{-4}\phi] + \phi = 1. \qquad [A.35]$$

When ξ is large and negative, ϕ is constant and equal to unity, as the particle simply follows the flow. At the surface of the drop,

$$-3\,\mathrm{St}\,\phi^2 + \phi = 1 \qquad [A.36]$$

therefore

$$\phi(-1) = \frac{1 \pm \sqrt{1 - 12\,\mathrm{St}}}{6\,\mathrm{St}}. \qquad [A.37]$$

The critical value of the Stokes number,

$$\mathrm{St}_c = \frac{1}{12} \qquad [A.38]$$

emerges from the requirement that ϕ must be real. It can also be shown that a series solution of [A.35] in ascending powers of St, which tends to unity as $\xi \to -\infty$, also converges to [A.37], with the minus sign in the numerator, at the surface.

There is no simple derivation of St_c when the boundary layer is included. However, a slight overestimate of St_c can be obtained by assuming that the particle must have sufficient residual velocity after travelling through the potential flow regime of the outer fluid to be able to cross the displacement thickness of the boundary layer. This approximation is valid if the particle velocity is several times as large as the fluid velocity at all points in its passage through the boundary layer. It follows, from [17] of the main text and [A.32], that

$$v_{\mathrm{res}} = 0.657\,\mathrm{Re}^{-1/2}\,\mathrm{St}^{-1}. \qquad [A.39]$$

As a first step towards a formula for v_{res}, [A.33] has the following approximate solution while v remains close to unity:

$$v \approx 1 + \exp\left(\frac{-\xi}{\mathrm{St}}\right) \int_{-\infty}^{\xi} \frac{\exp\left(\frac{\xi}{\mathrm{St}}\right)}{\mathrm{St}\,\xi^3}\,d\xi = 1 - \mathrm{St}^{-3} \exp\left(\frac{|\xi|}{\mathrm{St}}\right) \mathrm{E}_3\left(\frac{|\xi|}{\mathrm{St}}\right). \qquad [A.40]$$

For large values of St,

$$v_{\mathrm{res}} \approx 1 - \tfrac{1}{2}\,\mathrm{St}^{-1} + O(\mathrm{St}^{-2}). \qquad [A.41]$$

Equation [A.40] can be used to provide initial values of v for the numerical integration of [A.33]; residual velocities so obtained by the standard Runge–Kutta method in the range of St from 0.2 to 2.0 are given in table A2. These values can be very well fitted by an approximate formula,

$$v_{\mathrm{res}} = \frac{\mathrm{St} - 0.139}{\mathrm{St} + 0.323} \qquad [A.42]$$

Table A2. Residual velocity in potential flow

St	v_{res}	Equation [A.42]
0.2	0.117	0.117
0.5	0.439	0.439
1.0	0.653	0.651
2.0	0.799	0.801

which also compares well with the asymptotic formula [A.41] for large St but should not be extrapolated to $\mathrm{St} < 0.2$.

Substitution from [A.42] into [A.39] provides the following formula for the critical Stokes number:

$$\mathrm{St_c} = 0.070 + (0.0048 + 0.26\,\mathrm{Re}^{-1/2} + 0.11\,\mathrm{Re}^{-1})^{1/2} + 0.33\,\mathrm{Re}^{-1/2}. \qquad [\mathrm{A.43}]$$

Equation [A.43], which shows remarkably good agreement with the exact computations by Beard & Grover (1974), even when $\mathrm{Re} = 1$, has been used for the calculations summarized in table 1 of the main text.