The hydrodynamic interaction of two unequal spheres moving under gravity through quiescent viscous fluid

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The hydrodynamic forces and couples that act on two spherical particles in slow motion through a quiescent fluid are determined as functions of the relative configuration of the particles from the solution of the Stokes equation for the motion of the fluid in the vicinity of the particles. General formulae that relate the translational and rotational velocities of the particles to the ratios of their radii $a = a_2/a_1$ and net densities $I = (\rho_2 - \rho)/(\rho_1 - \rho)$ are obtained, as are asymptotic forms for the velocities in the limiting cases of very large and very small interparticle separation. Relative trajectories of the particles when they move solely under gravity and their own interaction are calculated for several values of I and a. A particularly interesting feature of the results is that, for certain ranges of values of I and a, trajectories of finite length and trajectories having the form of closed periodic orbits may occur.

1. Introduction

The motion of particles through fluid media under the combined action of gravitational forces and hydrodynamic interaction is an important aspect of a number of problems in suspension and aerosol mechanics. Settling velocities of suspensions, efficiencies of spray scrubber devices for removing particulates from gas streams and rates of agglomeration of aerosol particles in the atmosphere all depend on the nature of relative motion of particles in suspension. Previous studies of this problem have dealt with the interaction of two spherical particles of equal density, their relative motion being caused by a difference in size. In this paper the combined effects of unequal size and density on the motion of two spherical particles will be examined.

Several simplifying assumptions are made in the analysis presented here. The ambient fluid is taken to be incompressible and Newtonian; the fluid is assumed to be unbounded and to be at rest far from the particles. The particles are taken to be rigid and sufficiently small that their inertia is negligible in comparison with the pressure and viscous forces exerted on them and that

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FIGURE 1. Co-ordinate geometry and notation.

inertial forces in the fluid are everywhere small compared with local viscous forces. When the clearance between the particles is of the order of the particle radii, the latter condition is met when the translational Reynolds numbers

$$Re_i = a_i u_i^{(\infty)} \rho / \mu$$

of the particles based on their Stokes terminal velocities $u_i^{(\infty)} = \frac{2}{9}(\rho_i - \rho)a_i^2g/\mu$ are much less than unity. Here a_i and ρ_i denote the particle radii and densities, respectively, μ and ρ are the viscosity and density of the fluid and g is the magnitude of the gravitational acceleration. However when the particles are close together the time and length scales of the fluid motion in the gap between them, being proportional to the clearance, become very small, so it would appear that the condition of negligible inertial forces in the fluid might place additional restrictions on the Reynolds number. That this is not the case will be shown later in §3.

Under these conditions the steady-state velocity and pressure fields $\mathbf{v}(\mathbf{x})$ and $p(\mathbf{x})$ in the fluid satisfy equations of continuity and motion of the forms

$$\nabla \cdot \mathbf{v} = 0, \quad \mu \nabla^2 \mathbf{v} = \nabla p, \tag{1.1a,b}$$

together with the boundary conditions of no slip on the particle surfaces and no motion at infinity. Let the instantaneous position of the centre of the particle of radius a_2 and density ρ_2 be \mathbf{x}_0 and that of the particle of radius a_1 and density ρ_1 be $\mathbf{x}_0 + \mathbf{r}$, as shown in figure 1. Further, let \mathbf{u}_i and $\boldsymbol{\omega}_i$ be, respectively, the instantaneous translational and rotational velocities of particle *i*, and let \mathbf{r}_i be the position vector of any point relative to the centre O_i of the particle. The boundary conditions are then

$$\mathbf{v} = \mathbf{u}_i + \mathbf{\omega}_i \times \mathbf{r}_i \quad \text{on} \quad r_i = a_i \quad (i = 1, 2) \tag{1.2a}$$

 $\mathbf{v} \to 0 \quad \text{as} \quad |\mathbf{x} - \mathbf{x}_0| \to \infty.$ (1.2b)

By virtue of the linearity of the Stokes equation and boundary conditions that relate to fluid and particle velocities, the analysis of the flow problem may be separated into two problems: (i) the axisymmetric translational motion of the particles along their line of centres and (ii) their asymmetric translational motion along and rotational motion about a direction perpendicular to the line of centres. An exact solution for the axisymmetric translational motion was first given by Stimson & Jeffery (1926) for two spherical particles moving with the same velocity along their line of centres. Using bipolar co-ordinates, they obtained the solution in the form of an infinite series, which converges rapidly except when the particles are close together. Calculated values of the forces were reported only for equal-sized spheres. With slight modification of the boundary conditions they used, the forces may also be calculated for the general case when two unequal spheres move with different velocities. Dean & O'Neill (1963) and O'Neill (1964a) have extended the use of bipolar co-ordinates to asymmetric problems. This technique has been used by O'Neill (1964b) and independently by Goldman, Cox & Brenner (1966) and Wakiya (1967) to solve the asymmetric problem of two equal spherical particles translating along and/ or rotating about axes perpendicular to their line of centres. For unequal spheres calculations have recently been reported by Davis (1969) and O'Neill & Majumdar (1970a, b). Here also the solution is expressed as an infinite series; however in these instances the coefficients of the various terms cannot be expressed in terms of known analytical functions but rather take the form of a solution of an infinite set of difference equations. As with the other cases mentioned, the rate of convergence of these series decreases with smaller clearance between the particles; the series, in fact, diverge if the particles are in contact. The solution in bispherical co-ordinates of the Stokes equation for the fluid motion in the vicinity of spherical particles moving in a linear shear flow has been discussed by Curtis & Hocking (1970), Davis (1971) and Lin, Lee & Sather (1970). The last of these solutions, which treats the general case of arbitrary fluid and particle motions, was used in making the calculations of the present study.

Under conditions of negligible particle inertia the equations of motion of the particles are obtained by equating both the total forces and total couples exerted on the particles to zero. The forces exerted on particle i are the hydrodynamic force \mathbf{F}_i and the gravitational and buoyancy forces, hence we have

$$\mathbf{F}_{i} + \frac{4}{3}\pi a_{i}^{3}(\rho_{i} - \rho)\,\mathbf{g} = 0, \tag{1.3}$$

where \mathbf{g} is the gravitational acceleration. If the distribution of mass within the particles is uniform, the centres of buoyancy and mass coincide. Then no external couples act on the particles, so for negligible particle inertia the hydrodynamic couple \mathbf{T}_i (about O_i) satisfies

$$\mathbf{T}_i = 0. \tag{1.4}$$

From (1.1)-(1.4) it is clear that the translational motion of the particles lies in the plane of the vectors **r** and **g** and the only non-vanishing component of their rotational velocities is that about the direction normal to the plane of **r** and **g**. Let us denote by u_i and v_i , respectively, the components of \mathbf{u}_i along the line

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of centres and perpendicular to it in the plane of the motion, and by F_i^{ϵ} and F_i^{θ} the corresponding components of the hydrodynamic force \mathbf{F}_i . Also let ω_i and T_i be the components of $\boldsymbol{\omega}_i$ and \mathbf{T}_i in the direction normal to the plane of the motion. Then (1.3) and (1.4) become

$$\begin{pmatrix} F_i^r \\ F_i^\theta \end{pmatrix} + \frac{4}{3}\pi a_i^3(\rho_i - \rho) g \begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$
(1.5*a*)

(1.5b)

and

where θ is the angle between **r** and $-\mathbf{g}$.

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Since the hydrodynamic force and couple are linear functions of the fluid velocity **v**, it follows from (1.1) and (1.2) that they must be linearly related also to \mathbf{u}_i and $\mathbf{\omega}_i$. The relations between these quantities for particle *i* are (Brenner 1963, 1964*a*, *b*; Brenner & O'Neill 1972)

 $T_{i} = 0,$

$$F_{i}^{r} = -6\pi\mu a_{i} \sum_{j=1}^{2} B_{ij} u_{j}, \qquad (1.6a)$$

$$F_{i}^{\theta} = -6\pi\mu a_{i} \sum_{j=1}^{2} \left(A_{ij}v_{j} + a_{2}C_{ji}\omega_{j}\right)$$
(1.6b)

and

$$T_{i} = -8\pi\mu a_{i}^{2} \sum_{j=1}^{2} \left(\frac{3}{4} \frac{a_{2}a_{j}}{a_{i}^{2}} C_{ij} v_{j} + a_{2} D_{ij} \omega_{j} \right).$$
(1.6c)

The resistance coefficients A_{ij} , B_{ij} , C_{ij} and D_{ij} are non-dimensional scalar functions of only the ratio $a = a_2/a_1$ of the radii of the particles and the distance $\xi = r/a_1$ between the particle centres. Because of the dynamical equivalence of the two problems that are obtained when the particles are interchanged, the resistance coefficients satisfy the following useful relations:

$$K_{11}(a^{-1},\xi a^{-1}) = K_{22}(a,\xi), \quad K_{12}(a^{-1},\xi a^{-1}) = K_{21}(a,\xi), \qquad (1.7a,b)$$

$$L_{11}(a^{-1},\xi a^{-1})=aL_{22}(a,\xi),\quad L_{12}(a^{-1},\xi a^{-1})=aL_{21}(a,\xi),\qquad (1.7\,c,d)$$

where K_{ij} denotes either A_{ij} or B_{ij} and L_{ij} denotes either C_{ij} or D_{ij} . By means of these relations values of the resistance coefficients are known for all a if we have values for them for $0 < a \leq 1$.

Not all of the sixteen resistance coefficients that appear in (1.6) are independent because of the axial symmetry of the two-sphere system about the line of centres. The number of independent coefficients is, in fact, thirteen, the other three being related to them by

$$A_{12} = aA_{21}, \quad B_{12} = aB_{21}, \quad D_{12} = a^2D_{21}.$$
 (1.8*a*, *b*, *c*)

The equations of motion of the particles that result when (1.5) is combined with (1.6) can be inverted to give expressions for the components u_i , v_i and ω_i of the particle velocities in terms of the particle radii and densities and the coordinates ξ and θ . These expressions can be put into non-dimensional form by using a_1 , the radius of the larger sphere, as the unit of length, $u_1^{(\infty)}$, the terminal Stokes velocity of the larger sphere, as the measure of the translational velocity and $u_1^{(\infty)}/a_1$ as the characteristic rotational velocity. The expressions for the nondimensional velocity components $U_i = u_i/u_1^{(\infty)}$, $V_i = v_i/u_1^{(\infty)}$ and $\Omega_i = a_1\omega_i/u_1^{(\infty)}$ involve only two parameters, a and $I = (\rho_2 - \rho)/(\rho_1 - \rho)$:

$$\hat{U}_1 = U_1 / \cos \theta = (B_{22} - Ia^2 B_{12}) / (B_{11} B_{22} - a^{-1} B_{12}^2)$$
(1.9*a*)

and

$$D_1 = U_1 / \cos \theta = (B_{22} - Ia^2 B_{12}) / (B_{11} B_{22} - a^{-1} B_{12})$$
(1.9*a*)

$$U_2 = U_2 / \cos \theta = (Ia^2 B_{11} - a^{-1} B_{12}) / (B_{11} B_{22} - a^{-1} B_{12}^2)$$
(1.9b)

with similar equations for $\hat{V}_i = V_i / \sin \theta$ and $\hat{\Omega}_i = \Omega_i / \sin \theta$. Using the interchange relation (1.7), we obtain the following:

$$\widehat{U}_1(\xi a^{-1}, a^{-1}, I^{-1}) = \widehat{U}_2(\xi, a, I) / Ia^2, \tag{1.10a}$$

$$\hat{V}_1(\xi a^{-1}, a^{-1}, I^{-1}) = \hat{V}_2(\xi, a, I)/Ia^2$$
(1.10b)

$$\widehat{\Omega}_1(\xi a^{-1}, a^{-1}, I^{-1}) = \widehat{\Omega}_2(\xi, a, I)/Ia^2.$$
(1.10c)

and

The relative motion of the particles is conveniently described in terms of particle trajectories, which express how the non-dimensional separation varies with the polar angle θ . To determine the trajectories we resolve the particle velocities in spherical co-ordinates (ξ, θ, ϕ) with the origin at the centre of particle 2, as depicted in figure 1. The translational velocity of the centre of the ith particle may be expressed in the form

$$\mathbf{u}_i/u_1^{(\infty)} = -\hat{U}_i(\xi, a, I)\cos\theta \,\mathbf{e}_r + \hat{V}_i(\xi, a, I)\sin\theta \,\mathbf{e}_\theta. \tag{1.11}$$

For the non-dimensional velocity of particle 1 relative to that of particle 2 we have

$$\mathbf{u_{12}}/u_1^{(\infty)} \equiv (\mathbf{u_1} - \mathbf{u_2})/u_1^{(\infty)} = -\hat{U}_{12}(\xi, a, I)\cos\theta \,\mathbf{e_r} + \hat{V}_{12}(\xi, a, I)\sin\theta \,\mathbf{e_{\theta}}.$$
 (1.12)

The centre of the particle of radius a_1 represents a 'material' point which moves in the plane of \mathbf{e}_r and \mathbf{e}_{θ} with velocity \mathbf{u}_{12} . This point does not move with a fluid element except for $\xi \to \infty$; therefore the divergence of \mathbf{u}_{12} is different from zero for all finite ξ .

The kinematical equations determining the paths of the spheres and hence the change in relative configuration are $d\mathbf{r}/dt = \mathbf{u}_{12}$, with \mathbf{u}_{12} given by (1.12). The trajectories of particle 1 relative to particle 2 are the integrals of the two scalar equations

$$d\xi/d\tau = -\hat{U}_{12}(\xi, a, I)\cos\theta$$
 (1.13*a*)

and

$$d\theta/d\tau = \xi^{-1} V_{12}(\xi, a, I) \sin \theta, \qquad (1.13b)$$

where $\tau = (a_1/u_1^{(\infty)})t$ is the dimensionless time. By eliminating τ from these equations we obtain

$$\sin\theta / \sin\theta_0 = \exp\left\{-\int_{\xi_0}^{\xi} \frac{\hat{V}_{12}}{\xi \hat{U}_{12}} d\xi\right\},\tag{1.14}$$

where (ξ_0, θ_0) gives an initial relative position of the particles and hence serves to designate the particular trajectory.

2. Far-field asymptotic forms for the particle velocities

When the distance r between the centres of the two particles is large compared with two radii a_1 and a_2 , the hydrodynamic forces and couples exerted on the particles can be approximated by asymptotic forms in inverse powers of ξ . Expressions for the forces on two unequal particles translating along their line of centres have been evaluated to terms $O(\xi^{-5})$ by Happel & Brenner (1965, chap. 6) using the method of reflexions. Wakiya (1957) has solved the problem of two spherical particles which are free to rotate as they translate perpendicular to their line of centres by a similar technique.[†] Equating these expressions with the appropriate external forces in (1.5*a*) and solving for the velocity components results in the following expressions for the velocities of two widely separated particles subjected to gravitational forces:

$$\hat{U}_1 = 1 + \frac{3}{2}Ia^3\xi^{-1} - \frac{1}{2}Ia^3(1+a^2)\xi^{-3} - \frac{15}{4}a^3\xi^{-4} + o(\xi^{-6}), \qquad (2.1a)$$

$$\hat{U}_2 = Ia^2 + \frac{3}{2}\xi^{-1} - \frac{1}{2}(1+a^2)\xi^{-3} - \frac{15}{4}Ia^3\xi^{-4} + o(\xi^{-6}), \qquad (2.1b)$$

$$\hat{V}_1 = 1 + \frac{3}{4}Ia^3\xi^{-1} + \frac{1}{4}Ia^3(1+a^2)\xi^{-3} + o(\xi^{-6})$$
(2.1c)

and

$$V_2 = Ia^2 + \frac{3}{4}\xi^{-1} + \frac{1}{4}(1+a^2)\xi^{-3} + o(\xi^{-6}).$$
(2.1d)

The expressions for the rotational velocities of the particles under couple-free conditions $(T_1 = T_2 = 0)$ are

$$\hat{\Omega}_{1} = -\hat{V}_{1}\left[\frac{9}{16}a\xi^{-3} + \frac{3}{16}a(1 + \frac{27}{16}a + a^{2})\xi^{-5}\right] + \hat{V}_{2}\left(\frac{3}{4}a\xi^{-2} + \frac{27}{64}a^{2}\xi^{-4}\right) + o(\xi^{-6}) \quad (2.2a)$$

and

$$\hat{\Omega}_2 = -\hat{V}_1(\frac{3}{4}\xi^{-2} + \frac{27}{64}a\xi^{-4}) + \hat{V}_2[\frac{9}{16}a\xi^{-3} + \frac{3}{16}a(1 + \frac{27}{16}a + a^2)\xi^{-5}] + o(\xi^{-6}). \quad (2.2b)$$

It is easy to show that these results are consistent with the interchange relations (1.10).

3. Near-field asymptotic forms for the particle velocities

Analytic asymptotic forms for the resistance coefficients may also be found for cases where the distance between the centres of the two spheres approaches the limiting value $a_1 + a_2$. It has been found necessary to use these asymptotic forms in order to be able to describe the motion of the spheres when they are very close to each other because, as already noted, the series forms of the exact solutions either fail to converge or converge too slowly for computational convenience. Because of the singular nature of the formal solutions in the limit $r \rightarrow a_1 + a_2$, the procedure usually adopted (see for instance O'Neill & Stewartson 1967; Cooley & O'Neill 1968) is to match asymptotic expansions of the solutions for the inner region of strong shear flow in the small gap between the particles and the outer region away from the gap where the velocity gradients are relatively weaker. For our purposes, however, the complete matched asymptotic solution is not required, and we can use instead the partial solutions obtained by Cooley & O'Neill (1969b) for the problem in which a sphere of radius a_1 approaches a stationary sphere of radius a_2 with constant speed along the line of centres and O'Neill & Majumdar (1970b) for the case in which a sphere of radius a_1 translates along and rotates about the direction perpendicular to the line joining its centre to that of a stationary sphere of radius a_2 . In both cases only the terms of the asymptotic expansion of the solution for the inner region

[†] These results may be found also in chapter 6 of Happel & Brenner (1965).

were derived. However, as they have shown, this is sufficient to describe completely the singular nature of the forces and couples in the limit as the dimensionless clearance $\delta = \xi - (1 + a)$ tends to zero, provided that a matching outer solution exists. The contributions to the forces and couples from the flow in the outer region, which are required in order to obtain their non-singular parts. must be O(1). Although these contributions may, at first, seem insignificant for very small gap widths, it is evident that they are of the same order of magnitude as the gravity and buoyancy forces and hence must be included in the determination of the particle motions. In lieu of calculating the contributions of the outer solution by continuing the singular perturbation analysis, an estimate of them can be found by matching the asymptotic expansion of the inner solution with the numerical data obtained from the series solution at some small value of δ . as has been suggested by Goldman, Cox & Brenner (1967)[†] and O'Neill & Majumdar (1970b). The expressions for the velocity components, as we later show, may then further be improved by making use of the exact solutions for two touching spheres of Goren (1970), Cooley & O'Neill (1969a) and Nir & Acrivos (1973).

Using the result of Cooley & O'Neill (1969b) for the singular part of the forces acting on the spheres when one of them moves along the line of centres and the other is stationary and the interchange relations (1.7) together with the symmetry relations (1.8), we obtain the following asymptotic forms for the resistance coefficients when $\delta \ll 1$:

$$B_{11} = -\beta(\delta, a) + C_{01}(a), \quad B_{12} = \beta(\delta, a) + C_{02}(a)$$
(3.1*a*, *b*)

and

$$B_{22} = -a^{-1}\beta(\delta, a) + C_{03}(a), \qquad (3.1c)$$

$$\beta(\delta, a) = a^2(1+a)^{-2}\delta^{-1} + \frac{1}{5}(1+a)^{-3}a(1+7a+a^2)\log\delta^{-1} \qquad (3.2)$$

where (3.2)and the $C_{0i}(a)$ are contributions O(1) which are obtained from the matching of

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the inner and outer solutions. When the spheres are in contact, $u_1 = u_2 = u$ and the forces on each sphere given by (1.6) become

$$F_1^r = -6\pi\mu a_1 u f_1, \quad F_2^r = -6\pi\mu a_1 u f_2 \quad (\delta \ll 1), \tag{3.3a,b}$$

where $f_1 = C_{01} + C_{02}$ and $f_2 = a(C_{02} + C_{03})$. Values of f_1 and f_2 were calculated for a range of values of a by Cooley & O'Neill (1969a) and by Goren (1970). Combining (1.5) and (1.6) with (3.1) and solving for U_1 and U_2 , we find that the dimensionless relative velocity U_{12} is given by

$$U_{12} \simeq -\frac{Ia^3 f_1 - f_2}{\beta(f_1 + f_2)} \cos \theta \quad (\delta \ll 1),$$
(3.4)

where terms O(1) in the denominator have been neglected. The expressions for the non-dimensional velocities of the individual spheres are, to the same order of accuracy,

$$U_1 \simeq U_2 \simeq \frac{1 + Ia^3}{f_1 + f_2} \cos \theta \quad (\delta \ll 1).$$
(3.5)

Calculated values of \hat{U}_1 and \hat{U}_2 for a = 0.2 and 0.5 and for several values of I are given in table 2.

† These authors have found that the slopes of the asymptotic and the exact resistance coefficients vs. the dimensionless gap width δ are in good agreement for a range of very small values of δ , and thus may be used to evaluate the leading term of the outer solution.

Resistance		
coefficient		
$= L_n \log \delta^{-1} + l_n$	$L_n(a)$	l_n
A_{11}	$L_{5} = \frac{4}{15}a(1+a)^{-3}(2+a+2a^{2})$	l_{5}
A_{12}	$L_6 = -L_5$	l ₆
C_{11}	$L_7 = -\frac{2}{15}(1+a)^{-2}(4+a)$	l,
C_{21}	$L_8 = -L_7(a^{-1})$	l_8
A_{22}	$L_{10} = a^{-1}L_5$	l ₁₉
C_{12}	$L_{11} = -a^{-1}L_7$	<i>l</i> ₁₁
C_{22}	$L_{12} = a^{-1} L_7(a^{-1})$	l_{12}
D_{11}	$L_{15} = \frac{2}{5} (1+a)^{-1}$	l ₁₅
D_{12}	$L_{16} = \frac{1}{4}aL_{15}$	l ₁₆
D_{22}	$L_{20} = L_{15}$	l_{20}
TABLE 1. Near-	field asymptotic forms for the resistance	coefficients

With these results we can examine the restrictions imposed on the magnitude of the Reynolds number when $\delta \ll 1$ by the condition that inertial forces of the fluid motion in the gap be much smaller than the viscous forces. The length and time scales of the motion in the gap are $a_1\delta$ and $a_1\delta/u_{12}$, respectively, so the ratio of inertial to viscous forces is $a_1 \delta \rho u_{12}/\mu$. The condition that inertial forces are negligible can be written as $Re_1 \ll (U_{12}\delta)^{-1}$, which is $O(\delta^2)$ from (3.4) and (3.2). Consequently if $Re_i \ll 1$ inertial forces are everywhere negligible compared with the viscous forces for all configurations of the particles.

In the case of motion perpendicular to the line of centres O'Neill & Majumdar (1970b)⁺ have shown that each resistance coefficient in (1.6) exhibits a logarithmic singularity of the form $L_n(a)\log \delta^{-1} + l_n(a).$

Analytical expressions for the $L_n(a)$ of each of the resistance coefficients are given in table 1. Values of the $l_n(a)$ may be estimated by means of the exact computed coefficients in the manner described above. By virtue of the symmetry relations only ten of the resistance coefficients are independent.

Substitution of the asymptotic forms of the resistance coefficients given in table 1 into (1.6) gives, after some cumbersome algebra, the following results for the velocities V_i and Ω_i :

$$V_{1} = \hat{V}_{1} \sin \theta = \frac{\lambda_{11} (\log \delta^{-1})^{2} + \lambda_{21} \log \delta^{-1} + \lambda_{31}}{(\log \delta^{-1})^{2} + \Delta_{2} \log \delta^{-1} + \Delta_{3}} \sin \theta \quad (\delta \ll 1), \qquad (3.6a)$$

$$V_{2} = \hat{V}_{2}\sin\theta = \frac{\lambda_{12}(\log\delta^{-1})^{2} + \lambda_{22}\log\delta^{-1} + \lambda_{32}}{(\log\delta^{-1})^{2} + \Delta_{2}\log\delta^{-1} + \Delta_{3}}\sin\theta \quad (\delta \ll 1), \qquad (3.6b)$$

$$a\Omega_{1} = a\widehat{\Omega}_{1}\sin\theta = (\gamma_{1} + \gamma_{2}\widehat{V}_{1} + \gamma_{3}\widehat{V}_{2})\sin\theta \quad (\delta \leqslant 1)$$

$$a\Omega_{2} = a\widehat{\Omega}_{2}\sin\theta = (\gamma_{4} - \gamma_{5}\widehat{V}_{1} - \gamma_{6}\widehat{V}_{2})\sin\theta \quad (\delta \leqslant 1).$$
(3.7*a*)
(3.7*b*)

(3.7b)

and

The coefficients γ_i , Δ_i and λ_{ij} are functions of the coefficients L_n and l_n and the parameters a and I.⁺ Taking the limit of V_1 and V_2 as $\delta \rightarrow 0$ in these expressions,

[†] Results for two equal spheres have been given by Goldman et al. (1967), O'Neill (1969) and by Zia, Cox & Mason (1967).

[‡] Copies of a summary of the relations among the coefficients are available on request from either the editor or the authors.



FIGURE 2. Notation used for description of motion of two touching spheres. All quantities are dimensionless. The forces F_{h}^{agg} and F_{g}^{agg} act at O_{ζ} in the direction of V, and the couples T_{h}^{agg} act in the direction of Ω .

we see that λ_{11} and λ_{12} are the exact values of \hat{V}_1 and \hat{V}_2 when the gap width equals zero, that is, $\lambda_{1j} = \hat{V}_j^{(T)} \equiv \lim_{\delta \to 0} \hat{V}_j$ for j = 1 and 2.

It is of interest to examine the relations between V_1 , V_2 , Ω_1 and Ω_2 in the limit $\delta \rightarrow 0$ to see what conclusions can be obtained for the motion of spherical particles in contact. From (3.6) and (3.7)

$$\Omega_1 = \Omega_2 = \Omega \quad \text{when} \quad \delta = 0 \tag{3.8}$$

and

$$V_1^{(T)} - V_2^{(T)} = (1+a)\Omega$$
 when $\delta = 0.$ (3.9)

Now, since Ω_i is a free vector which may be displaced parallel to itself, it is obvious that there is no relative rotation between the touching spheres and that Ω is the common angular velocity of the two-sphere aggregate about the instantaneous axis of rotation. Consequently, the difference $V_1 - V_2$ between the translational velocities of the particle centres is solely due to the rigid-body rotation of the pair. This provides a rigorous proof of the fact that two touching spheres moving under gravity in an inertialess quiescent viscous fluid undergo a rigidbody rotation. Similar behaviour is found also when an aggregate of two touching spheres is freely suspended in a linear shear flow, as has been shown both by the analysis of Nir & Acrivos (1973) and by the observations of Mason and coworkers (Bartok & Mason 1957; Zia *et al.* 1967).

We can use the results in the paper by Nir & Acrivos to determine exact limiting values for V_i and Ω_i when $\delta \to 0$ in order to improve the results (3.6) and (3.7). Because of the linearity of the Stokes equation of motion, one can write the total hydrodynamic force F_h^{agg} in the direction perpendicular to the line of centres of the two-sphere aggregate and the total hydrodynamic couple T_h^{agg} about a point O_{ζ} (see figure 2) as

$$F_h^{\text{agg}} = -\pi\mu a_1 u_1^{(\infty)} (\alpha_1 V + \delta_1 \Omega)$$
(3.10)

$$T_{h}^{\text{agg}} = -\pi\mu a_{1}^{2} u_{1}^{(\omega)}(\delta_{1} V + \beta_{1} \Omega), \qquad (3.11)$$

in which V and Ω are, respectively, the (dimensionless) translational and angular velocities of the aggregate relative to the point O_{ζ} , which is located along the line of centres at a distance $a_1\zeta$ from the point of contact towards O_1 . The dimensionless distance ζ and resistance coefficients α_1 , β_1 and δ_1 are given in tables 1 and 2 of Nir & Acrivos as functions of the particle size ratio.† It should be noted that the point O_{ζ} represents in their work the location of the centre of rotation at which, in the absence of external forces on the agggregate, the translational velocity component perpendicular to the line of centres exactly equals the corresponding component of the undisturbed field. In our case O_{ζ} is simply the point about which the resistance elements of the coupling and rotation dyadics are calculated. Now, the net external force on the aggregate in the direction perpendicular to the line of centres of the particles is

$$F_g^{\text{agg}} = -\frac{4}{3}\pi a_1^3(\rho_1 - \rho)g(1 + Ia^3)\sin\theta$$
(3.12)

and the external couple about O_{ζ} is

$$T_g^{\text{agg}} = -\frac{4}{3}\pi a_1^4(\rho_1 - \rho)g(1 + Ia^3) \left\{ \frac{1 - Ia^4}{1 + Ia^3} - \zeta \right\} \sin \theta.$$
(3.13)

Equating the sums of the forces and couples acting on the aggregate to zero and solving for V and Ω , we have

$$V = -6(\alpha_1\beta_1 - \delta_1^2)^{-1} \{ (1 + Ia^3)(\beta_1 + \delta_1\zeta) - (1 - Ia^4)\delta_1 \} \sin\theta$$
(3.14)

and
$$\Omega = -6(\alpha_1\beta_1 - \delta_1^2)^{-1}\{(1 - Ia^4)\alpha_1 - (1 + Ia^3)(\delta_1 + \alpha_1\zeta)\}\sin\theta.$$
 (3.15)

Since the aggregate is in rigid-body rotation, the translational velocities of the particule centres are

$$V_{1}^{(T)} = V + (1 - \zeta) \Omega \quad (\delta = 0)$$
(3.16a)

and

$$V_1^{(T)} = V + (1 - \zeta) \Omega \quad (\delta = 0)$$

$$V_2^{(T)} = V - (a + \zeta) \Omega \quad (\delta = 0),$$
(3.16b)
(3.16b)

from which the values in table 2 were calculated. These expressions are consistent with the result given in (3.9).

4. Relative particle trajectories

The principal results of this paper are the relative trajectories $\xi(\theta)$ of two spherical particles as they fall (or rise) in a quiescent fluid. Because the trajectories are directly dependent on the relative velocity functions $U_{12}(\xi, a, I)$ and $\hat{V}_{12}(\xi, a, I)$, it will prove helpful to preface the description of the trajectories with a discussion of the velocity functions. These are displayed in graphical form in

† In the paper by Nir & Acrivos, dimensional coefficients a_1, b_1 and d_1 are used instead of α_1, β_1 and δ_1 . The relationship between the two sets of coefficients is $\alpha_1 = a_1/\pi\mu a_I$, $\beta_1 = b_1/\pi\mu a_I^3$ and $\delta_1 = d_1/\pi\mu a_I^2$, where a_I is the radius of the larger of the two spheres.

and

	For spi	heres in contact (ξ =	= 1 + a)				
a = 0.5							
Ι	$\vec{U}_1=\vec{U}_2$	$\hat{V}_{1}^{(T)}$	$\hat{V}_{2}^{(T)}$	$\hat{V}_{12}^{(T)}$			
0	0.953631	0.927309	0.713115	0.214194			
1	1.072834	1.016449	0.904161	0.112288			
1.5	$1 \cdot 132436$	1.061018	0.999683	0.061335			
1.68406	1.154377	1.077425	1.034847	0.042578			
2	$1 \cdot 192038$	$1 \cdot 105588$	1.095206	0.010382			
$2 \cdot 10188$	$1 \cdot 204183$	$1 \cdot 114670$	$1 \cdot 114670$	0.000000			
3	1.311242	$1 \cdot 194728$	1.286252	-0.091524			
4	$1 \cdot 430446$	$1 \cdot 283867$	$1 \cdot 477297$	-0.193430			
5	1.549650	1.373006	1.668343	-0.295337			
		a = 0.2					
0	0.997871	0.982764	0.947294	0.035470			
1	1.005854	0.990343	0.962644	0.027699			
2.5	1.017828	1.001710	0.985669	0.016041			
2.89379	1.020732	1.004467	0.991253	0.013214			
4	1.029802	1.013078	1.008694	0.004384			
4.56414	1.034284	1.017353	1.017353	0.000000			
6	1.045768	1.028234	1.039394	-0.011160			
15	1.117615						

For spheres far apart $(\xi \to \infty)$ $\dot{U}_1, \dot{V}_1 \rightarrow 1, \quad \dot{U}_2, \dot{V}_2 \rightarrow Ia^2, \quad \dot{U}_{12}, \dot{V}_{12} \rightarrow 1 - Ia^2$

TABLE 2. Limiting values for the particle velocity functions

figures 3 and 4, which show how \hat{U}_{12} and \hat{V}_{12} vary with ξ for a = 0.2 and 0.5and for several values of the relative particle density I. Several interesting features of these curves may be noted. For small values of I the functions \hat{U}_{12} and V_{12} , for a given value of a, are positive for all values of ξ and increase monotonically from their limiting values when the spheres are in contact at $\xi = 1 + a$, which are given in table 2. As I is increased a critical value is reached where $(d\hat{U}_{12}/d\xi)_{\xi=1+a}$ equals zero. For larger values of I the limiting slope is negative, and \hat{U}_{12} itself is negative for a range of values of ξ . The critical value of the relative density I_c^U , which depends on the particle size ratio a, can be calculated from the values of f_1 and f_2 [cf. equations (3.3a, b)] given in the papers by Cooley & O'Neill (1969a) and Goren (1970). Differentiation of \hat{U}_{12} in (3.4) with respect to ξ and setting $(d\hat{U}_{12}/d\xi)_{\xi=1+a}$ equal to zero gives

$$I_c^U = f_2 / a^3 f_1. (4.1)$$

Values of I_c^U for several values of a are given in table 3. As I is increased above I_c^U , the range of values of ξ (between 1 + a and ξ_U^* , say) for which \hat{U}_{12} remains negative increases with I. An estimate of the upper limit ξ_U^* of the range is obtained from the three-term approximations of the far-field equations (2.1a, b), which give

$$\xi_U^* \simeq \frac{1}{2}\kappa[3 - (1 + a^2)/\xi_U^*], \qquad (4.2)$$

where $\kappa = (1 - Ia^3)/(1 - Ia^2)$ has values between 0 and 1. More accurate values, obtained from the calculations of \hat{U}_{12} , are given in table 4 for several (I, a) pairs.



Similar behaviour is found for the relative velocity function \hat{V}_{12} for the perpendicular direction. As I is increased a second critical value I_c^V , always greater than I_c^U except in the limit as $a \to 1$, when both I_c^U and I_c^V equal unity, is reached at which \hat{V}_{12} changes from a monotonic positive-valued function to one which has negative values for ξ in some range $1 + a \leq \xi < \xi_{\mathcal{V}}^*$. Negative values of \widehat{V}_{12} first appear for $\xi = 1 + a$; consequently the critical values I_c^{∇} can be determined by



FIGURE 4. The relative velocity functions $\hat{U}_{12}(\xi)$ and $\hat{V}_{12}(\xi)$ for a = 0.2.

equating the expression for $\hat{V}_1^{(T)} - \hat{V}_2^{(T)}$ obtained from (3.15) and (3.16) to zero. This gives $I^V = \sigma^{-3} \frac{\alpha_1(1-\zeta) - \delta_1}{\alpha_1(1-\zeta) - \delta_1}$ (4.3)

$$I_{c}^{\nu} = a^{-3} \frac{\alpha_{1}(1-\zeta) - \delta_{1}}{\alpha_{1}(a+\zeta) + \delta_{1}},$$
(4.3)

from which the values in table 3 were calculated. The upper limit ξ_{V}^{*} can also be estimated from the far-field forms of (2.1), which yield

$$\xi_V^* \simeq \frac{1}{4}\kappa[3 + (1 + a^2)/\xi_V^{*2}]. \tag{4.4}$$

It is easy to see from (4.2) and (4.4) that $\xi_U^* > \xi_V^*$ for all values of *I* and *a* such that $Ia^2 < 1$. This is found to be the case also for the more accurate values of ξ_V^*

	a^{-1}	I_c^U	I_c^V	
	8	4.844	00	
	20	$4 \cdot 20000$		
	10	3.64982	6.66060	
	8	$3 \cdot 42128$		
	6	3-12838	<u></u>	
	5	2.86379	4.56414	
	4	2.57344		
	3	2.19330	3.07743	
	2.5	1.95777		
	2	1-68406	2.10188	
	1.5	1.36600	1.55857	
	$1 \cdot 2$		1.22281	
	$1 \cdot 125$	1.09592		
	1.01		1.01103	
	1	1	1	
	TABL	E 3. Values of I_c^U and	I_c^V	
a	I	ξ * ξ _U	ζ* 5γ	
0.5	2	1.78227	+	
0.5	3	3.5378	2.01740	
0.2	4	1.31632	†	
0.2	5	3.0949	1.80303	
	\dagger Not	applicable because I	$< I_c^V$	
	TABI	E 4. Values of ξ_U^* and	1 5 *	

obtained from the numerical calculations of \hat{V}_{12} and given in table 4. The dependence of I_c^U and I_c^V on a is summarized in figure 5.[†]

The hydrodynamic forces exerted on the particles when they move with equal velocity through a quiescent fluid can readily be determined from the resistance coefficients. When the particles move parallel to their line of centres with velocity u and separation ξa_1 , the force components F_1^r and F_2^r are given by (3.3). The dimensionless forces $f_1(\xi)$ and $f_2(\xi)$ have been calculated by Cooley & O'Neill (1969*a*) for different relative sphere sizes. They find that the force f_2 on the smaller sphere is a monotonically increasing function as ξ varies from 1 + a to ∞ . However, the force on the larger sphere is monotonic in ξ only if a is less than a critical value, which they estimate to be about 0.7. For $a \geq 0.7$, f_1 exhibits a minimum at some value of ξ larger than 1 + a. Identical behaviour is found for the hydrodynamic forces exerted on the particles when they move under couple-free conditions perpendicular to their line of centres with the same velocity v. The dimensionless forces g_1 and g_2 , defined by

and

$$F_{1}^{\theta} = -6\pi\mu a_{1}vg_{1}, \quad F_{2}^{\theta} = -6\pi\mu a_{1}vg_{2}$$
$$T_{1} = T_{2} = 0,$$

† The individual particle velocity functions \hat{U}_i and V_i are affected in a similar manner by the value of I. It is found from the calculations (for a = 0.5) that \hat{U}_1 is a monotonic function of ξ , while \hat{U}_2 is monotonic for I = 0 and 1.5 but exhibits maxima for I = 2 and 3. Similarly, \hat{V}_2 is monotonic, while \hat{V}_1 is monotonic when I = 0 but has maxima when I = 1.5, 2 and 3.



FIGURE 5. Dependence of I_c^U and I_c^V on a.

are shown as functions of ξ in figures 6(a) and (b). We note that the force g_2 on the smaller sphere increases monotonically with ζ , while g_1 is monotonic only if a is below about 0.7. For $a \lesssim 0.7$, g_1 also passes through a minimum at some separation greater than that at contact.

As is to be expected from the preceding discussion, the shape $\xi(\theta)$ of the relative particle trajectories is strongly affected by the relative particle density. For each value of the size ratio *a* four types of trajectories are obtained, depending on the value of *I*. Figure 5 shows these four regions on a plot of Ia^2 , the ratio of the terminal velocities of the particles moving by themselves in the ambient fluid, *vs.* the particle size ratio.

(a) $0 \leq I < I_c^U$. In this case both \hat{U}_{12} and \hat{V}_{12} are positive-valued functions of ξ . Hence, from (1.3) $d\xi/d\tau < 0$ and $d\theta/d\tau > 0$ for $0 < \theta < \frac{1}{2}\pi$, so the larger particle moves downward relative to the smaller one, passing around and below it. The trajectories extend to $\xi = \infty$ both above and below the smaller sphere and are symmetric about $\theta = \frac{1}{2}\pi$. Some sample trajectories for this case, calculated for a = 0.5 and I = 1.5 from (1.14), are shown in figure 7(a).

(b) $I_c^U < I < I_c^V$. For I between I_c^U and I_c^V only \hat{V}_{12} is positive for all values of ξ . The relative velocity \hat{U}_{12} along the line of centres is negative for $1 + a < \xi < \xi_U^*$ and positive for $\xi > \xi_U^*$; hence for $0 < \theta < \frac{1}{2}\pi$, $d\xi/d\tau$ is positive for $\xi < \xi_U^*$ and negative for $\xi > \xi_U^*$, while $d\theta/d\tau$ is always positive. The trajectory plane of $\xi(\theta)$ divides, therefore, into two parts according to whether ξ is greater or less than ξ_U^* . No trajectories cross the curve $\xi = \xi_U^*$ (since $d\xi/d\tau = 0$ there). Outside this



FIGURE 6. The force on (a) the larger and (b) the smaller of two spheres moving perpendicular to their line of centres.

curve the trajectories extend to infinity in both directions as in (a), the larger sphere again moving downward relative to the smaller one. The trajectories inside the curve $\xi = \xi_U^*$ begin at $\theta = 0$ and $\xi = 1 + a$; as the larger sphere moves around the smaller one the centre-to-centre distance ξ increases to a maximum value at $\theta = \frac{1}{2}\pi$, whereupon it decreases in a symmetrical fashion until the particles reach a stationary configuration in which they are in contact with the smaller one directly above the larger one, i.e. $\theta = \pi$ and $\xi = 1 + a$. The duration of trajectory, denoted by T, is infinite because the hydrodynamic forces exerted on the particles become infinite as the clearance approaches zero, while

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FIGURE 7. Relative particle trajectories $\xi(\theta)$ for a = 0.5 and (a) I = 1.5, (b) I = 2($\xi_U^* = 1.78227$) and (c) I = 3 ($\xi_U^* = 3.5378$, $\xi_V^* = 2.0174$).

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the gravitational force which drives the motion is always finite. This property of the trajectories is reflected in the formula for T obtained from the integral of (1.13a) using (1.14):

$$T = \int_{0}^{T} dt = -\left(2u_{1}^{(\infty)}/a_{1}\right) \int_{1+a}^{\xi_{\max}(\xi_{0},\,\theta_{0})} \hat{U}_{12}^{-1} \left\{1 - \sin^{2}\theta_{0} \exp\left(-2\int_{\xi_{0}}^{\xi} \frac{\hat{V}_{12}}{\xi\hat{U}_{12}} d\xi\right)\right\}^{-\frac{1}{2}} d\xi,$$
(4.5)

where $\xi_{\max}(\xi_0, \theta_0)$, the maximum value of ξ (at $\theta = \frac{1}{2}\pi$), is obtained from

$$\int_{\xi_0}^{\xi_{\max}} \frac{\hat{V}_{12}}{\xi \hat{U}_{12}} d\xi = \log \sin \theta_0.$$

It is evident from (3.4) and (3.2) that the integral in (4.5) diverges. Sample trajectories for this case calculated for a = 0.5 and I = 2 are displayed in figure 7(b).

(c) $I_{U}^{V} < I < a^{-2}$. Again there are two kinds of trajectories: those in the space $\xi > \xi_{U}^{*}$, which extend to infinity both above and below the smaller sphere, and those confined to the region $1 + a < \xi < \xi_{U}^{*}$. However, in this case the finite trajectories are of a different character because \hat{V}_{12} changes sign from negative to positive as ξ becomes greater than ξ_{V}^{*} . For $\xi < \xi_{V}^{*}$ and $0 < \theta < \frac{1}{2}\pi$, $d\theta/d\tau$ is negative so θ decreases with time while ξ increases. Hence ξ is a minimum (equal to ξ_{\min} , say) at $\theta = \frac{1}{2}\pi$. The trajectory that passes through $\xi = \xi_{\min}$ and $\theta = \frac{1}{2}\pi$ continues in time with θ decreasing and ξ increasing until $\xi = \xi_{V}^{*}$, whereupon θ begins to increase along with ξ until $\theta = \frac{1}{2}\pi$. Because of the symmetry of the motion about $\theta = \frac{1}{2}\pi$. The trajectories are thus closed orbits around which the larger sphere moves relative to the smaller one in a cyclic fashion.† The period of the motion, which is finite in this case, is given by (4.5), but with the lower limit of 1 + a replaced by $\xi_{\min}(\xi_{0}, \theta_{0})$, which is determined from

$$\int_{\xi_{\min}}^{\xi_0} \frac{\hat{V}_{12}}{\xi \hat{U}_{12}} d\xi = \log \sin \theta_0.$$

Typical trajectories for this case calculated for a = 0.5 and I = 3 are shown in figure 7 (c).

(d) $I > a^{-2}$. Both \hat{U}_{12} and \hat{V}_{12} are negative for all $\xi > 1 + a$, so $d\xi/d\tau > 0$ and $d\theta/d\tau < 0$ for $0 < \theta < \frac{1}{2}\pi$. Thus, all trajectories extend to infinity in both directions, but the direction of the motion is now upward, reflecting the fact that the relative density of the smaller sphere is so large that it always falls faster than the larger sphere.

The existence of closed orbits in case (c) is reminiscent of a similar characteristic of the relative trajectories of neutrally buoyant spherical particles in laminar shear flows. Under conditions of negligible fluid and particle inertia and no external forces or couples or Brownian effects, Batchelor & Green (1972a, b) have shown analytically that there exists around each particle a region of closed trajectories along which the other particle moves in a cyclic manner. The existence of orbiting trajectories for pairs of spherical particles in linear shear flows had

[†] The trajectories as seen by a stationary observer are not closed, of course; they only appear so to an observer moving with the smaller sphere.

previously been established by the observations of Darabaner & Mason (1967). Because there has apparently been no experimental demonstration of closed orbits for the case at hand, we made some qualitative observations of a nylon sphere and a smaller glass sphere falling in castor oil. The value of a was approximately 0.38, and that of I was about 6.6, which is between $I_c^V \simeq 2.7$ and $a^{-2} \simeq 7$. Periodic motion of the larger sphere in the 'no-escape' region near the smaller sphere was clearly evident.

One of the important problems in particle mechanics is the prediction of rates of collection of small particles by larger ones as they fall through a viscous medium. Since the hydrodynamic force resisting the approach of the particles towards each other becomes infinite as $\delta \to 0$, contact evidently is never achieved. Observations show, however, that contact resulting in permanent capture does occur, apparently because electrostatic forces, slip flow and/or other effects become significant as δ becomes very small. In order to allow for these effects in model studies of the collection process, capture is said to occur whenever δ reaches a small non-zero value δ_c . The quantity of interest in these studies is the collection efficiency E of the large particle, defined as

$$E = R^2/a_1^2,$$

where a_1 is the radius of the large particle and R is the radius of the circle far above the large sphere through which the centre of the small one must pass in order for capture to occur. There have been several investigations of this problem, but none has taken account of the effect of unequal particle densities. The present work has shown that the particle densities can in certain cases have a decisive influence on the possibility of collection. In particular, if the values of I and aof a pair of particles belong to either case (b) or (c) and the particles are initially separated by a distance $\xi > \xi_U^*$, then they will never result in capture. Around each sphere there is a spherical region of radius $\xi_U^*(I, a)$ which spheres outside cannot penetrate and from which spheres inside cannot escape. The size of this region can be very large; for $Ia^2 = 1$, corresponding to equal terminal velocities of the particles, it is infinite. As an illustration of the consequence of this result consider the use of a spray of $10 \,\mu m$ water droplets to remove smaller dust particles having a density of 3g/c.c. from a gas stream. From table 3 or figure 5, $I_c^U = 3$ corresponds to $a \simeq 0.18$; thus only particles smaller than about $1.8 \,\mu \text{m}$ can be collected in this case.

Finally, the results presented here can be used to determine the effect of hydrodynamic interactions during particle encounters on sedimentation rates in suspensions. Batchelor (1972) has developed a procedure for calculating the contribution of two-sphere encounters to the sedimentation velocity of spheres and has evaluated the contribution for a suspension of spheres of uniform size and density. In order to obtain the average velocity of the particles the twoparticle configuration probability function $P(\mathbf{r}, t)$ must be known. Since equal spheres fall at the same velocity for all separations \mathbf{r} , configurations remain constant with time and hence are the same as at some initial time. Thus if the initial configuration in the suspension is random, $P(\mathbf{r}, t)$ is a constant independent of \mathbf{r} and t. Such is not the case for suspensions of dissimilar particles. The relative motion of the particles and the possibility of finite and closed trajectories will have a profound effect on the form of $P(\mathbf{r}, t)$ and consequently on the determination of the sedimentation rate. This problem will be taken up in a later report.

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