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A Mathematical Model for the Transport of Environmental Plutonium*

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The time history of particulate plutonium in the environment is governed by convective, diffusive, fluid-particle, particle-surface, and fluid-particle-surface interactions. For example, a plutonium aerosol that is initially released in the atmosphere will be convected and diffused in a manner characterized by fluid-particle interactions. Furthermore, particulate plutonium distributed on the earth's surface will adhere to the soil and either diffuse into it in a particle-surface interaction or be entrained by the earth's surface fluid layer in a fluid-particle-surface interaction. The purpose of the proposed model is to mathematically describe the behavior of particulate plutonium in the lower atmosphere, which accounts for particles settling or impinging on the earth's surface as well as particle re-entrainment or resuspension.

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We make the following assumptions: (1) The particle is spherical in shape, (2) the particle is small compared to the smallest wavelength of atmospheric motions, and (3) the particle concentration is characterized by a dilute gas-solid suspension. The first two assumptions allow us to neglect the effects of particle orientation and rotation in a turbulent shear flow. By using an experimentally determined shape factor,[1] we are able to transform the analysis of an irregularly shaped particle to an equivalent analysis of a spherical particle of either Stokes or aerodynamic diameter. As for the second assumption, we are generally concerned with respirable particulate plutonium, which is less than 10 μm in diameter. These length scales are several orders of magnitude smaller than even the microscale of turbulence[2] in the lower atmosphere. The third assumption enables us to treat each particle individually. That is, particles are separated from one another so that direct and indirect particle-particle interactions are negligible. Direct and indirect interactions involve particle collisions and fluid velocity field modifications in the space between particles, respectively.

Atmospheric motions are described through a solution of the incompressible fluid dynamical (Navier-Stokes) equations

$$\rho_f \frac{Du_{f,i}}{Dt} = - \frac{\partial p}{\partial x_i} - \rho_f g \delta_{i3} + \mu \frac{\partial^2 u_{f,i}}{\partial x_j \partial x_j} \qquad (1)$$

and the continuity equation

$$\frac{\partial u_{f,i}}{\partial x_i} = 0, \qquad (2)$$

where $u_{f,i}$ are the fluid velocity components spanning the physical space x_i, p is the pressure, ρ_f is the fluid density, μ is the dynamic viscosity, and δ_{ij} denotes the Kronecker delta (coordinate 3 is opposite to the direction of gravity). The Lagrangian or substantial derivative $\frac{D}{Dt}$ in equation (1) is defined

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + u_{f,j}\frac{\partial}{\partial x_j} ,$$

where the subscripts i and j introduce conventional Cartesian tensor notation. If we consider a momentum balance[3] on a small solid spherical rigid particle in a fluid velocity field described by equations (1) and (2), we write

$$\frac{\pi d^3}{6}\rho_p \frac{Du_{p,i}}{Dt_p} + \frac{1}{2}\frac{\pi d^3}{6}\rho_f\left(\frac{Du_{p,i}}{Dt_p} - \frac{Du_{f,i}}{Dt_p}\right) = F_i , \tag{3}$$

where $u_{p,i}$ are the particle velocity components, ρ_p is the particle density, d is the particle diameter, and F_i is the contribution of all forces acting on the particle. The two terms on the left hand side of equation (3) are, respectively: the force required to accelerate the particle and the force associated with the acceleration of the apparent mass of the particle relative to the fluid. That is, when a particle is accelerated relative to the surrounding fluid, a complex three-dimensional flow field that possesses kinetic energy is set up around the particle. Work must be supplied to move the particle in addition to that required to accelerate it alone. This extra energy requirement shows up as an additional force on the particle. The particle has inertia and thus does not necessarily follow fluid streamlines; therefore, the particle Lagrangian derivative $\frac{D}{Dt_p}$ is written

$$\frac{D}{Dt_p} = \frac{\partial}{\partial t} + u_{p,j} \frac{\partial}{\partial x_j} \quad .$$

For a particle completely immersed in a fluid, the contributing forces on the right hand side of equation (3) are:

(1) gravitational force

$$-\frac{\pi d^3}{6} \; g(\rho_p - \rho_f) \; \delta_{i3},$$

where the bouyancy force is included;

(2) viscous drag force

$$-C_D \frac{\pi d^2}{4} \; \rho_f (u_{p,i} - u_{f,i})^2/2 \quad ,$$

where C_D is the particle drag coefficient;

(3) Basset force[4] or the effect of acceleration on the viscous drag, where the growth of the viscous boundary layer on a particle is a function of the entire previous history of the particle motion,

$$- 3d^2 \, (\pi \mu \rho_f)^{1/2}/2 \int_0^t \frac{\frac{Du_{p,i}}{D\tau_p} - \frac{Du_{f,i}}{D\tau_p}}{(t - \tau)} \, d\tau ;$$

and (4) pressure force or the effect of the pressure gradient on the particle

$$-\frac{\pi d^3}{6} \frac{\partial p}{\partial x_j} \quad ,$$

where $\frac{\partial p}{\partial x_j}$ is found from the Navier-Stokes equations (1). For a particle resting on the earth's surface, we include another force, the adhesive force between the particle and the surface, which is perpendicular to the substrate. In general, the force of adhesion is very complex and may be influenced by many factors;[5] however, in the first approximation, we consider only the molecular attraction between a particle and a plane surface,[6] which is given by

$$- 2\pi \gamma d \delta_{13},$$

where γ is the total surface energy of the particle.

Equations (1), (2), and (3) constitute a set of seven equations in the seven unknowns $u_{f,i}$, $u_{p,i}$, and p. This set of equations is a coupled nonlinear stochastic integrodifferential system which, in general, is very complex and difficult to solve. To date, Hotchkiss and Hirt[7] have had the most success in solving a similar system. They numerically treat a full three-dimensional Navier-Stokes flow field [equations (1) and (2)] with each discrete particle characterized by a simplified version of equation (3). The system of equations is numerically simulated, and the results are presented by means of a computer generated film.

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