Coagulation of Smoke Aerosol in a Buoyant Plume

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Received August 1, 1978; accepted February 8, 1979

The mechanism of particulate coagulation in a turbulent plume is studied by combining the Morton–Taylor–Turner theory of turbulent buoyant plumes with the present authors' earlier analysis of coagulation in a homogeneous system. The conservation of fluid mass, particulate matter, momentum, and energy leads to a set of differential equations for horizontal averages of hydrodynamic quantities. These relations are combined with the horizontally averaged coagulation equation to yield an equation which is transformed to be exactly equivalent to the problem of coagulation in a homogeneous medium. The effective time scale is a known function of the vertical plume height which is determined by solving the plume hydrodynamic equations. This permits the coagulation process in a homogeneous system to be quantitatively related to that in a buoyant plume. Sample calculations are performed to illustrate the effects of the initial number and mass concentrations of the particulate, rate of heat release, initial plume momentum, and atmospheric stratification on the aging process. Results indicate that the coagulation process can be "frozen" if the entrainment of uncontaminated air into the plume sufficiently dilutes the particulate concentration. Calculations of the number flux and the particle size D_{32} as a function of plume height are included.

1. INTRODUCTION

Aerosol formation and growth in a buoyant plume occurs in a number of combustion processes including fires, the combustion of coal in power plants, and the production of carbon black and high purity silicon. The initial particle formation is a complex process involving chemical dynamics, nucleation, condensation, and coagulation. As the plume rises, further growth of the particles will occur primarily as a result of coagulation and condensation. Finally at some height the vapor concentration is depleted by condensation and dilution and further growth can only occur by coagulation.

It is this last stage of particle growth that is of interest in this paper. The particle formation processes of nucleation and condensation are assumed to be complete. The problem of interest is the calculation of the dependence of the size distribution of an aerosol on the plume height allowing for coagulation and air entrainment in the rising plume.

In order to proceed, it is necessary to combine a tractable model of the buoyant plume hydrodynamics with the Smoluchowski equation which describes the particulate coagulation. Since most plumes of practical interest are turbulent, it is not possible to proceed from first principles. The most useful model for the present study is that due to Morton *et al.* (1). They assumed that the mean velocity and temperature profiles across the plume were Gaussian functions of a suitably normalized radial position in the plume. They further postulated a relation between the rate at which mass was entrained by turbulent mixing into the plume and the mean vertical momentum flux. The model was extended by Morton (2) to consider variations in the character of the source and the environment. It has been used subsequently in a variety of applications. A survey of these results and

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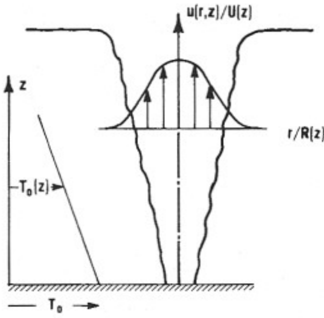

FIG. 1. Schematic of buoyant plume showing assumed mean velocity profile $U(r,z)/v(z)$ and ambient stratification $T(z)$.

their experimental verification is given in the recent monograph by Turner (3).

This model has been incorporated into our analysis in a slightly extended form, to allow for the presence of particles in the plume. No assumption is made about the size distribution, but we do take the coagulation coefficient to be constant. This permits the size distribution to emerge as a solution of the Smoluchowski equation, rather than evolve as a prescribed functional form with specified parameters determined by taking moments of the Smoluchowski equation. A key result of our work is the demonstration that the problem of coagulation in a buoyant plume can be transformed to that of the well-studied problem of coagulation in a uniform system such as a smoke box (4–7).

Delattre and Friedlander (8) have treated a somewhat similar problem, coagulation in a turbulent jet. They assume an eddy diffusion model for the turbulence and assume that the size distribution remains self-preserving. The eddy diffusion turbulence model, while quite successful in turbulent jet analyses, has not proved useful in the study of buoyant plumes. The assumption of a self-preserving size distribution avoids the necessity of solving the coagulation equation for the size distribution. However, it does so at the expense of an ability to study the size distribution in detail.

In the present work, the Smoluchowski equation is cast into a form consistent with the Morton–Taylor–Turner model in Section 2. The plume model is derived in extended form in Section 3 and the solution displayed in Section 4. The general solution of the coagulation equation is obtained in Section 5 and the correspondence between the plume solution and the homogeneous (smoke box) solution is shown in detail. Finally, numerical solutions illustrating the effects of the parameters controlling coagulation are displayed in Section 6. The number flux and particle size D_{32} are calculated for the parameter range appropriate for aging in a smokestack plume and in a plume generated by a smoldering source in an enclosure.

2. THE COAGULATION MODEL

We consider a turbulent, weakly buoyant plume whose mean (time-averaged) properties depend upon the vertical coordinate z above the plume source and a radial coordinate r measured from the mean plume centerline. The plume is laden with particulate matter and rises into a particle free atmosphere at rest. Let the mean particle distribution over the particle volume size v be $n(r,z,v)$, where $n(r,z,v)dv$ represents the number density of particles with volume between v and $v + dv$ and the mean vertical component of the fluid velocity be $u(r,z)$ (Fig. 1). The Smoluchowski equation governing the evolution of particulate matter in the plume may then be written in integral form as

$$\frac{\partial}{\partial z}\left\{\int_0^\infty n(r,z,v)u(r,z)rdr\right\} = \int_0^\infty K(r,z,v)rdr,$$

$$K(r,z,v) = \Gamma\left\{\int_0^v n(v - v')n(v')dv' - 2n(v)\int_0^\infty n(v')dv'\right\}. \quad [1]$$

In the definition of the K function, the first term corresponds to an increase in particles of size v resulting from collisions of two smaller particles, and the second term corresponds to a loss resulting from particles of size v colliding with particles of any other size. The spatial dependence of n is suppressed in the definition of $K(r,z,v)$ in the interest of clarity. In obtaining Eq. [1], it is assumed that the contribution of turbulent fluctuations to the mean vertical flux of particulate matter and to the coagulation rate is small compared with the contributions of the mean properties. Relative motion between gas and particles is ignored.

The coagulation frequency Γ has been taken as constant. The integral formulation, based on averages over horizontal cross sections through the plume, is forced on us by our inability to make a detailed description of the turbulent state in the plume. Instead, we adopt the Morton–Taylor–Turner (1–3) description which has proved quite successful in the study of the hydrodynamics of buoyant plumes. Specifically we assumed that $u(r,z)$ and $n(r,z,v)$ can be represented in the form

$$u(r,z) = U(z) \exp\{-(r/R(z))^2\},$$
$$n(r,z,v) = N(v,z) \exp\{-(r/R(z))^2\}. \quad [2]$$

When Eq. [2] is substituted into Eq. [1], the coagulation equation becomes

$$\frac{\partial}{\partial z}\{N(v,z)U(z)R^2(z)\} = \bar{K}(v,z),$$

$$\bar{K}(v,z) = \Gamma R^2(z)\left\{\int_0^v N(v-v')N(v')dv' - 2N(v)\int_0^\infty N(v')dv'\right\}. \quad [3]$$

Again, the dependence of N on z in the definition of $\bar{K}$ is understood.

The problem can be formally reduced to the equation which governs the study of homogeneous coagulation by the following transformation. Let $\phi(\tau,v)$ and $\tau(z)$ be defined by

$$\phi(\tau,v) = N(v,z)U(z)R^2(z),$$
$$\frac{d\tau}{dz} = \{R(z)U(z)\}^{-2}. \quad [4]$$

The coagulation equation now takes the form

$$\frac{\partial\phi}{\partial\tau} = \Gamma\left\{\int_0^v \phi(v-v')\phi(v')dv' - 2\phi(v)\int_0^\infty \phi(v')dv'\right\}. \quad [5]$$

Physically, the quantity $\pi\phi dv$ represents the mean flux of particulate matter in a size range dv about v through a cross section of the plume at a height z, while $(RU)^2$ is proportional to the vertical momentum flux at that cross section. The homogeneous form of the coagulation equation has been the subject of numerous investigations (4–7). It has been applied to size distributions of the type found experimentally in a smoke box by the authors (9) and shown to follow the data quite accurately. The transformation given in Eqs. [4] opens the possibility of using smoke box data in a buoyant plume. In order to proceed, however, it is necessary to determine $U(z)$ and $R(z)$ from a study of hydrodynamics of the particle-laden plume.

Equation [5] can be generalized to include the v dependence of Γ. For simplicity we take Γ to be a constant. In a study of smoke coagulation in a well-mixed chamber (9), good agreement was obtained with a constant Γ equation.

3. PLUME HYDRODYNAMICS

The Morton–Taylor–Turner model is based on an integral formulation of the conservation of mass, momentum, and energy in a fluid. Density variations $\delta\rho$ about a mean background level ρ are assumed to be a small fraction of ρ, so that only the weakly buoyant portion of the plume is considered. The pressure in the plume is taken to be hydrostatic, which is consistent with the horizontal components of the momentum conservation equation for a long narrow plume (i.e., one for which R/z is small). The mean temperature perturbation $\theta(r,z)$ from the ambient temperature $T(z)$ is small compared with $T(z)$. Moreover, the variation of $T(z)$ about a reference value T_0 (e.g., the value at $z = 0$) is also assumed to be small compared with T_0.

Under the above conditions, the conservation of momentum and energy in the plume may be expressed in the form

$$\frac{d}{dz}\int_0^\infty u^2 r dr = -g\int_0^\infty \frac{\delta\rho}{\rho} r dr,$$

$$\frac{d}{dz}\left\{c_p\int_0^\infty u\theta r dr\right\} + \left(c_p\frac{dT}{dz} + g\right)\int_0^\infty u r dr = 0. \quad [6]$$

Here g is the magnitude of the gravitational acceleration and c_p is the specific heat of the gas. Since the fluid is actually a gas–particle mixture, the specific heat should be that of the mixture. However, since the particulate mass fraction $x(r,z)$ is typically quite small, there is little loss in accuracy in this approximation. The fractional density difference $\delta\rho/\rho$ is given in terms of the temperature excess $\theta(r,z)$ and the particulate mass fraction x by the expression

$$\delta\rho/\rho = -\frac{\theta}{T_0} + x. \quad [7]$$

The first term expresses the decrease in density with increased temperature based on the ideal gas equation of state while the second term takes into account the increase in density resulting from the presence of the particulate. The mass fraction x satisfies a conservation of particulate mass equation obtained from the coagulation equation, Eq. [1], in the form

$$\frac{d}{dz}\left(\int_0^\infty u x r dr\right) = 0. \quad [8]$$

Finally, effective plume radius $R(z)$ is obtained from consideration of the mass entrained into the plume. Let $\Psi(r,z)$ be the stream function, defined so that $2\pi\rho\Psi$ is the mean mass flux through a tube of radius r at height z in the plume. Mathematically Ψ is the vector potential satisfying the equation expressing conservation of mass in the plume,

$$\frac{\partial\Psi}{\partial r} = ru(r,z), \quad \frac{\partial\Psi}{\partial z} = -rw(r,z). \quad [9]$$

The quantity $w(r,z)$ is the mean radial component of velocity. The conservation of mass can be expressed in integral form as

$$\frac{d}{dz}\left(\int_0^\infty u r dr\right) = \frac{d\Psi}{dz}(\infty,z). \quad [10]$$

The entrainment hypothesis (1) states that $(d\Psi/dz)(\infty,z)$ may be determined from the expression

$$\frac{d\Psi(\infty,z)}{dz} = \alpha R(z)U(z). \quad [11]$$

Here, $U(z)$ and $R(z)$ are respectively the mean velocity and radius defined in Eq. [2], and α is an empirically determined entrainment constant.

The entrainment hypothesis, together with the Guassian profile assumption for all quantities of interest, enables us to obtain a closed system of equations for the mean plume variables. Specifically, we define a mean temperature $\Theta(z)$ and a particulate concentration $X(z)$ as follows:

$$\theta(r,z) = \Theta(z)\exp\{-(r/R(z))^2\},$$
$$x(r,z) = X(z)\exp\{-(r/R(z))^2\}. \quad [12]$$

(Note that the form for $x(r,z)$ follows from that for $n(r,z,v)$ in Eq. [2] since x is proportional to the first moment of n with respect to the size variable v.) Substitution of forms [2] and [12] into Eqs. [6], [8], and [10], together with the elimination of $(d\psi/dz)(\infty,z)$ and $\delta\rho/\rho$ through the use of Eqs. [7] and [11], leads to the desired set of equations

$$\frac{d}{dz}(R^2U) = 2\alpha RU,$$

$$\frac{d}{dz}(R^2UX) = 0,$$

$$\frac{d}{dz}(R^2U^2) = 2gR^2\left(\frac{\Theta}{T_0} - X\right),$$

$$\frac{d}{dz}(R^2U\Theta) = -(2g/c_p)(1 - \Delta)UR^2. \quad [13]$$

These equations, which are a slight generalization of those used by Morton *et al.*, represent conservation of fluid mass, particulate mass, vertical momentum, and energy, respectively. The quantity Δ appearing in the last of Eqs. [13] is defined as

$$\Delta = -(c_p/g)\frac{dT}{dz}. \quad [14]$$

In the atmosphere Δ is the lapse rate; in a fire situation Δ is a measure of the room stratification induced by the fire. For an adiabatic atmosphere, $\Delta = 1$ and the flux of excess mean energy in the plume ($R^2U\Theta$) is constant. This possibility arises because in the absence of any molecular heat transport dissipative process, or distributed heat sources and sinks (as opposed to the source at $z = 0$), the plume is adiabatic. If the surroundings are also adiabatic, there can be no net energy exchange and the constancy of the energy flux follows. The lapse rate will be treated as a prescribed constant, consistent with the assumption that there are no large variations in ambient properties. The entrainment constant α will be taken as 0.07, the value recommended by Turner (3).

Equations [3] and [13], or equivalently, Eqs. [4], [5], and [13], must be supplemented by initial conditions which describe the nature of the source at $z = 0$ which generates the plume. Let m_0 be the mass flux released by the source, of which x_0m_0 is particulate. It is further assumed that the size distribution of the particle flux is given as $\pi\phi_0(v)$. If Q_0 and p_0 are respectively the flux of heat and momentum released by the source, then the following conditions hold at $z = 0$.

$$\phi = \phi_0(v),$$

$$\rho\pi R^2U = m_0,$$

$$X = 2x_0,$$

$$c_pm_0\Theta = 2Q_0,$$

$$\pi\rho U^2R^2 = 2p_0. \quad [15]$$

We turn in the next section to the solution of the plume equation [13], subject to initial conditions given in the last four of Eqs. [15]. With these solutions in hand, we are then in a position to return to the main problem in the following section and complete the study of the coagulation process itself.

4. THE PLUME SOLUTION

It is convenient to introduce nondimensional variables by scaling U, Θ, R, and X by the initial conditions. We choose the following normalization.

$$z = z_0t,$$

$$R = \alpha z_0\eta(t),$$

$$\Theta = (2Q_0/c_pm_0)\bar{\Theta}(t),$$

$$U = (4gQ_0z_0/m_0c_pT_0)^{1/2}\bar{U}(t),$$

$$z_0 = (m_0/2\rho\pi\alpha^2)^{2/5}(m_0c_pT_0/gQ_0)^{1/5}. \quad [16]$$

The choice of z_0 will become clear below. We also introduce new dependent variables ξ, M, P^2, and H, which are respectively proportional to the particulate mass, fluid mass, momentum, and energy flux in the plume. They are defined as

$$X = 2x_0\xi,$$

$$\bar{U}\eta^2 = M,$$

$$\bar{U}\eta = P,$$

$$\bar{U}\eta^2\bar{\Theta} = H. \quad [17]$$

In terms of these variables, the plume equations (Eqs. [13]) take the form

$$\frac{dM}{dt} = 2P,$$

$$M\xi = 1,$$

$$P^2 d\,\frac{(P^2)}{dt} = M(H - \chi),$$

$$\frac{dH}{dt} = -\nu^2 M. \qquad [18]$$

The equations contain two dimensionless parameters, χ and ν^2, defined by

$$\chi = (x_0 c_p m_0 T_0/Q_0),$$

$$\nu^2 = (z_0 m_0 g/Q_0)(1 - \Delta). \qquad [19]$$

The initial conditions, Eqs. [15], may be rewritten in terms of the new variables as

$$\xi(0) = M(0) = H(0) = 1,$$

$$P(0) = (m_0 c_p T_0 p_0/2\pi\alpha^2 \rho g Q_0 z_0^3)^{1/2} \equiv P_0. \quad [20]$$

Thus, the problem is reduced for it is seen to depend upon the three dimensionless parameters χ, ν^2, and P_0. Physically, χ measures the relative effectiveness of particulate loading and the heat flux on the plume buoyancy. The quantity χ must be less than unity if a buoyant plume (as opposed to a forced jet) is to exist at all. The quantity ν^2 measures the importance of ambient stratification over the length scale z_0; while P_0 measures the ratio of initial to induced plume momentum. The scale z_0 is chosen by the requirement that the normalized particle mass fraction ξ vary through the range $0 \le \xi \le 1$.

The solution procedure is essentially that due to Morton (2). We introduce a new buoyancy variable W, defined by

$$W(t) = H - \chi,$$

$$W(0) = 1 - \chi. \qquad [21]$$

Two integrals can be obtained by eliminating t between the first three of Eqs. [18]. They are

$$P^4 + W^2/\nu^2 = ((1 - \chi)/\nu)^2 + P_0^4 \equiv K_1^4,$$

$$M^2 = 1 + (4K_1^3/\nu)\{f((1 - \chi)/K_1^2\nu) - f(W/K_1^2\nu)\},$$

$$f(y) = \int_0^y (1 - s^2)^{1/4} ds. \qquad [22]$$

The dimensionless vertical position t can then be obtained from

$$(\nu/K_1^2)t = \int_{W/K_1^2\nu}^{(1-\chi)/K_1^2\nu} \{1 + (4K_1^3/\nu) \times [f((1 - \chi)/K_1^2\nu) - f(y)]\}^{-1/2} dy. \quad [23]$$

The solution thus consists of Eqs. [22] and [23], and the second of Eqs. [18]. It is expressed parametrically in terms of W. The source is at $W = 1 - \chi$. The buoyancy force W decreases as the vertical height increases. At $W = 0$, the buoyant force becomes negative, and steadily retards the plume until the value $W/K_1^2\nu = -1$ is reached. At this point, the vertical momentum P^2 vanishes. Thus, the maximum plume height $t_{\max}$ is given by

$$t_{\max} = (K_1^2/\nu) \int_{-1}^{(1-\chi)/K_1^2\nu} \{1 + (4K_1^3/\nu \times [f((1 - \chi)/K_1^2\nu) - f(y)]\}^{-1/2} dy. \quad [24]$$

Above this height, the fluid spills out of the plume into the surrounding atmosphere and the plume ceases to exist as an organized structure. With these results in hand, we now return to complete the solution of the coagulation equation.

5. THE SOLUTION OF THE COAGULATION EQUATION

We wish to solve Eq. [5] for the particle flux distribution $\phi(v,z)$ subject to the initial condition $\phi = \phi_0(v)$ given in the first of Eqs. [15]. The particle flux distribution plays the same role as the number distribution does in homogeneous coagulation. Similarly, the total number flux $\Phi(\tau)$ plays the role of the homogeneous number density. The total number flux is defined by the relation

$$\Phi(\tau) = \int_0^\infty \phi(\tau,v)dv. \qquad [25]$$

To proceed, it is convenient to introduce the first two moments, Φ_0 and Φ_1, of the initial distribution

$$\Phi_0 = \int_0^\infty \phi_0(v)dv,$$

$$\Phi_1 = \int_0^\infty v\phi_0(v)dv. \quad [26]$$

Integrating Eq. [5] over v, one readily obtains the solution for the total number flux $\Phi(z)$ as

$$\Phi = \Phi_0/(1 + \Phi_0\Gamma\tau). \quad [27]$$

Using Eq. [27], we can transform Eq. [5] into the reduced form used in our previous study (9) of homogeneous coagulation. Note that the ratio Φ_1/Φ_0 defines a natural volume size scale. Thus, we define new dependent and independent variables as follows:

$$\phi = (\Phi^2/\Phi_1)\psi(\bar{v},\lambda),$$

$$\bar{v} = v(\Phi_0/\Phi_1),$$

$$\Phi(\tau)/\Phi_0 = 1 - \lambda. \quad [28]$$

This transformation of variables is essentially the same as the similarity transformation introduced by Swift and Friedlander (10) and Wang and Friedlander (11).

Equation [5] then becomes

$$\frac{\partial\psi(\bar{v},\lambda)}{\partial\lambda} = \int_0^{\bar{v}} \psi(\bar{v}',\lambda)\psi(\bar{v} - \bar{v}')d\bar{v}'. \quad [29]$$

Equation [29] is now identical to Eq. [12] of Ref. (9), with Eqs. [28] replacing Eqs. [13]–[15] of that work as the definitions of ψ, λ, and $\bar{v}$. The initial conditions may also be written in the form (compare Eqs. [17] and [18] of (9))

$$\psi(\bar{v},0) = \psi_0(\bar{v}),$$

$$\int_0^\infty \psi_0(\bar{v})d\bar{v} = 1,$$

$$\int_0^\infty \bar{v}\psi_0(\bar{v})d\bar{v} = 1. \quad [30]$$

Finally, the solution can still be obtained by Laplace transform techniques as

$$\psi(\lambda,\bar{v}) = \int_{\alpha-i\infty}^{\alpha+i\infty} e^{p\bar{v}} \frac{\bar{\psi}_0(p)}{1 - \lambda\bar{\psi}_0(p)}dp. \quad [31]$$

Here, as in Eq. [16] of Ref. (9), $\bar{\psi}_0(p)$ is the Laplace transform of the initial condition.

At this point, however, the analysis diverges sharply from that appropriate to homogeneous coagulation. This follows from the relation between τ (or λ) and the dimensionless vertical position in the plume t. Using the definition of τ in Eq. [4] together with the normalization introduced in Eqs. [16] and the plume solutions given in Eqs. [22] and [23], one can write τ in the form

$$\tau = \tau_0 \int_{W/K_1^2\nu}^{(1-\chi)/K_1^2\nu} \{(1 - y^2)[f((1 - \chi)/K_1^2\nu) - f(y) + \nu/4K_1^3]\}^{-1/2}dy,$$

$$\tau_0 = \frac{m_0 c_p T_0}{4\alpha^2 g z_0^2 Q_0}\frac{1}{\nu}\left(\frac{\nu}{4K_1^3}\right)^{1/2}. \quad [32]$$

Equations [32] and [23] parametrically define the relation between τ and t. Since the integral appearing in Eq. [32] is bounded as $W/K_1^2\nu \to -1$, τ has a finite maximum value $\tau_{\max}$, given by Eq. [32] with $W/K_1^2\nu = -1$. Physically, this means that the coagulation process "freezes," in the sense that the equivalent homogeneous process abruptly stops at a fixed point in time. We now consider this phenomenon in detail.

6. RESULTS

The problem has now been reduced to the evaluation of two dimensionless integrals, one corresponding to a reduced height, h_r (Eq. [23]), and the other to a reduced time, $\bar{\tau}$ (Eq. [32]):

$$h_r = 2t(\nu/K_1)^{1/2},$$

$$\bar{\tau} = \tau/\tau_0. \quad [33]$$

The quantities $\bar{\tau}$ and h_r are parametrically related through their dependence on the quantity $W/K_1^2\nu$. Some care must be taken in performing the integration in Eq. [32] because of the singularity of the integrand at $y = \pm 1$ and because of the smallness of

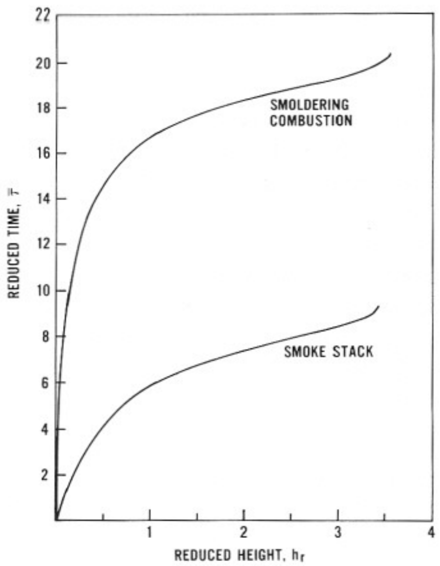

FIG. 2. The relationship between coagulation time and height in the plume for a smokestack-type plume ($\nu/4K_1^3 = 4.32 \times 10^{-5}$ and $(1 - \chi)/K_1^2\nu = 0.99765$) and for a smolder-generated plume ($\nu/4K_1^3 = 3.03 \times 10^{-6}$ and $(1 - \chi)/K_1^2\nu = 1.0$).

the quantity $\nu/4K_1^3$. The method used is outlined in the Appendix.

The analysis developed above is now applied to a smokestack plume and a plume from a smoldering punk stick. The effluent of a 1-m-radius smokestack is assumed to be at 400°K with a flow velocity of 15 m/sec. The fraction of fuel converted to particulate, x_0, is taken to be 2×10^{-3}. These values are estimates for a local coal power plant. Also, a value of -6.5°C/km is assumed for the temperature gradient in the atmosphere with a base temperature of 300°K.

The punk (imported from the Orient as incense sticks) consists of a bamboo stick core with a square or rectangular cross section, 1 to 2 mm in width. A finely ground mixture of cellulosic material, binder, and oxidant coated the stick to a final diameter of about 3 mm. The mass flux was found to be 3.5×10^{-4} g/sec with about 10% of the fuel appearing as particulate. A heat flux of 1.3×10^6 ergs/sec was obtained on the basis of an estimated surface temperature of 800°C. The surface gas velocity is assumed to be zero and the temperature gradient in the enclosure is taken to be 10°C/m.

The relationship between $\bar{\tau}$ and h_r is shown in Fig. 2 for both the smokestack plume and a smoldering smoke-source plume. The curves terminate at the point where the velocity of the rising plume goes to zero. One unit of the reduced height h_r corresponds to about 100 m for the smokestack and to about 8 cm for the smoldering source.

An important result of our study is the calculation of the dependence of the particle number flux on height. It is convenient to display the result in terms of the reduced number flux and the dimensionless plume parameter A.

$$\Phi/\Phi_0 = 1/(1 + A\bar{\tau}), \qquad [34]$$

where

$$A = \Phi_0 \Gamma \tau_0. \qquad [35]$$

From the plots of $\bar{\tau}$ versus h_r in Fig. 2, one can readily obtain the curves for the reduced number flux as a function of the reduced height. The curves in Fig. 3 correspond to the smokestack plume while those in Fig. 4 correspond to the smoldering source. It is seen in both figures that for values of A greater than 0.1 the particle number flux is significantly affected by coagulation.

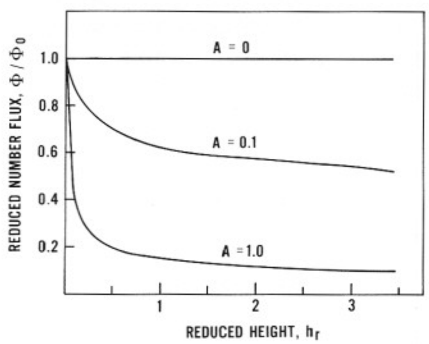

FIG. 3. The particle number flux is plotted versus height for various values of the plume parameter A. The parameters $(1 - \chi)/K_1^2\nu$ (=0.99765) and $\nu/4K_1^3$ (=4.32×10^{-5}) were chosen to be representative of a smokestack-type plume.

The shape of the aging curves (Figs. 3 and 4) is controlled by the time versus height curves (Fig. 2). The heavily laden, slowly rising smoldering plume produces a very fast initial growth in $\bar{\tau}$. This is responsible for the rapid decay in particle flux shown in Fig. 4. The smokestack plume, on the other hand, behaves initially like a forced momentum jet and is very lightly loaded. The coagulation is both slower and more uniformly spread over the vertical extent of the plume than is the case in the smoldering plume. In both cases, there is a noticeable tendency for the coagulation process to freeze due to the dilution produced by the entrainment of particulate-free air. The small apparent increase in coagulation implied by the decrease in Φ/Φ_0 at the top of the plume (see Fig. 3) is an artifact of the model. It is caused by the large increase in time $\bar{\tau}$ induced by the vanishing of the vertical velocity at this point. In fact, the model breaks down here, predicting an infinitely wide plume at the final height.

The estimation of the plume parameter A can be useful for determining whether coagulation will have a significant effect in a given situation. Assuming Q is given by $m_0 c_p(T - T_0)$, one finds

$$\tau_0 = \left(\frac{\nu}{4K_1^3}\right)^{1/2} \frac{T_0}{4\alpha^2 g z_0^2 (T - T_0)\nu}. \quad [36]$$

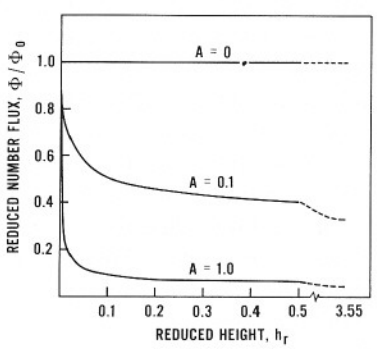

FIG. 4. The particle number flux is plotted versus height for various values of the plume parameter A. The parameters $(1 - \chi)/K_1^2\nu$ (=1.0) and $\nu/4K_1^3$ (=3.03 $\times 10^{-6}$) were chosen to be representative of a smoldering source in an enclosure.

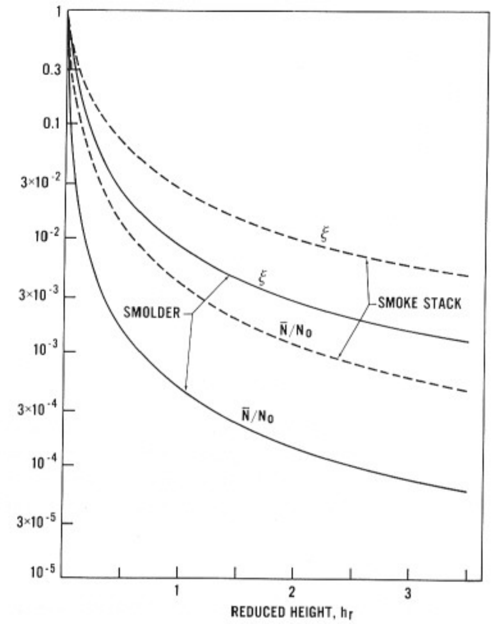

FIG. 5. The particle mass concentration ξ and reduced number concentration $\bar{N}/N_0$ are plotted versus height for a smokestack plume and a smoldering source plume. The plume constant A is 1.0 for these plots.

Substituting the appropriate values for a smokestack plume, one obtains $\tau_0 \sim 10^{-8}$ sec^2/cm. Assuming a coagulation frequency Γ of 10^{-9} cm^3/sec, one finds that Φ_0 must be greater than about 10^{16} particles/sec for coagulation to be significant ($A > 0.1$). This flux corresponds to a number concentration on the order of 10^8 particles/cm^3. The corresponding result for the smoldering case is a number flux $\sim 5 \times 10^9$ particles/sec and a number concentration $\sim 10^{10}$ particles/cm^3.

Once the parameter A has been determined, the timelike variable λ defined in Eq. [28] is known. Thus, any moment of the flux distribution function can be determined in terms of its initial value. These quantities, together with the reduced fluid mass flux M, determine the moments of the particle distribution function $N(v,z)$ (see Eq. [4]). The first two moments are shown plotted in Fig. 5 for the two cases considered when $A = 1.0$. The ratio of the total par-

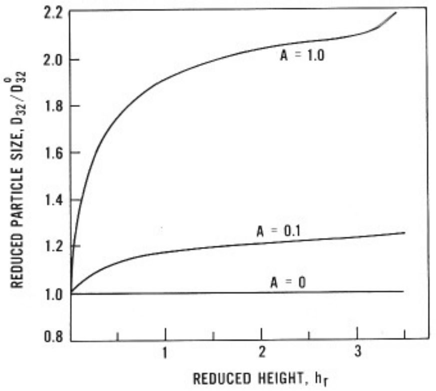

FIG. 6. The particle size is plotted versus height for various values of the plume parameter A. The parameters $(1 - \chi)/K_1^2\nu$ $(=0.99765)$ and $\nu/4K_1^3$ $(=4.32 = 10^{-5})$ were chosen to be representative of a smokestack.

ticle number density to its initial value, denoted by $\bar{N}/N_0$ in the figure, is determined from the relation

$$\bar{N}M/N_0 = \Phi/\Phi_0.$$

The reduced particle mass fraction ξ is obtained from the second of Eqs. [18]. The mass fraction, being a conserved quantity, is determined solely by the hydrodynamics. Thus, the difference between the ξ curves is solely due to the different mass entrainment rates caused by differences in ambient stratification and initial momentum. The additional relative discrepancy between the number density curves may be attributed to the differing coagulation rates described above. In the absence of coagulation, the plots of ξ and $\bar{N}/N_0$ would be identical.

A final application concerns the mean particle size. The decrease in number flux with increased height is accompanied by an increase in particle size as the particles coagulate. Because of recent advances in optical sizing techniques (12), it is perhaps easiest to follow the coagulation process by measuring the volume surface mean particle diameter D_{32} as a function of plume height. As an aid to such an experiment, it is shown below how the quantity D_{32} can be determined from the general analysis given in Sections 2 and 5.

The quantity D_{32} is defined by

$$D_{32} = \frac{\int_0^\infty D^3 n_D dD}{\int_0^\infty D^2 n_D dD}, \qquad [37]$$

where $n_D dD$ is the number concentration in the particle diameter range D to $D + dD$. The quantity D_{32} when expressed in terms of the flux variables assumes the form

$$D_{32} = (6/\pi)^{1/3}(\Phi_1/\Phi)^{1/3}\left\{\int_0^\infty \psi\eta^{2/3}d\eta\right\}^{-1}, \qquad [38]$$

where η $(=V\Phi/\Phi_1)$ is closely related to $\bar{v}$ in Eq. [28]. Our previous study (Ref. (9)) showed that ψ to a good approximation depended on only the variable η for smoke aerosols generated both by flaming combustion and by smoldering combustion. Assuming this to be strictly true, then the only term in Eq. [38] depending on plume height is the number flux Φ. Thus D_{32} can be expressed as a function of the initial values of D_{32} and Φ and as a function of the number flux, $\Phi(z)$,

$$D_{32} = D_{32}^{\circ}[\Phi_0/\Phi(z)]^{1/3}. \qquad [39]$$

Once the particle number flux is calculated as a function of plume height from Eqs. [33] and [34], the dependence of particle size on height is readily obtained using Eq. [39]. As an example, particle size is plotted as a function of height in Fig. 6 for the smokestack example. It is seen from Figs. 4 and 6 for the case $A = 1$ that as the particle size has increased by about a factor of two, the number flux has dropped by about a factor of nine. This is expected based on the cube root dependence between particle size and number flux in Eq. [39].

APPENDIX

The method used for performing the integration in Eq. [32] is outlined below. In order to simplify notation, Eq. [32] is expressed as

$$\bar{\tau} = \int_{\zeta}^{\alpha} \{(1 - y^2) \times [f(\alpha) - f(y) + \beta]\}^{-1/2} dy, \quad \text{[A1]}$$

where

$$\alpha = (1 - \chi)/K_1^2\nu,$$
$$\beta = \nu/4K_1^3,$$
$$\zeta = W/K_1^2\nu.$$

The function $f(y)$ can be expressed in one of the following forms to a precision of better than 1%:

$$\begin{aligned} f(y) &= C/6 - 2^{1/4}[4/5 - 1/18(1 - y)](1 - y)^{5/4}, && 0.7 < y < 1, \\ f(y) &= y - y^3/12 - 3y^5/160, && -0.7 \le y \le 0.7, \\ f(y) &= -C/6 + 2^{1/4}[4/5 - 1/18(1 + y)](1 + y)^{5/4}, && -1 \le y \le -0.7, \\ C &= \Gamma(1/4)\Gamma(1/2)/\Gamma(3/4). && \text{[A2]} \end{aligned}$$

Due to the smallness of the parameter β when the running variable ζ is near one, the integral must be rescaled to obtain reasonable numerical accuracy. The integral can be simplified under these circumstances to

$$\bar{\tau} = \lambda^{-1/10}(5/2)^{2/5} \int_{\theta_1}^{\theta} (1 + u^{5/2})^{-1/2} du, \quad \text{[A3]}$$

where

$$\theta = [1 - \zeta]^{1/2} \frac{5\lambda}{4(2)^{1/4}},$$

$$\lambda = \beta + f(\alpha) - f(1) = \beta - 2^{1/4}\,{}^4\!/_5(1 - \alpha)^{5/4}.$$

The above expression is valid for $\lambda > 0$. For $\lambda < 0$ the corresponding result is given by

$$\bar{\tau} = (-\lambda)^{-1/10}(5/2)^{2/5}(4/5) \int_{\delta_1}^{\delta} (1 + w^2)^{-3/5} dw,$$

where

$$\delta = \left[(1 - \zeta)^{5/4}\left(\left(\frac{5(-\lambda)}{4 \cdot 2^{1/4}}\right)^{-1} - 1\right)\right]^{1/2}. \quad \text{[A4]}$$

In the range $-0.95 < z < \alpha - 0.05$, the integration in Eq. [A1] is performed by straightforward application of the trapezoidal rule. For $\zeta \to -1$, it is again necessary to rescale because of the integrable singularity which results from the ambient stratification.

$$\bar{\tau} = (2)^{1/2} \int_{u}^{u_0} [f(\alpha) + f(1) + \beta - 2^{1/4}(4/5)u^{5/2}]^{-1/2} du,$$

$$u = (1 + \zeta)^{1/2}. \quad \text{[A5]}$$

The integral in Eq. [23] is evaluated by a similar method. Again the change of variables identified in case 1 or case 2 is used. The trapezoidal rule integration may be applied to $\zeta = -1$ because there is no singularity in the integrand of Eq. [23] as $\zeta \to -1$.

ACKNOWLEDGMENT

The authors wish to thank Professor Sheldon Friedlander for a stimulating conversation that led to this study.

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