

MODELLING DRY DEPOSITION

National Power¹ and CERC²

CONTENTS

1. Introduction	2
2. Modelling Dry Deposition	3
3. Deposition Velocity – Diffusive Part	6
3.1 Aerodynamic Resistance	8
3.2 Sub-layer Resistance	8
3.3 Surface Resistance.....	9
4. Deposition Velocity – Gravitational Settling	11
4.1 Modelling of Specific Effects.....	11
4.1.1 Particle Fall-Out During Plume Rise.....	11
4.1.2 Descending Plume Model	11
4.1.3 Plume Spread.....	12
4.1.4 Enhanced Dry Deposition.....	12
4.2 Calculating the Gravitational Settling velocity	13
5. Nomenclature	14
6. References	16

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1. Introduction

ADMS models three plume depletion processes: dry deposition, wet deposition and radioactive decay. This paper describes the dry deposition module. All three processes lead to a variation with downwind distance, or travel time, of the plume strength Q :

$$Q(x) = \int_{-\infty}^{\infty} dy \int_0^{\infty} C(x, y, z) U dz \quad (1.1)$$

where U is the mean wind speed and C is the concentration. In addition, dry deposition may alter the *shape* of the vertical concentration profile because removal of source material by dry deposition occurs only at the surface.

It is assumed that:

- (i) Dry deposition may be modelled by deposition velocity
- (ii) Removal processes act independently;
- (iii) Removal processes are irreversible

The first assumption implies that all the physical and chemical processes involved in the removal of pollutant from the plume by dry deposition are adequately represented by simple proportionality between removal rates and the near-surface concentration of local airborne material. The second assumption implies that plume strength can be written as a product of source strength and depletion factors corresponding to each removal process. Where species with significantly different deposition parameters are present in the release a separate calculation must be performed for each. Variation of dry deposition parameters with the formation of new isotopes following radioactive decay cannot, however, be modelled and the values of dry deposition velocity should reflect the contents of the initial isotope inventory. Assumption (iii) precludes re-suspension of small particles.

Removal processes are integrated into the dispersion model as follows. Let $C_1(x,y,z)$ be the mean concentration *per unit source strength* in the absence of deposition and assume that it can be factorised as:

$$C_1 = \chi_1(x, z) C_y(y) \quad (1.2)$$

in terms of the crosswind-integrated concentration χ_1 and normalised crosswind profile $C_y(y)$. Let Q_s be the source strength of the species under consideration in [mass units] s^{-1} .

Given the ground level concentration, the Dry Deposition Module calculates a depletion factor $q_d(x)$ and crosswind-integrated dry deposition flux $f_d(x)$.

At any downwind distance the plume strength in [mass units] s^{-1} is then given by:

$$Q = q_d(x) Q_s \quad (1.3)$$

The mean concentration in [mass units] m^{-3} is given by:

$$C = Q(x) \chi'_1(x, z) C_y(y) \quad (1.4)$$

and the dry deposition flux at a point in [mass units] $\text{m}^{-2} \text{s}^{-1}$ is:

$$F_{dry} = Q(x) f_d(x) C_y(y) \quad (1.5)$$

The dry deposition flux is calculated using a deposition velocity. Default values of the deposition velocity are supplied, or the user may specify parameters that allow the system to estimate the deposition velocity: nature of pollutant, surface roughness, particle diameter and particle density. The algorithms for calculating deposition velocity are given in sections 3 and 4.

2. Dry Deposition

Dry deposition is modelled through a single parameter v_d - the deposition velocity, which includes a diffusive part, v'_d , often referred to simply as dry deposition, and a gravitational settling or terminal velocity part, v_t . Removal of material at the surface leads to plume depletion with distance and to a modified vertical distribution of concentration.

In modelling dry deposition using the parameter v_d it is assumed that the rate of deposition per unit area is proportional to the near-surface concentration:

$$F_{dry} = v_d C(x, y, 0) \quad (2.1)$$

v_d is a function of pollutant species, the nature of the surface and the wind speed. It may be input directly by the user or estimated by the system, see sections 3-4.

Deposition velocities are assumed to be constant throughout the domain, except when modelling complex terrain for a reactive or unreactive gaseous pollutant and the user has specified the deposition velocity as unknown. In this case the model calculates the deposition velocity from the gas type: reactive, non-reactive or inert and uses the local value of friction velocity, u^* , in the equations in §§3.1-3.2. In flat terrain, as the surface roughness is single-valued, u^* takes a constant value and, therefore, the dry deposition velocity is constant in value.

Gaseous pollutants are assumed to have zero gravitational settling velocity because of the negligible weight of the molecules. In general, for particular pollutants, gravitational settling as well as diffusion will make some contribution to the overall rate of deposition. Many authors have simply taken the deposition velocity to be the sum of a gaseous deposition velocity and the gravitational settling velocity v_t . A more consistent method of incorporating gravitational settling can be derived from diffusivity theory [1] which results in an expression for the overall deposition velocity v_d :

$$v_d = \frac{v_t}{1 - \exp(-v_t / v'_d)} \quad (2.2)$$

Clearly, in the limit as $v_t \rightarrow 0$ we have $v_d \sim v'_d$.

Removal of source material at the surface:

- (i) reduces the total amount of airborne material
- (ii) redistributes material within the plume

Thus, both plume strength and the shape of the vertical concentration profile are modified as shown in Figure 1.

Plume strength may be written as a product of the initial source strength Q_s (possibly depleted by rain i.e. wet deposition) and a depletion factor $q_d(x)$:

$$Q = Q_s q_d(x) \quad (2.3)$$

The concentration per unit plume strength $C_1' = \chi_1'(x,z)C_1(y)$ may be written as the product of the concentration in the absence of deposition, C_1 , and a shape factor $S(x,z)$:

$$C_1' = S(x,z)C_1(x,y,z) \quad (2.4)$$

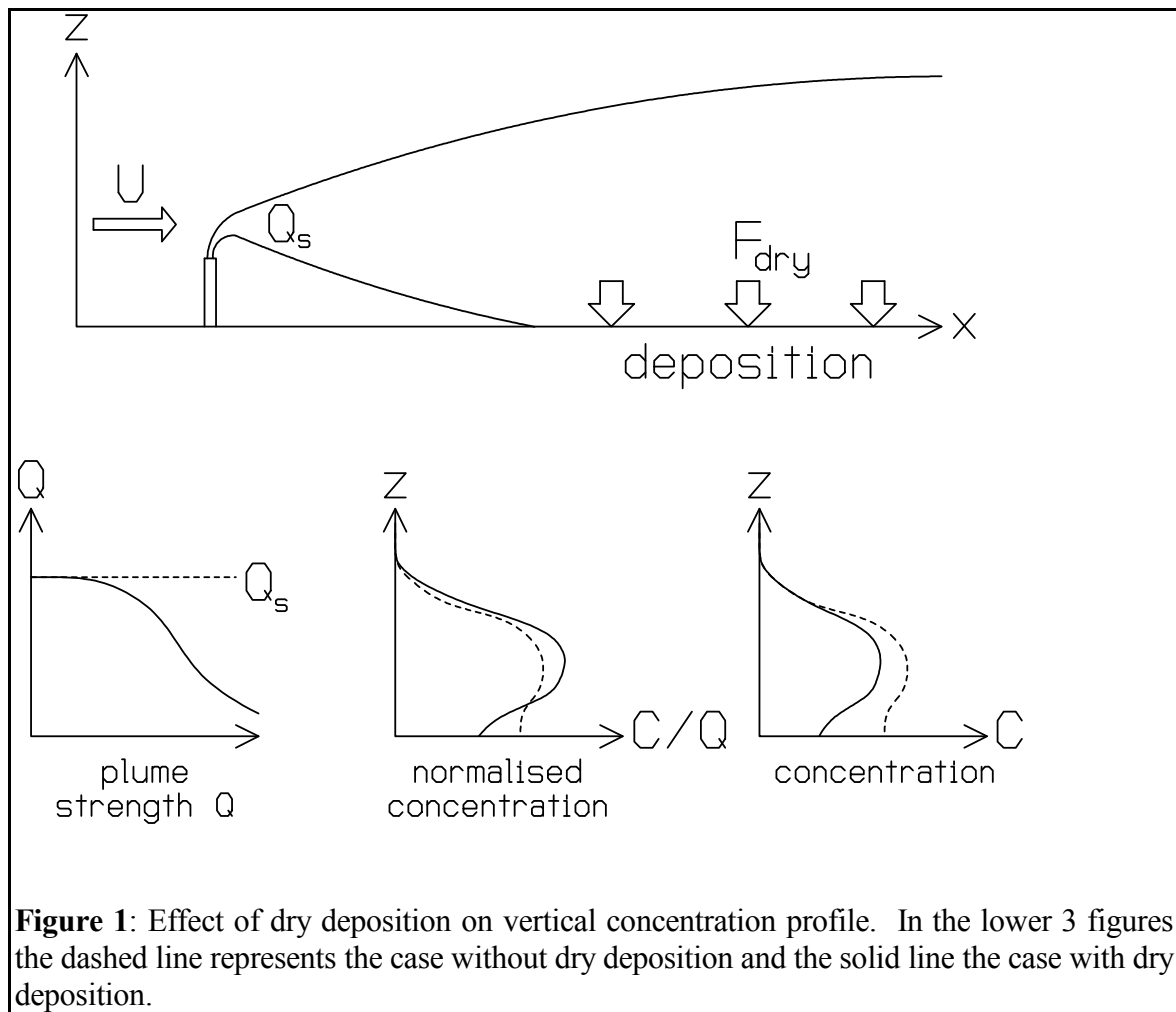


Figure 1: Effect of dry deposition on vertical concentration profile. In the lower 3 figures the dashed line represents the case without dry deposition and the solid line the case with dry deposition.

The lateral distribution of deposition is assumed to be the same as that of concentrations. Absolute concentrations are then given by:

$$C = Q(x) \chi_1'(x, z) C(y) \quad (2.5)$$

and the dry deposition flux at a point is given by equation (1.5), where $f_d = v_d \chi_1'(x, 0)$ is the crosswind-integrated dry deposition flux per unit plume strength.

The vertical concentration is required to satisfy the *surface flux condition*:

$$K \frac{\partial C}{\partial z} \sim v_d' C \quad (z \rightarrow 0+) \quad (2.6)$$

where K is the effective vertical eddy diffusivity and v_d' is the diffusive part of the deposition velocity (see section 3). For gases or particles with negligible settling velocity v_d' is the same as v_d . Since the boundary condition is linear in C and Q is independent of z , equation (2.6) becomes a condition on the shape factor S :

$$K \frac{\partial S}{\partial z} = v_d' S \quad \text{at } z = 0 \quad (2.7)$$

(since $\partial C_1 / \partial z = 0$ at $z=0$). Normalisation requires in addition that:

$$\int_0^\infty S \chi_1 U dz = 1 \quad (2.8)$$

Equations (2.7) and (2.8) do not determine S uniquely but a simple shape factor satisfying these conditions is:

$$S = \frac{1 + v_{ds} z / K}{1 + v_{ds} z / K} \quad (2.9)$$

where $\bar{z} = \int z C_1 dz / \int C_1 dz$ is the mean plume height (in the absence of deposition). K is given by

$$K = \sigma_w \Lambda_w (1 - \exp(-t / Tl)) \quad (2.10)$$

where t is travel time from the source and σ_w , Λ_w and Tl are the vertical turbulence, vertical turbulent lengthscale and turbulent timescale respectively, evaluated at \bar{z} . K is evaluated at $\min(\bar{z}, H/2)$. v_{ds} is given by

$$v_{dS} = \min \left(v_d, \frac{u_*}{\frac{\ln(z_d / z_0)}{\kappa} + 8} \right) \quad (2.11)$$

if the user has specified v_d , or $v_{dS} = v_d$ otherwise, where $z_d = \max(1, z_0)$.

The plume strength Q , is required to satisfy *conservation of material*:

$$\frac{dQ}{dx} \Big|_{dry} = - \int_{-\infty}^{\infty} F_{dry} dy \quad (2.12)$$

On substitution of (2.1), (2.3) and (2.5) produces:

$$\frac{dq_d}{dx} = -q_d f_d \quad f_d = v_d \chi'_1(x, 0) \quad (2.13)$$

to satisfy conservation of material.

3. Dry Deposition Velocity – Diffusive Part

The modelling of dry deposition in terms of a single dimensional quantity v_d tends to obscure the whole variety of physical and chemical processes whereby airborne material may be transported to and removed at a surface. The recommendations on deposition velocity of the Working Group on Atmospheric Dispersion [2] were addressed largely to the dispersion of radionuclides, notably molecular iodine. They gave only broad order of magnitude limits for the deposition velocity of selected groups: noble gases, reactive gases, $1\mu\text{m}$ and $10\mu\text{m}$ diameter particles, with no attempt to distinguish between different atmospheric and surface conditions. Despite considerable scatter in experimental data [3] there is general agreement on the major processes involved [4] and scope for a more rational approach to estimating the dry deposition velocity v_d .

The deposition velocity is *defined* by:

$$v_d = \frac{F(z_d)}{C(z_d)} \quad (3.1)$$

where F is the downward flux of material and C the local mean concentration. In the theoretical model we set $z_d=0$, which gives equation (2.1), although, in practice, z_d is some finite reference height, usually 1m. The diffusive deposition velocity v'_d may depend on the reference height z_d , the physico-chemical form of the pollutant, the characteristics of the surface and atmospheric turbulence. The reciprocal, $1/v'_d$ is termed the *resistance* r .

Physically, pollutant is transported to the vicinity of the surface by the turbulent eddies of the flow (possibly augmented by gravitational settling as described in section 4) and crosses the final sub-layer by one or more of a number of processes. The processes are determined by the nature of the surface (smooth, rough or water) and by the form of the pollutant (gas or particulate). For gases, permanent retention will then depend on reaction or absorption at the surface. Resuspension of small particles will be neglected. A resistance can be defined for each zone that the pollutant has to cross:

$$r(z_2, z_1) = \frac{C(z_2) - C(z_1)}{F} = \int_{z_1}^{z_2} \frac{dz}{K_z} \quad (3.2)$$

where K_z is a height-dependent diffusivity. Assuming that F , the dry deposition flux, is independent of height, (true on a sufficiently small vertical scale far enough from the source) then the *total* resistance is the sum of individual resistance terms:

$$r = r(z_d, z_n) + r(z_n, z_{n-1}) + \dots + r(z_2, z_1) \quad (3.3)$$

The resistance, r , in the absence of gravitational settling may be conveniently broken down into three resistances:

$$r = r_a + r_b + r_s \quad (3.4)$$

- (i) r_a is the ***aerodynamic resistance*** determined by the ability of the turbulent eddies to bring material close to the surface. Except in the case of very large particles it is independent of the physical form of the pollutant, but is determined by the structure of the turbulent flow.
- (ii) r_b is the ***sub-layer resistance***, the resistance to transfer across the final zone adjacent to the surface. Transport across this layer may occur by a variety of mechanisms, depending primarily on the roughness of the surface and the physical form of the pollutant. For smooth surfaces, r_b represents molecular and turbulent diffusion across the (laminar and transition) sublayers closest to the surface. It is necessary to admit a non-zero eddy diffusivity even in the laminar layer closest to the surface and the functional dependence of r_b on the Schmidt number $Sc = \nu/D$ depends on the form assumed for this.
- (iii) r_s is the ***surface resistance*** which is determined by the affinity of the surface for gaseous pollutants. For a perfectly absorbing surface $r_s = 0$. In general, however, a positive surface resistance is needed to account for non-zero surface concentrations of gases, particularly over vegetation. This is dependent on the dampness of the surface, the chemical form of the gas and, for vegetation, the degree of stomatal opening, which varies from day to night. This resistance term is very uncertain and experimental measurements vary by several orders of magnitude.

Since deposition velocity is the reciprocal of resistance, each individual resistance term sets an

upper bound on the rate of deposition. Thus, aerodynamic resistance alone determines an upper limit to the deposition velocity (although one which may be orders of magnitude too high).

Reviews of dry deposition modelling have been undertaken by Slinn [5], McMahon and Denison [6], Sehmel [7], Hosker and Lindberg [8], Underwood [4] and Nicholson [3]. The major qualitative conclusions that may be drawn from those studies are as follows:

For gaseous pollutants:

- (i) The limiting resistance terms are generally r_b and r_s .

For particulate pollutants:

- (ii) Surface resistance is usually negligible; particles adhere on contact, although bounce-off and re-suspension may be important in a few circumstances.
- (iii) There is usually a minimum deposition velocity in the size range 0.1 - 1 μm where neither Brownian diffusion (dominant sublayer transport mechanism for small particles) nor inertial impaction/gravitational settling (dominant sublayer transport mechanisms for large particles) are particularly effective.
- (iv) Deposition is often greater over water than over dry surfaces of similar roughness. This may result from particle growth through condensation.

Sections 3.1-3.3 describe how the resistance terms are modelled.

3.1 Aerodynamic resistance: r_a

The modelling of aerodynamic resistance is based on momentum transfer. $u_*^2/(U(z_d)-U(z_1))$ is the net downward velocity required to carry momentum from height z_d to height z_1 at a rate of u_*^2 . r_a is therefore given by:

$$r_a = \frac{U(z_d) - U(z_1)}{u_*^2} \quad (3.5)$$

where

$$z_d = \max(1 \text{ m}, z_0) \quad (3.6)$$

The same result is obtained by setting $K_z = u_*^2/(dU/dz)$ in equation (3.2). z_1 depends on whether the surface is rough or smooth and is defined below in §3.2.

3.2 Sub-Layer resistance: r_b

For *smooth* surfaces (defined by $u_* h' / \nu < 4$ where $h' = 30z_0$ is the equivalent sand roughness) surface protrusions are completely imbedded in the viscous sub-layer:

$$r_b = \frac{1}{\kappa u_*} \ln(1 + Sc) \quad Sc = \nu / D \quad (3.7)$$

$$z_1 = \nu / \kappa u_* \quad (3.8)$$

This is derived by taking $K_z = \kappa u_* z + D$ up to the height z_1 at which $v = \kappa u_* z_1$ in equation (3.2). For gases, D is the molecular diffusivity and we shall assume $Sc \sim 1$. For particles, D is the Brownian diffusion coefficient given by:

$$D = f kT / 3\pi\mu D_p \quad (3.9)$$

where k is Boltzmann's constant ($1.38 \times 10^{-23} \text{ J K}^{-1}$) and T is the thermodynamic temperature.

When the particle size approaches the molecular mean free path λ_m , the drag law must be modified since the no-slip boundary condition no longer holds. Hence the factor f is introduced into the molecular diffusivity in equation (3.9). The factor is defined by:

$$f = 1 + Kn \left(2.514 + 0.8 \exp \left(\frac{-0.55}{Kn} \right) \right) \quad (3.10)$$

and the Knudsen number Kn is given by

$$Kn = \frac{\lambda_m}{D_p} \quad (3.11)$$

For *rough* surfaces:

$$r_b = \frac{1}{Bu_*} \quad (3.12)$$

For gases we follow Underwood [4] and use:

$$\frac{1}{B} = 8 \quad (3.13)$$

$$z_1 = z_0 \quad (3.14)$$

whilst for particles we use Sehmel and Hodgson's 4th correlation ([12]) which can be written in the present notation as:

$$\begin{aligned} \frac{1}{B} = & \exp[-378.051 + 16.498 \ln(Sc) \\ & + \ln(\tau) \{ -11.818 - 0.2863 \ln(\tau) + 0.3226 \ln(D_p / z_0) - 0.3385 \ln(D / u_* z_0) \} \\ & - 12.804 \ln(D_p / D_{pref})]. \end{aligned} \quad (3.15)$$

Here τ is a non-dimensional inertial timescale given by

$$\tau = 10^{-8} v_{ss} u_*^2 / g\nu, \quad (3.16)$$

where v_{ss} is the Stokes Law settling velocity:

$$v_{ss} = \rho_{ref} g D_p^2 / 18\mu, \quad (3.17)$$

where ρ_{ref} is a reference particle density (1500kg/m^3) and D_{pref} is a reference diameter (0.01m).

If the resistance integral ($1/B$) is greater than the particles diffusion integral ($u \cdot z_1/D$, where $z_1 = 0.01\text{m}$), it should be replaced by the particle diffusion integral. In this case, equation (3.12) gives $r_b = z_1/D$.

3.3 Surface resistance: r_s

For particles, $r_s=0$, whereas for gases this is often the dominant term. Unfortunately, it is also the one for which there is most uncertainty. For vegetative surfaces, in particular, it may vary diurnally, seasonally and depending on the soil moisture conditions. The uptake of gases reflects the opening of stomata by day and closing by night and, for instance, the closing of stomata in times of drought. For this reason we choose to give only approximate values for broad categories of gases, see Table 1. The choice of resistance is based largely upon experimental data reviewed by McMahon and Denison [6] and Sehmel [7].

Type	Definition	Examples	r_s (s m^{-1})
REACTIVE GASES	Gases expected to undergo significant chemical reaction with the surface	SO_2 , O_3 , NO_x , I_2 , Cl_2 , HF	30
UNREACTIVE GASES	Gases not undergoing significant chemical reaction with the surface	CO_2 , CH_3I	1000
INERT GASES	Noble gases	He, Ne, Ar, Kr, Xe, Rn	∞

Table 1: Surface resistance for gases

4. Gravitational Settling

Gravitational settling causes the following effects all of which are modelled:

- fall-out of heavy particles during plume rise;
- global descent of particulate material during the subsequent passive dispersion stage;
- reduced turbulent spread as a consequence of particle inertia and motion relative to the flow;
- enhanced dry deposition;

Detailed accounts of the treatment of gravitational settling in atmospheric dispersion modelling can be found in references [1] and [9]. In ADMS the modelling of gravitational settling depends on a single parameter v_t , the settling velocity, which is used by the dry deposition module. The actual value of the settling velocity may be specified by the user or can be determined from diameter and density on the assumption of spherical particles. A procedure for evaluating the settling velocity (which reduces to the Stokes' Law range for small particles but is not restricted to such particles) is given in section 4.2.

In practice, releases need not be assumed monodisperse (i.e. of uniform settling velocity) and separate calculations can be performed for different size particles in the model. To avoid excessive computing time particle distributions should be broken down into ranges of settling velocity.

4.1 Modelling of Specific Effects

4.1.1 Particle Fall-Out During Plume Rise

Particles do not immediately settle out of a buoyant gaseous plume but are retained by re-entrainment at the base of the plume until the rise velocity falls below a threshold multiple of the settling velocity. Particles are retained within the plume provided:

$$w_p > cv_t \quad (4.1)$$

where w_p is the rise velocity of the gaseous plume. Foster [10] argues that value $c = 0.6$.

4.1.2 Descending Plume Model

During the passive dispersion stage the particulate component of the release descends relative to the mean streamlines of the flow:

$$\left. \frac{dz_p}{dt} \right|_{\text{settling}} = -v_t \quad (4.2)$$

where t is travel time. If this gravitational settling causes the plume to descend to ground level, it is assumed to travel along the ground from that point onwards. 'Image' plumes in the mean

concentration model are determined by direct reflection in the surface (ground) or inversion layer.

4.1.3 Plume Spread

Two effects reduce the spread of particles compared with gaseous pollutants [9].

1. **Trajectory-crossing effect** - each particle is continually falling out of the turbulent eddies which influence it, reducing the timescale over which eddy motion is correlated:

$$T_L^* = \frac{T_L}{\sqrt{1 + v_t^2 / \sigma_w^2}} \quad (4.3)$$

where T_L is the Lagrangian timescale of a fluid element and T_L^* that of the particle.

2. **Inertial or low-pass filter effect** - the inertia of the particles reduces their response to high frequency turbulent eddies:

$$\frac{\sigma_w^*}{\sigma_w} = \frac{\sigma_v^*}{\sigma_v} = \frac{1}{\sqrt{1 + T_p / T_L^*}} \quad (4.4)$$

where σ_v , σ_w are the crosswind and vertical root mean square (rms) velocity fluctuations in the ambient flow and σ_v^* , σ_w^* are those of the particle. $T_p = v_t/g$ is the particle response time.

If the vertical spread coefficient for gaseous constituents is given by $\sigma_z = \sigma_w tf(t/T_L)$ then the corresponding quantity for coarse particles is $\sigma_z^* = \sigma_w^* tf(t/T_L^*)$, with analogous result for σ_y^* .

4.1.4 Enhanced Dry Deposition

Gravitational settling enhances dry deposition through its effect on the deposition velocity v_d . This is explained in section 2 and shown in equation (2.2).

4.2 Calculating the Gravitational Settling Velocity

The terminal or settling velocity v_t in air of a particle of mass m satisfies:

$$\frac{1}{2} \rho_a v_t^2 A_p C_D = mg \quad (4.6)$$

where ρ_a is the density of the air and A_p is the horizontal projected area. C_D is the drag coefficient which, for any particular shape and orientation, is a function of the Reynolds number $Re = v_t D_p / \nu$. A practical approximation to Schlichting's [11] graph of C_D against Re for spherical particles is:

$$C_D = A Re^{-n} \quad (4.7)$$

where A and n are constants for decadal ranges of Reynolds number Re and are given in Table 2. The calculated values of v_t in Table 2 are based on a particle density of $1,000 \text{ kg/m}^3$ for illustrative purposes.

Re	A	n	For $\rho_p = 1,000 \text{ kg/m}^3$	
			D_p (μm)	v_t (m/s)
< 0.1	24	1	< 36	$3.07 \times 10^{-5} (D_p)^2$
0.1-1	28.5	0.925	36-83	$6.57 \times 10^{-5} (D_p)^{1.79}$
1-10	28.5	0.830	83-200	$1.77 \times 10^{-4} (D_p)^{1.56}$
10-100	16.4	0.591	200-600	$1.78 \times 10^{-3} (D_p)^{1.13}$
100-1000	6.54	0.391	600-2050	$9.67 \times 10^{-3} (D_p)^{0.86}$
> 1000	0.44	0	> 2050	$1.55 \times 10^{-1} (D_p)^{0.5}$

Table 2 Values of constants A and n , equation (4.7)

¹ D_p in μm .

The simultaneous solution of equation (4.6) and (4.7) yields the settling velocity v_t . For $Re < 0.1$ the relationships $C_D = 24/Re$, and $v_t = (\rho_p / \rho_a)(g/\nu)D_p^2/18$ are equivalent to Stoke's Law:

$$\text{Drag} = 3\pi\mu v_t D_p \quad (4.8)$$

As the particle size approaches the molecular mean free path λ_m (where $\lambda_m \approx 0.065$ in standard conditions), the drag law must be modified because the no-slip boundary condition no longer holds. The settling velocity v_t is multiplied by the factor f , that is given in equation (3.10). This factor becomes important for particles of diameter less than $1 \mu\text{m}$.

Note that the settling velocity can reach values on the order of 1 m/s . A range of observed values for

water droplets (i.e. particles of density 1000kg/m^3) can be found in Table B.1 of Mason [13].

5. Nomenclature

A	constant in power law dependence of Re on C_D
A_p	projected horizontal area of particle
B	factor in the sub-layer resistance term
C	concentration
C_D	drag coefficient
D	Brownian or molecular diffusion coefficient
D_p	particle diameter
f	slip correction factor
$F_{\text{dry}}, F_{\text{wet}}$	deposition fluxes at a point
f_d, f_w	crosswind-integrated dry and wet deposition fluxes
g	gravitational acceleration
Kn	Knudsen number
K	diffusion coefficient
n	exponent in power law dependence of Re on c_D
q_d, q_w	plume depletion factors for dry and wet deposition
Q	plume strength
Q_s	source strength
r	resistance
r_a, r_b, r_s	aerodynamic, sub-layer, surface resistances
Re	Reynolds number $=v_s d_p / \nu$
S	vertical profile shape factor from dry deposition
S^*	'stopping distance' $=u^* T_p$
Sc	Schmidt number $=\nu/D$
t	travel time
T	thermodynamic temperature
T_L	Lagrangian timescale
T_p	particle inertial timescale or response time $=v_t/g$
u^*	friction velocity
U	mean wind speed
v_{sr}	settling velocity at reference particle density
v_d	deposition velocity
v_d'	diffusive (non-settling) part of deposition velocity
V_t	gravitational settling velocity
v_{ss}	Stokes law settling velocity
w_p	plume rise velocity

x, y, z	Cartesian co-ordinate system: x along-wind, y crosswind; z vertical
z_1	height of the sub-layer
z_d	reference height
z_0	roughness length
z_p	plume height
χ	crosswind-integrated concentration
κ	von Karman's constant (=0.4)
μ, ν	dynamic, kinematic viscosities
ρ	density
ρ_{ref}	reference particle density (=1500kg/m ³)
ρ_v, ρ_w	root mean square (rms) horizontal and vertical velocity fluctuations
ρ_y, ρ_z	root mean square (rms) horizontal and vertical plume spread

Subscripts

a	approach flow
i	isotope index
s	source
l	per unit plume strength

Superscripts

*	effective flow quantity experienced by particle
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