PREDICTIONS FOR PARTICLE DEPOSITION ON NATURAL WATERS*

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Abstract—Deposition velocity predictions, for aerosol particles depositing on natural waters, are obtained by modifying earlier theory for deposition on smooth, solid surfaces. If particle growth by water vapor condensation occurs in the humid region adjacent to the air/water interface, then the deposition velocity for particles with (dry) radii near 1 μm is predicted to be essentially independent of particle size and equal to the limiting value set by atmospheric turbulence. That is, for these particles, \( \frac{d}{du} = C_D \) where (for open waters, constant wind, near neutral stability, and a 10 m reference height) the drag coefficient \( C_D \) = \((1.3 \pm 0.3) \times 10^{-2}\).

NOMENCLATURE

- \( a \) = particle radius
- \( a_w \) = geometric mean particle radius
- \( C_D \) = drag coefficient
- \( D \) = Brownian or molecular diffusivity
- \( D_f \) = dry deposition flux
- \( g \) = gravitational acceleration
- \( h \) = reference height
- \( k \) = transfer velocity without gravity
- \( \kappa \) = total transfer velocity
- \( m^* \) = mass flux of evaporated water
- \( S_c \) = \( \nu/D \) = Schmidt number
- \( S_t \) = \( tu^2/v \) = Stokes number based on the height of the viscous sublayer
- \( u_s \) = friction velocity
- \( u \) = mean wind speed
- \( u_i \) = deposition velocity
- \( v \) = gravitational settling velocity
- \( z \) = height
- \( \alpha \) = \( 10^{10} \text{ cm s}^{-1}/(\text{g cm}^{-2} \text{s}^{-1}) \), a constant
- \( \delta \) = height of the deposition layer
- \( \kappa \) = von Karman's constant
- \( v \) = kinematic viscosity of air
- \( \sigma \) = geometric standard deviation
- \( \tau \) = particle relaxation time
- \( \chi \) = trace constituent's concentration in air

Subscripts

- \( c \) = constant flux layer
- \( D \) = deposition layer
- \( d \) = dry
- \( \delta \) = evaluated at \( z = \delta \)
- \( h \) = evaluated at \( z = h \)
- \( i \) = evaluated at the air/water interface
- \( w \) = wet

Other symbols

- \( C \) = mass-average
- \( \text{mmr} \) = mass mean radius.

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INTRODUCTION

Improved estimates for particle dry deposition on natural waters are needed for the calculation of air pollution fluxes to lakes and oceans (e.g., Winchester and Nifong, 1971; Elder, 1975; Gatz, 1975; Whelpdale, 1976; Hicks, 1977; Prospero, 1978). The improvement investigated here is to attempt to account for particle growth by water vapor condensation. That some (and probably most, aged) aerosol particles grow in the neighborhood of water surfaces and that this growth would influence particle deposition rates have been discussed qualitatively by many authors. This growth was investigated quantitatively by Toba (1965) in his studies of sea-salt particle concentrations above the oceans.

In the present communication, a crude model to investigate the influence of particle growth on particle deposition will be used. For the model, the atmosphere beneath a convenient reference height (usually 10 m) is conceptually divided into two layers; see Fig. 1. In the lower layer, next to the interface, atmospheric turbulence is assumed to have negligible direct influence on particle transport (since fluctuations in the vertical component of the air's velocity must vanish at an impermeable boundary) and particles are assumed to possess their "wet radius", \( a_w \), the equilibrium size for the high value of the humidity to be specified. In the upper layer, the portion of the constant flux layer up to the reference height, atmospheric turbulence and gravitational settling of dry particles are assumed to govern particle transport. Clearly, this simple model provides opportunities for many improvements; here, the desire is to demonstrate that the model appears to contain sufficient realism to explain major features of available data.

ANALYSIS

Mathematically, the model is as follows. At the
Fig. 1. Schematic of two layer model and definition of some symbols; a more complete list of symbols can be found in the nomenclature.

The problem addressed now is the specification of $k_c$ and $k_D$. For transport across the deposition layer, it is proposed to modify earlier results for deposition to smooth, solid surfaces (Slinn, 1976). One proposed modification is to change the dependence of the flux on particle diffusivity from $D^2$ (e.g., see Appendix C of Slinn et al., 1978) to $D$. This change is suggested because the surfaces of natural waters slip; in contrast, for some reported water wind tunnel studies, it may be that the water surfaces did not slip because of short fetch. The slip for natural waters is in the direction of the mean wind and with a speed equal to a few percent of the 10-m wind speed (e.g., Krauss, 1972). Therefore, a characteristic air velocity in the air's viscous sublayer is approximately $u_*$ rather than $(z/\kappa)u_*$ (cf. Appendix C of Slinn et al., 1978). The second proposed change of the results for deposition to solid surfaces is, as already suggested, to use the equilibrium, wet particle radius, $a_w$, in all terms in $k_D$. For particles smaller than a few microns in radius, particle growth is sufficiently rapid to justify this equilibrium assumption (e.g., Toba, 1965; Ferron, 1977). For larger particles, it will be seen that the influence of particle growth is negligible because the transfer is rate-limited by transfer across the constant flux layer. Modifications to account for particle impaction on waves have been considered but, for most cases, the impaction parameter is too small for impaction to be important; exceptions may have occurred in the high-wind/small-wave wind tunnel studies reported by Sehmel and Sutter (1974). Also, modifications to account for particle capture by spray appears to be unnecessary, except at very high "spray rates"; this conclusion can be checked by analogy with scavenging by rain.

In summary, as a first approximation, we take

$$k_D = -\frac{\tau m^*}{\kappa} + \frac{1}{\kappa} C_D \rho_h [Sc^{-1.2} + 10^{-3.3}]$$

where $m^*$ represents the contribution from diffusophoresis (in which $x = 10^5$ cm s$^{-1}$(1 g cm$^{-2}$ s$^{-1}$) and $m^*$ is the rate of water evaporation); $Sc = v/D$ is the Schmidt number (in which $\nu$ is the air's kinematic viscosity and $D(a_w)$ is the particle's Brownian diffusivity in air); $St = \tau u_*^2/v$ is the Stokes number or impaction parameter (in which $u_*$ is the friction velocity and $\tau = u_*(a_w)/g$ is the particle's relaxation time, with $g$ the acceleration of gravity); $\kappa = 0.4$ is von Karman's constant; $u_*$ is the mean wind speed at the reference height; and $C_D = u_*^2/u_*$ is the drag coefficient (which, for open waters, steady wind, near neutral stability, and a 10 m reference height, Krauss (1972) gives $C_D = (1.3 \pm 0.3) \times 10^{-3}$; corrections for other than neutral stability are also given by Krauss). Finally, for transport by turbulence across the constant flux layer, we take

$$k_c = \frac{1}{(1+\kappa)} \frac{\tau u_*^2}{\kappa} = \frac{1}{(1+\kappa)} C_D \rho_h$$

and therefore $v_e = v_e(a_w)$, as desired.
Fig. 2. Plots of Equation (4), using Equations (5) and (6), for three wind speeds and three types of particles: dotted curves — hydrophobic (dry) particles; dashed curves — for particles that grow to the equilibrium size for \((\text{NH}_4)_2\text{SO}_4\) particles exposed to a relative humidity of 99%; solid curves — expected behavior of \((\text{NH}_4)_2\text{SO}_4\) particles for the case of deposition to lakes with relative humidity near 100% in the deposition layer. Note that for a range of particle radii near \(1 \mu m\), it is predicted that the dry transfer of hygroscopic particles can be rate limited by turbulent transfer through the constant flux layer; i.e., for these particles \(v_d = C \mu u_b\).

Note that, for the transport of momentum (with \(v_g = 0\) and \(D = v\)) then (4)–(6) give \(v_d = C \mu u_b\), as desired.

## RESULTS AND DISCUSSION

Figure 2 shows plots of (4) for three wind speeds, \(\mu = 0\), and for three cases of particle growth: (i) hydrophobic particles (i.e., \(\zeta = \zeta_0\)); (ii) particles that would grow to the equilibrium size of ammonium sulfate particles for a relative humidity of 99% in the deposition layer; and (iii) the expected behavior of ammonium sulfate particles (i.e., using the equilibrium size as given by Fitzgerald, 1975) for a relative humidity in the deposition layer of 100%.

These results suggest to the authors that for most, aged, atmospheric aerosol particles with mass mean radius (mmr) near \(1 \mu m\), it would be an acceptable approximation to assume that their deposition on natural waters is rate limited by turbulent transfer through the constant flux layer and to use

\[
\langle v_d \rangle \approx C \mu u_b = 1.3 \times 10^{-3} \langle \mu u_b \rangle
\]

(7)

This suggestion is consistent with recently reported data (Lodge, 1978; Delumyea and Petel, 1979; Sievering et al., 1979).

However, there are at least three factors that complicate data interpretation:

(i) inferences of dry deposition velocities from differences in pollutant air concentrations across water bodies are sensitive to small changes in the pollutant's vertical profile (or height of the atmosphere's mixed layer);

(ii) inferences of the mass-average deposition velocity are strongly influenced by the particle size distribution (e.g., for a log-normal distribution with geometric mean radius \(\alpha_g\) and standard deviation \(\sigma_g\), and for \(v_d(\alpha) = \alpha^2\) for Stokes' law then

\[
\langle v_d \rangle = \exp\left[\frac{8}{3} (\ln \sigma_g^3)\right] v_d(\alpha_g)
\]

\[
= \exp\left[\frac{2}{3} (\ln \sigma_g^3)\right] v_d(\text{mmr}),
\]

(8)

where \(\langle v_d \rangle\) is the mass-average deposition velocity; and

(iii) inferences of \(v_d\) from profiles of \(\chi(\alpha)\) can easily be misleading because of statistical, counting-errors when particle concentrations are low.

Nevertheless, there appear to be a number of independent hints that (7) is correct.

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## REFERENCES


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