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DRY DEPOSITION VELOCITIES

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DRY DEPOSITION VELOCITIES

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ABSTRACT

Dry deposition velocities are very difficult to predict accurately. In this article, reported values of dry deposition velocities are summarized. This summary includes values from the literature on field measurements of gas and particle dry deposition velocities, and the uncertainties inherent in extrapolating field results to predict dry deposition velocities are discussed. A new method is described for predicting dry deposition velocity using a least-squares correlation of surface mass transfer resistances evaluated in wind tunnel experiments.

INTRODUCTION

Data on dry deposition velocities are available from a variety of experimental techniques. Although several measurement techniques have been used to evaluate dry deposition velocities, the generalization of measured dry deposition velocities to predict or estimate dry deposition velocities is a topic of continued uncertainty and discussion. Particulate and gaseous dry deposition results have been summarized in both proceedings of conferences and in journal articles. Two international conferences have been held directed toward summarizing the current state of knowledge on dry deposition research (Pruppacher et al. 1983; Engelmann and Sehmel 1976). Much of the current research on dry deposition was summarized in 1982 in the Proceedings of the Fourth International Conference on Precipitation Scavenging, Dry Deposition, and Resuspension (Pruppacher et al. 1983). Although it would be desirable to review the recent literature here, time does not permit the development of an extensive review for this workshop. Hence, in order to assist the California Air Resources Board in assembling background data that might be used to further

develop their plans for measuring dry deposition in California, published values of dry deposition velocities are summarized from reports written by the author.

Data summarized include literature values from field measurements of gas and particle dry deposition velocities (Sehmel 1980a, 1980b) and dry deposition velocity predictions from a new least-squares correlation of surface mass transfer resistances evaluated in wind tunnel experiments (Sehmel 1984). This correlation is an improvement of the correlation published by Sehmel and Hodgson (1980).

RESISTANCES TO MASS TRANSFER

Gaseous and particulate dry-deposition removal rates from the atmosphere are a function of contaminant properties, deposition substrate properties, and meteorological parameters. To simplify analysis of the various processes contributing to transfer of airborne contaminants to the surface, contaminants are considered to be transported through a series of adjacent layers, each of which has a characteristic resistance r to vertical transfer. The resistance layers are often described in order of decreasing distance from the surface as the aerodynamic, the surface, and the substrate resistances. For particulates, a surface resistance and an aerodynamic resistance are usually considered. For gases, a substrate resistance is also considered. In general, the relationship between the resistances and the net deposition velocity is

$$V_d = (r_a + r_s + r_g)^{-1}, \quad (1)$$

where r_a is the aerodynamic resistance, r_s is the surface resistance, and r_g is substrate resistance.

The aerodynamic resistance, r_a , is solely a function of vertical transport by turbulent atmospheric eddies. Thus, it depends only on the properties of the atmosphere and the aerodynamic roughness of the surface. This turbulent transport of material can be assumed equal to either the turbulent transport of heat or momentum. Since momentum is a second order tensor and mass is not,

the transport of mass is often assumed equal to the transport of sensible heat. Although this latter equality is generally accepted as a better assumption, the experimental evidence is limited. The evidence for the similarity of the turbulent transport of mass to that of heat includes the recent results of Droppo and Doran (1983). The turbulent transport of heat is well known from micrometeorological observations (Businger et al. 1971). The aerodynamic resistance, r_a , is inversely proportional to the atmospheric turbulent mixing. Its relative importance to the surface resistance, r_s , and substrate resistance, r_g , depends in part on the height for which the deposition velocity is defined.

The surface resistance has been studied extensively, but is less well known than the aerodynamic resistance. The surface resistance, r_s , accounts for transfer across the thin layer of air in contact with the surface elements. Because atmospheric mixing becomes small at the surface, r_s for a gas depends primarily on molecular diffusion and hence on the properties of the gas, as well as the surface properties. For particulate contaminants, r_s also depends on particle inertia, impaction efficiency, and interception effects. For gases, the surface resistance is often expressed as

$$r_s = B^{-1}/u_* \quad (2)$$

where B^{-1} is an inverse Stanton number (Owen and Thompson 1963).

The substrate resistance, r_g , is least well known and reflects the processes by which the gaseous contaminants are captured and retained. This resistance is highly dependent on the properties of the gas and the deposition substrate.

DRY DEPOSITION VELOCITY DEFINITIONS

The dry deposition velocity of a pollutant is defined (Chamberlain and Chadwick 1953) as the negative ratio of the deposition flux, F , (amount depositing per unit area per unit time) divided by the airborne pollutant concentration at some height above the deposition surface. The minus sign is required because the flux downward is a negative quantity, whereas the

deposition velocity is defined as positive. Usually, the deposition velocity is reported in units of centimeters per second.

The dry deposition flux is calculated for each contaminant as the product of its airborne concentration near ground level and its dry deposition velocity. However, the height 'near ground level' has been defined as ranging from 1 cm to about 10 m. The author uses 1 cm when describing the surface mass transfer resistance for particles adjacent to the deposition surface. It is emphasized that the surface mass transfer resistance for oceans is often considered to include all mass transfer resistances below 10 m. Nevertheless, over land surfaces, airborne concentrations are often defined as those occurring at about a 1-m height.

For particulate contaminants, the particle size dependency of dry deposition velocities must always be considered. Because dry deposition velocities are a function of particle diameter, two different deposition velocity parameters are used in the literature. The symbol v_d is used sometimes for describing the average deposition velocity for a polydisperse particle size distribution of mass concentration χ . That is, the dry deposition velocity is

$$v_d = - F/\chi. \quad (3)$$

Also, the deposition velocity can be defined in terms of the number concentration, C , of each particle size per unit volume of air. The deposition velocity nomenclature, K , rather than v_d , is used to reflect that deposition velocities are a function of particle diameter. For a monodispersed particle concentration, C , the deposition velocity, $K_{1\text{-cm}}$, is defined as

$$K_{1\text{-cm}} = - F/C. \quad (4)$$

Depending on the particle size distribution, the numerical value of v_d is highly dependent on the characteristics of the size distribution. For a polydispersed aerosol, the relationship between v_d and individual K_i is

$$v_d = \frac{\sum K_i x_i}{\sum x_i} . \quad (5)$$

DRY DEPOSITION VELOCITIES OF GASES MEASURED IN FIELD EXPERIMENTS

Independent measurements of dry deposition velocities for gaseous contaminants have been reported in the literature for many experiments. Measurements through 1980 were summarized by Sehmel (1980a, 1980b) for 16 different gases. Since these data were summarized, experimental techniques using the micrometeorological gradient and eddy flux methods have advanced. Most of the data summarized are for inorganic gases. The removal of gaseous organic air pollution by dry deposition has not been studied extensively and few experimental data are available. Vapor pressure, however, is anticipated to be a very important property influencing dry deposition removal rates for organic gases.

Seventeen Gases

Recent results are decreasing the uncertainties about deposition velocities for some gases. Deposition velocities for gases are expected to be a function of diurnal variations. During the day, gaseous exchange on plant leaves is enhanced through the open stomata. At night, the stomata are closed, which results in decreased gaseous exchange through leaves. In some early work, these effects were often considered of second-order importance because even the order of magnitude for dry deposition velocities was unknown. For earlier work, reported dry deposition velocities for gases summarized by Sehmel (1980a, 1980b), and for CCl_4 reported by Cupitt (1980) are shown in Table 1. The gases are listed in order of the maximum reported deposition velocities, from the largest value to the smallest value. The ranges of deposition velocities are usually at least one order of magnitude for each gas. Some of the range might reflect atmospheric stability effects. For nitrogen oxides (NO and NO_x), the range is actually from minus to positive numbers. For all gases other than the noble gas krypton, which is unreactive (hence, a very low

TABLE 1. Dry Deposition Velocity Ranges for Gases in Order of Decreasing Maximum Reported Deposition Velocities

Depositing Gas		Dry Deposition Velocity Range, cm/s
Inorganic Gases	Organic Gases	
SO ₂		0.04 to 7.5
I ₂		0.02 to 26
HF		1.6 to 3.7
ThB		0.08 to 2.6
Fluorides		0.03 to 2.4
Cl ₂		1.8 to 2.1
O ₃		0.002 to 2.0
NO ₂		1.9
NO		minus to 0.9
	PAN	0.8
NO _x		minus to 0.5
H ₂ S		0.015 to 0.38
CO ₂		0.3
(CH ₃) ₂ S		0.064 to 0.28
	CH ₃ I	10 ⁻⁴ to 10 ⁻²
	CCl ₄	10 ⁻⁴
Kr		2.3 x 10 ⁻¹¹ maximum

deposition velocity), dry deposition velocities for gases range from 10⁻⁴ to 7.5 cm/s. For krypton the maximum deposition rate is seven orders of magnitude lower, 2.3 x 10⁻¹¹ cm/s.

Iodine and Sulfur Dioxide

Deposition velocities for iodine and sulfur dioxide have been studied extensively because these gases could be effluents from nuclear and nonnuclear industries, respectively. Even though I₂ and SO₂ have been studied extensively, their deposition velocities are so widely scattered that they cannot be predicted confidently from the data ranges shown in Table 1. As shown in Table 1, the deposition velocity of I₂ ranges over three orders of magnitude, from 2 to 26 cm/s; the deposition velocity of SO₂ ranges over two orders of magnitude, from 0.04 to 7.5 cm/s.

The dry deposition velocity data for I_2 and SO_2 are further organized in Figures 1 and 2. Reference numbers in these figures refer to references given in Sehmel (1980a). In each figure, data have been organized or ranked according to the maximum deposition velocity reported in each experiment. Commonly, deposition velocities are a function of experimental conditions and show a wide range even for the same types of deposition surfaces. This wide range is most evident for grass and water surfaces. Although a 1 cm/s deposition velocity is often assumed for gases, Figures 1 and 2 show that 1 cm/s for I_2 and SO_2 may have an uncertainty range from about 10^{-1} to 10 cm/s. In addition, maximum SO_2 deposition velocities determined in the laboratory are less than those determined under certain field conditions.

Effect of Atmospheric Stability on Gaseous Deposition Velocities

Evidence exists that the deposition velocities for gases may depend on atmospheric stability (Bunch 1968). Whelpdale and Shaw (1974) have also shown an experimental dependency on atmospheric stability. They report that the deposition velocities to grass, snow, and water surfaces under neutral conditions are greater than deposition velocities under stable conditions.

DRY DEPOSITION VELOCITIES OF PARTICULATES MEASURED IN FIELD EXPERIMENTS

Particle dry deposition velocities for many materials and various deposition surfaces were summarized by Sehmel (1980a, 1980b) who listed reported deposition velocities for different materials as a function of particle diameter. The range of experimental deposition velocities for each field experiment was presented rather than an 'average' deposition velocity. This range is critical in emphasizing the experimental uncertainties in many dry deposition field experiments and in our ability to predict dry deposition mass transfer rates. The deposition velocities reported range over five orders of magnitude from a minimum of 10^{-3} cm/s to 180 cm/s.

Sehmel (1980a) listed dry deposition velocities to land surfaces as a function of particle diameter. These particle deposition velocity data are organized graphically in Figure 3. The reference numbers refer to references

FIGURE 1. Iodine deposition velocity summary. Numbers refer to references for cited data in Sehmel (1980a).

given in Sehmel (1980a). The ranges of deposition velocities for each set of experimental conditions are shown as a function of particle diameter range. The dashed lines are for field experiments determined with polydispersed aerosols. In contrast, the solid lines are data obtained with much narrower size distributions. In general, the data show the following:

- the deposition velocities in any individual experiment range over several orders of magnitude
- a minimum deposition velocity exists and is approximately 10^{-2} cm/s for particle diameters in the range of 0.1 to 1 μ m diameter.

FIGURE 2. Sulfur dioxide deposition velocity summary. Numbers refer to references for cited data in Sehmel (1980a).

Because of meteorological and deposition surface complexities in the field, deposition velocity predictors for large vegetative canopies are expected to be even more complex than predictors developed for simple surfaces in wind tunnel experiments. However, as discussed below, dry deposition velocity predictors are complex even for surfaces investigated in wind tunnel experiments. Particle dry deposition velocities are a function of the Schmidt number, the particle relaxation time, the ratio of Brownian to eddy diffusivity, and a length ratio. Also, dry deposition velocities should be a function of other parameters and variables including leaf area index and atmospheric stability.

Hence, generalizations made from these early field experiments have been limited, in part because experimental variables were not adequately controlled or reported, based on our current understanding of dry deposition (i.e., often

FIGURE 3. Particle deposition velocities measured in the field. Numbers refer to references for cited data in Sehmel (1980a).

the particle size distribution was either not known or not reported). Since deposition velocities are a function of particle diameter, most early field-determined mass transfer data must be interpreted carefully. It is hoped that field experiments conducted since the summary have had better control of experimental conditions.

PREDICTIONS OF DRY-DEPOSITION VELOCITIES AS A FUNCTION OF PARTICLE DIAMETER

Dry deposition velocities determined in early field experiments encompass the range predicted by Sehmel and Hodgson's empirical model (1980), which was based on wind tunnel measurements of dry deposition velocities for monodispersed particles. As discussed below, the predictive model was improved

in 1984. Similar to these field experiments, predictions indicate a minimum of about 10^{-2} cm/s for particle diameters ranging from 0.1 to 1 μm . Thus, for generalizing about dry deposition velocities, the 1984 predictive model is recommended. These predictions are a function of particle diameter, friction velocity, aerodynamic surface roughness, and particle density.

1980 Correlation

The predictive correlations are based on deposition velocities evaluated in wind tunnel experiments for non-living surfaces of different aerodynamic surface roughness. The 1980 correlation is based on a correlation of the integral surface resistance, Int , defined in equations 6 and 7. Dry deposition velocities with a concentration reference height of 1 cm are calculated from the expression

$$K_{1\text{-cm}} = \frac{v_t}{1 - (1/\alpha)} \quad (6)$$

in which v_t is the gravitational settling velocity of particles and

$$\alpha = \exp(-v_t \text{Int}/u_*) \quad (7)$$

The expression for Int was developed using a weighted least-squares technique to correlate the logarithm of mass-transfer resistances, Int , in terms of the logarithms of dimensionless groups and particle diameter. The dimensionless groups are the Schmidt number (ν/D), the particle relaxation time (τ_+), a ratio of Brownian to eddy diffusivity ($D/z_0 u_*$), and a length ratio (d/z_0). The multiple correlation coefficient was 0.92, and all coefficients were statistically significant at the 99% level.

Subsequently, deposition velocities were predicted as a function of particle diameter from 0.03 to over 50 μm . However, the 1980 correlation was inadequate for predictions below 0.03 μm diameter. That is, predicted deposition velocities started to decrease as particle diameter was decreased below 0.03 μm .

The 1980 correlation was developed to predict the surface mass transfer resistance (reciprocal deposition velocity) in the lower 1 cm adjacent to uniform deposition surfaces without deep vegetative canopies. In this case, the objective was to develop a mass transfer resistance predictor that could be used as a boundary condition for meteorological transport and diffusion models in which aerodynamic resistances were described for heights closer than 1 m to the deposition surface. In addition, the 1980 correlation was used to predict particle dry deposition velocities for a height of 1 m. This height is often used for a boundary condition over surfaces without large vegetative canopies in meteorological transport and diffusion models. In this case, the modeling approach was to combine mass-transfer resistances in the 1 cm adjacent to the surface determined by the correlation, with aerodynamic mass transfer resistances from 1 cm to 1 m determined by the diffusivity equations of Businger et al. (1971).

When the 1980 correlation was developed, particle diffusivities were assumed equal to air-momentum diffusivities. However, the validity of this assumption is dependent on the relative mass transfer resistances at and above the deposition surface. Currently, the equality to heat diffusivity is often considered more appropriate; however, the diffusivity selection may be a second-order effect. In practice, the use of either momentum or heat diffusivity is only of secondary importance for predicting deposition velocities for particle diameters ranging from about 0.1 to 1 μm . For these diameters, deposition velocities are controlled by surface resistances in the 1 cm adjacent to surfaces of low relief. In addition, selecting the proper diffusivity may be of minor importance compared to the uncertainties in predicting integrated surface resistances as a function of height and spatial variation in vegetative canopies.

Dry deposition velocities are complex functions of the parameters determined. The 1980 correlation (as well as the 1984 correlation) predicts dry deposition velocities as a function of particle diameter, friction velocity, and aerodynamic surface roughness. Also, because the experimental particle density was 1.5 g/cm³, the influence of particle density is predicted by including particle settling velocities in the prediction equations.

Because predictions are complex functions of these variables, some researchers have attempted to approximate the predicted deposition velocity curves obtained with the 1980 correlation. However, such simplification may compound prediction uncertainties, because uncertainties exist in the original correlation.

Hence, in 1984, particle dry deposition velocities evaluated with monodispersed-diameter particles in wind tunnel experiments were reexamined using least-squares techniques to develop an improved predictive correlation.

1984 Correlation

Least-squares techniques were used to correlate dry deposition velocities evaluated in wind tunnel experiments (the same data base used in developing the 1980 correlation) as a function of both particle diameter and dimensionless groupings. Logarithmic transformations were used in the least-squares analyses. Many correlations were examined for the highest multiple correlation coefficient. This examination included the evaluations of the statistical significance of the first through third powers of each dimensionless group examined and interactions between groups. Third powers were statistically insignificant. The correlation finally selected has the highest multiple correlation and is without variables that are highly correlated with other predictor variables. The multiple correlation coefficient is 0.95 (versus 0.92 in the 1980 correlation), and all coefficients are statistically significant at the 99% level.

This correlation predicts deposition velocities, $K_{1\text{-cm}}$ with an airborne concentration at 1 cm, and relates the logarithm of deposition velocity as a function of the logarithms of dimensionless groups and particle diameter. Particle diameter is expressed in centimeters. The correlation of deposition velocities as an exponential function of the logarithms of dimensionless groups and particle diameter follows:

$$\begin{aligned}
K_{1-\text{cm}} = & \exp (7.1108 \\
& + [\ln(\text{Sc}) \ln(d/z_0)] [0.2529 - 0.00273 \ln(d/z_0)] \\
& + [\ln(\tau^+)] [0.03239 \ln(\tau^+) - 0.09177 \ln(d/z_0)] \\
& - 0.14919 [\ln(d)]^2 \\
& - 4.180 \ln(d/z_0)) .
\end{aligned} \tag{8}$$

The Brownian diffusivity was calculated from (Davies 1966):

$$D = \frac{kT}{6\pi\mu a} \left(1 + \frac{10^{-4}}{pa} [6.32 + 2.01 \exp(-2190 pa)] \right) \tag{9}$$

where a = particle radius, cm

d = particle diameter, cm

D = Brownian diffusion coefficient, cm^2/s (see Equation 9)

k = Boltzmann's constant, 1.38×10^{-16} erg/(molecule K)

p = pressure, cm of mercury (76.0 cm used)

Sc = Schmidt number, ν/D

T = temperature, K (296°K used)

u_* = friction velocity, cm/s

z_0 = aerodynamic surface roughness, cm

μ = air viscosity, g/(cm·s) [1.78×10^{-4} g/(cm·s) used]

ν = kinematic viscosity, μ/ρ

ρ = air density, g/cm³, (1.2×10^{-3} g/cm³ used)

ρ_p = particle density, g/cm³

$\tau^+ = \text{dimensionless relaxation time, } (\rho_p d^2 u_*^2)/(18 \mu \nu)$

Ratios of predicted deposition velocities divided by experimental deposition velocities were evaluated as indices for agreement between predicted and experimental results. The average ratio is 1.46, the maximum ratio is 19.3, the minimum ratio is 0.82, and the standard deviation is 1.78.

Also, ratios of predicted deposition velocities of the correlation developed in 1980 and Equation 8 were evaluated to show agreement between the two correlations for the experimental conditions used (i.e., the ratio K_{1980}/K_{1983}). In this case, the average ratio is 1.13, the maximum ratio is 3.88, the minimum ratio is 0.44, and the standard deviation is 0.68.

Predictions for Constant Friction Velocity

Predicted deposition velocities for a 1-cm concentration reference height, $K_{1\text{-cm}}$, from the 1980 correlation and 1984 correlation (developed in 1983 and published in 1984) are compared in Figure 4 as a function of aerodynamic surface roughness for a constant friction velocity of 30 cm/s and particle density of 1.5 g/cm^3 . Also shown are gravitational settling velocities for spherical particles. The following features are emphasized.

- Predictions from the 1984 correlation (as opposed to predictions from the 1980 correlation, which have a lower particle diameter limit of $0.03 \text{ }\mu\text{m}$) show trends expected for particle diameters as small as $10^{-2} \text{ }\mu\text{m}$. That is, in the 1984 correlation, deposition velocities continue to increase as particle diameter decreases below the particle diameter for minimum dry deposition velocity.

FIGURE 4. Predicted deposition velocities at 1-cm concentration reference height for a particle density of 1.5 g/cm^3 and a friction velocity of 30 cm/s from the 1980 and 1984 predictive correlations.

- Minimum deposition velocities are functions of the aerodynamic surface roughness.
- Dry deposition velocities are greater than gravitational settling velocities.
- Aerodynamic resistances from 1 cm to 1 m are increasingly important for large particles. Predicted deposition velocities for a 1-m concentration reference height would be similar to gravitational settling velocities. In comparison, because aerodynamic resistance is not included, deposition velocities for a 1-cm concentration reference height can be significantly greater than gravitational settling velocities for particle diameters as large as 50 μm .

CONCLUSIONS

Considerable research is yet needed to accurately evaluate and to generally predict dry deposition removal rates for gases and particulate contaminants. An improved predictive correlation was published in 1984 for predicting particle dry-deposition velocities with a concentration reference height of 1 cm. One conclusion from the correlation is that particle size dependencies always must be considered in designing field experiments. However, further improvements in predictive methods are needed including methods to predict effects of deeper canopies, vegetative canopies, nonuniform canopies, nonspherical particles, and simultaneous resuspension for particles 10 to 20 μm in diameter.

Based on wind tunnel experiments, deposition velocities are a function of several dimensionless groups: the Schmidt number (ν/D), the particle relaxation time (τ^+), a ratio of Brownian to eddy diffusivity ($D/z_0 u_*$), and a length ratio (d/z_0). Dry deposition velocities are minimum in the particle diameter range of about 0.1 to 1 μm . If improved predictors for field applications are to be developed, evaluations of these groups should be considered in designing field experiments. In addition, atmospheric stability, leaf area index, and fetch nonuniformities must be considered.

At present because information is not available for these and other variables, we must conclude that dry deposition velocities for both gaseous and particulate contaminants cannot be accurately predicted from published results of early field experiments. In addition, extrapolation of summarized field results of dry deposition velocities is confounded by the wide deposition velocity ranges that have been measured. Hence, field experiments should be designed to include deposition removal variations as a function of height and distance to determine reproducibility. Possibly inclusion of more recently published field data, and a critical review of experimental techniques, might decrease the expected ranges of deposition velocity.

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