A Linear Model for Predicting the Washout of Pollutant Gases from Industrial Plumes

A simplified method for predicting the washout of gaseous constituents from industrial plumes has been developed. This method is a linearized version of a more general treatment given previously. It is based upon an application of conventional mass-transfer theory in conjunction with existing plume dispersion models and accounts for reversibility of the adsorption process and liquid-phase mass-transfer resistance. The method presented in this paper is sufficiently simple to permit hand calculation, if necessary.

This model is recommended for calculation of washout of gases from plumes under conditions where the gas exhibits physical or pseudophysical absorption behavior. It is expected to apply with reasonable accuracy for gases with linear solubility relationships; accuracy for situations involving other gases is expected to be less satisfactory, depending upon the degree of deviation from this idealized behavior.

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SCOPE

When natural precipitation falls through polluted atmospheres, a washout process occurs which is often highly effective in removing pollutant constituents. This mechanism, therefore, can be a rather important factor in determining the environmental impact of chemical process emissions and, consequently, the allowable levels of their release. Development of methods for predicting the behavior of this process should lead to improved design criteria for control units, stacks, and associated equipment.

The effectiveness of precipitation washout as a sink for plumeborne particulate matter has been demonstrated experimentally in numerous investigations and has been treated with fair success theoretically using the washout-coefficient approach (Junge, 1963). The few existing reports of gas-washout studies, however, have indicated that pollutant gases are scavenged less effectively than predicted on the basis of washout-coefficient theory. It has been suggested that such behavior should be expected, owing to the increased importance of liquid-phase phenomena and reversibility under these circumstances.

Recently a model has been developed to describe the washout of SO_2 from industrial plumes (Dana et al., 1972). This model appears to describe washout in a satisfactory manner, at least under relatively simple conditions. Because of system nonlinearities and other complicating factors, however, numerical computations are required to provide washout estimates. The present paper describes a simpler, linearized model for reversible gas washout which does not require machine computation and can be used for engineering estimates of washout of pollutants from industrial plumes under a variety of circumstances.

CONCLUSIONS AND SIGNIFICANCE

A method for predicting washout of gaseous constituents from industrial plumes has been developed. This method is recommended for estimating gas washout from plumes at short to moderate distances from the source. For larger distances or different source configurations other forms which may be derived directly from basic equations in this development may be used, provided the spatial distribution of the gas-phase pollutant concentration is known.

Comparison of calculated and measured washout rates suggest that lumping of the raindrop size distribution to the mass-mean drop size is an acceptable simplification in applying this method, although it is preferable to treat these statistics in a more rigorous manner. Gases exhibiting nonlinear solubility behavior can be treated by choosing an intermediate, constant partition coefficient value; more refined estimates, however, can be obtained using a more general numerical approach presented previously.

THEORETICAL DEVELOPMENT

As discussed elsewhere (Hales, 1972) the interaction between rain and air pollutants may be described in terms of a two-phase system, the atmosphere and the rain composing the disperse and continuous phases, respectively. Assuming the rain to be composed of spherical, noninteracting drops of static size distribution, the corresponding equations for conservation of pollutant may be written in terms of divergence and source-sink terms as follows:

$$\frac{\partial \rho_{Ay}}{\partial t} = -(\nabla \cdot \rho_{Ay} \bar{v}_A) - w + r_{Ay} \qquad (1)$$

continuous phase (air)

$$\frac{\partial \rho_{Ax}}{\partial t} = -\frac{4\pi N_0}{3}\left(\nabla \cdot \int_0^\infty a^3 f(a)\, \overline{v}_t\, c\,(a)\, da \right) + w + r_{Ax} \quad (2)$$

disperse phase (rain)

Equation (1) is seen to be identical with the conventional single-phase continuity equation, except for the term w, which accounts for the rate of removal of pollutant A from the gas phase by washout. Since w appears in both Equations (1) and (2), it serves as a coupling entity, which necessitates simultaneous solution unless some simplifying assumption can be made. For the stipulated conditions of spherical, noninteracting raindrops, w can be given as

$$w = -4\pi N_0 \int_0^\infty a^2 f(a)\, N_{A0}\, da$$

$$= 4\pi N_0 \int_0^\infty a^2 f(a)\, K_y(a)\, (y_{Ab} - H'c)\, da \quad (3)$$

N_{A0} represents the average flux of material from the surface of a raindrop. For present purposes we will assume that the system is linear in the sense that the mass-transfer coefficient K_y and the effective Henry's-law constant H' are invariant with concentration. In addition we shall simplify Equations (1) and (2) by assuming that

1. a steady state exists
2. chemical reaction is negligible
3. w is small compared to the divergence term in Equation (1), and
4. all raindrops fall through vertical trajectories.

The above assumptions effectively decouple Equations (1) and (2), and reduce (2) to the form

$$\int_0^\infty a^3 f(a)\, v_z \frac{\partial c}{\partial z}\, da = 3\int_0^\infty a^2 f(a)\, K_y\, (y_{Ab} - H'c)\, da \quad (4)$$

which may be integrated with respect to z to obtain an expression for the flux of rainborne pollutant through an incremental area at any level z in the atmosphere, that is,

$$F_z = \frac{4\pi N_0}{3}\int_0^\infty a^3 f(a)\, v_z\, c(a,z)\, da \quad (5)$$

At $z = 0$ (ground level) Equation (5) reduces to

$$F_0 = \frac{4\pi N_0}{3}\int_0^\infty a^3 f(a)\, v_z\, c(a,o)\, da \quad (6)$$

which is of primary interest to our analysis.*

Because of the assumption that raindrops do not interact, it is allowable to solve these equations by first reducing Equation (4) to the form

$$\frac{dc}{dz} = \frac{3K_y}{v_z a}\,(y_{Ab} - H'c) \quad (7)$$

which pertains to any individual raindrop of radius a. This then may be integrated with respect to z and distributed over the raindrop size spectrum to calculate washout fluxes in accordance with Equation (6). Integration with respect to z, assuming an initially clean drop, results in the form

$$c(a,z) = \frac{3K_y}{v_z a}\exp\left(\frac{-3K_y H' z}{v_z a}\right)\int_\infty^z \exp\left(\frac{3K_y H' z}{v_z a}\right) y_{Ab}\, dz \quad (8)$$

for drop concentration at any point, or

$$c(a,o) = \frac{3K_y}{v_z a}\int_\infty^0 \exp\left(\frac{3K_y H' z}{v_z a}\right) y_{Ab}\, dz \quad (9)$$

at ground level.

The spatial distribution of pollutant in the gas phase (y_{Ab}) must be known prior to solving Equation (9) to obtain numerical values. y_{Ab} can be obtained through actual field measurements; it is more practical, however, to estimate this entity using published solutions to various forms of Equation (1). In the present paper, washout from plumes emitted from single stacks into the atmosphere, as depicted by Figure 1, is of primary interest. Description of gas-phase concentrations under such circumstances has been the subject of numerous modeling efforts; one of the better known of these models has resulted in the Pasquill-Gifford bivariate-normal plume equation (compare Turner, 1971),

$$y_{Ab}(x,y,z) = \frac{Q}{2\pi\,\sigma_y\,\sigma_z\,\overline{u}}\exp\left[-1/2\,(y/\sigma_y)^2\right]\left\{\exp\left[-1/2\left(\frac{z-h}{\sigma_z}\right)^2\right] + \exp\left[-1/2\left(\frac{z+h}{\sigma_z}\right)^2\right]\right\} \quad (10)$$

This equation is essentially a solution to Equation (1) subject to a number of assumptions, including zero w and r_{Ay}, constant eddy diffusivities in the horizontal and vertical directions, zero longitudinal mixing, point-source emission, total reflection at the surface, and constant wind speed $\overline{u}$. Values of the spread parameters σ_y and σ_z can be obtained from anemometer measurements or from the literature (Turner, 1971).

One should note that the y_{Ab} given in Equation (10) is intended to be a time-averaged entity, whereas the value appearing in Equation (7) was depicted as the instantaneous value experienced by the drop along its own particular trajectory to the ground. Since all plumes fluctuate with time in a complex manner, there is no

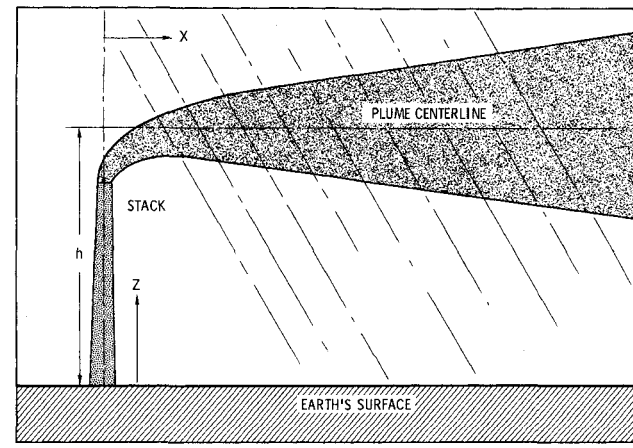

Fig. 1. Schematic of rain-plume interaction.

* Here it is convenient to note that the rainfall rate is defined by the equation

$$I = -\frac{4\pi N_0}{3}\int_0^\infty a^3 f(a)\, v_z\, da.$$

guarantee that the instantaneous concentration should resemble the time-averaged value at all. It has been shown elsewhere (Hales et al, 1971), however, that the use of time-averaged concentrations in (9) will result in the calculation of the mean concentration of the ensemble of drops that have fallen to the ground provided that the washout process is linear in the sense described earlier. For nonlinear systems this use of averages will give rise to discrepancies whose magnitudes will depend upon the extent of deviation from linearity.

Substitution of (10) into (9) and integration results in the form

$$c(a,o) = -\frac{Q\,F\,\xi}{2\sqrt{2\pi}\,\sigma_y\,\overline{u}} \exp\left(-\frac{y^2}{2\sigma_y^2} + \frac{\sigma_z^2\zeta^2}{2}\right) \left\{ \exp(\zeta h)\left[1 - \operatorname{erf}\left(\frac{-\sigma_z^2\zeta - h}{\sigma_z\sqrt{2}}\right)\right] + \exp(-\zeta h)\left[1 - \operatorname{erf}\left(\frac{-\sigma_z^2\zeta + h}{\sigma_z\sqrt{2}}\right)\right]\right\}, \quad (11)$$

where

$$\zeta = \frac{3K_yH'}{v_za} \quad (12)$$

$$\xi = \frac{3K_y}{v_za} \quad (13)$$

and F is a factor included to account for depletion of the plume by washout. One should note that representation of plume modification solely in terms of F implies the assumption of negligible distortion of plume geometry by the washout process. Such an assumption is permissible for small-to-moderate downwind distances from the source where F is still fairly close to unity. For moderate downwind distances the following asymptotic approximation to (11) is useful:

$$c(a,o) = \frac{-Q\,\sigma_z\,F\,\xi}{2\pi\,\sigma_y\,\overline{u}} \exp\left(-\frac{y^2}{2\sigma_y^2} - \frac{h^2}{2\sigma_z^2}\right) \left\{\frac{1}{-\sigma_z^2\,\zeta - h}\left[1 - \left(\frac{\sigma_z}{-\sigma_z^2\,\zeta - h}\right)^2\right] + \frac{1}{-\sigma_z^2\,\zeta + h}\left[1 - \left(\frac{\sigma_z}{-\sigma_z^2\,\zeta + h}\right)^2\right]\right\} + 0\left(\frac{\sqrt{2}\,\sigma_z}{\sigma_z^2\,\zeta - h}\right)^4. \quad (14)$$

The final term in (14) is a truncation-error estimate, 0 denoting order of.

Equations (11) or (14) can be employed directly with raindrop spectrum data to calculate ground-level washout fluxes using Equation (6). They also may be used to calculate average concentration in rain, and the total amount of pollutant washed out as a function of downwind distance from the source. These two quantities may be defined as

$$c_{\text{avg}} = \frac{F_0}{J} \quad (15)$$

and

$$W = \int_0^x \int_{-\infty}^{\infty} F_0\,\partial y\,\partial x \quad (16)$$

The plume depletion factor F may be calculated by the expression

$$F = 1 - \frac{W}{Q\,c_y} \quad (17)$$

Depending on how the raindrop size statistics are treated the previous equations may reduce to explicit forms upon incorporating (17), or they may be implicit in nature, necessitating iterative or numerical approximations to their solutions.

Although the bulk of effort in this paper is devoted to normally distributed plumes, the general applicability of Equation (9) should be emphasized. A step function or a top hat plume distribution, characterized by

$$y_{Ab} = 0 \text{ for } z > d,$$
$$y_{Ab} = S_n \text{ for } d < z < b, \ (S_n \text{ a constant})$$
$$y_{Ab} = 0 \text{ for } z < b \quad (18)$$

can be substituted into Equation (9) to provide the following expression for ground-level drop concentration;

$$c(a,o) = S_n\frac{\xi}{\zeta}F\,(e^{\zeta b} - e^{\zeta b}) \quad (19)$$

Similarly, general descriptions of plumes from line sources, area sources, puff releases, etc. can be employed to provide corresponding results.

ESTIMATION OF MASS-TRANSFER COEFFICIENTS

Mass-transfer coefficients for gas absorption by falling drops has been the subject of extensive discussion in the literature. For present purposes, however, this paper will confine discussion to limiting conditions, namely those posed by gas-phase limited and stagnant drop behavior. Behavior of real systems should fall somewhere between these extreme cases.

The gas-phase mass-transfer coefficient can be estimated from the well-known Froessling equation (compare Bird et al., 1960),

$$\frac{2\,a\,k_y}{D_{Ay}\,c_y} = 2 + 0.6\left(\frac{-2\,a\,v_z}{\nu}\right)^{1/2}\left(\frac{\nu}{D_{Ay}}\right)^{1/3} \quad (20)$$

For stagnant-drop conditions the liquid-phase mass transfer coefficient can be calculated using published solutions to the continuity equation describing diffusion into a sphere (Carslaw and Jaeger, 1959). Although the resulting values are concentration dependent, they can be approximated by constant values corresponding to drops experiencing ramp-function changes in surface concentration (Hales, 1972), thus,

$$k_x = \frac{5\,D_{Ax}c_x}{a}. \quad (21)$$

Equations (20) and (21) may be used to calculate the overall coefficient for the limiting cases using the well-known relationship

$$K_y = \frac{1}{\frac{H'c_x}{k_x} + \frac{1}{k_y}}. \quad (22)$$

EXAMPLE CALCULATIONS—COMPARISON WITH EXPERIMENTAL RESULTS

Equation (11) may be used directly to predict washout concentrations in rain upon obtaining estimates of the source, dispersion, and transport properties pertinent to the system of interest. Field experiments involving the analysis of rain samples collected downwind from continuous, elevated releases of SO_2 have been conducted recently (Dana et al, 1972), allowing some comparisons of predicted and measured results. For an example, consider an SO_2 plume being emitted to the atmosphere

under conditions characterized by Table 1. The parameters listed here correspond to an actual field experiment conducted at Quillayute, Washington, during March of 1971.

Raindrop-size spectrometer data obtained during this experiment are given in Table 2.

Terminal fall velocity as a function of drop radius can be estimated from the equation

$$v_z = -8710.9a + 18029.7a^2 + 32184.5a^3, \qquad (23)$$

which is a polynomial fit to the data of Gunn and Kinzer (1949). Solubility of SO_2 in water may be calculated on the basis of recently published low-concentration data (Dana et al., 1972). For the present calculations the solubility relationship was linearized, using an effective Henry's-law constant taken as the average between those calculated at ground level and release height at the downwind sampling distance of 121 meters.

Mass-transfer coefficients for the calculations were estimated using Equations (20) to (22). Molecular transport data utilized for this purpose are given in Table 3.

Washout concentrations pertaining to a given drop size may be calculated in a straightforward manner using Equation (11) in conjunction with the above data. Distributed systems may be approached by performing a number of such calculations for selected drop sizes (for example, those shown in Table 2), and then combining the contributions to evaluate a mixed-mean concentration corresponding to a collected rain sample. Alternatively, one could obtain a simpler and less exact estimate by lumping the raindrop statistics into one representative drop size (for example, the mass-mean drop diameter) and subsequently solving Equation (11) only once. Concentrations of SO_2 in rain calculated using both of these approaches are shown in Figure 2, along with actual concentrations measured during the experiment, depicted by vertical bars. Concentrations calculated on the basis of the well-mixed drop assumption are denoted by solid curves while those based on stagnant drop behavior are shown by the broken lines.

As can be observed from Figure 2, the calculations based on a well-mixed drop of mass-mean diameter provide closest agreement with peak concentration values for this example. Total washout rates (proportional to areas beneath the curves), however, are matched more closely by the distributed system calculations. Both the single and distributed drop-size calculations were found nearly to coincide for stagnant drop conditions, which

TABLE 1. CHARACTERISTICS OF SO_2 WASHOUT EXPERIMENT

SO_2 release rate, Q	0.0816 moles/s
SO_2 release height, h	7.62 meters
Mean wind velocity, $\bar{u}$	3.5 meters/s
Downwind rain sampling distance	121 meters
Dispersion parameters (calculated from anemometer measurements):	
σ_y	29.4 meters
σ_z	9.4 meters
Ambient temperature	9°C

TABLE 2. SUMMARY OF MEASURED RAINDROP SIZE STATISTICS

Raindrop diam. range, cm	Fraction of drops in size range
0-0.0249	0.098
0.0249-0.0299	0.123
0.0299-0.0360	0.105
0.0360-0.0430	0.241
0.0430-0.0515	0.173
0.0515-0.0615	0.073
0.0615-0.0740	0.054
0.0740-0.0900	0.023
0.0900-0.107	0.050
0.107-0.129	0.027
0.129-0.154	0.028
0.154-0.169	0.005

TABLE 3. MOLECULAR TRANSPORT DATA USED IN CALCULATING MASS-TRANSFER COEFFICIENTS

Property	Value	Source
Diffusivity of SO_2 in air	0.136 cm^2/s	(Reid and Sherwood, 1958)
Diffusivity of SO_2 in water	9×10^{-6} cm^2/s	(Lynn et al, 1955)
Kinematic viscosity of air	0.133 cm^2/s	(Reid and Sherwood, 1958)

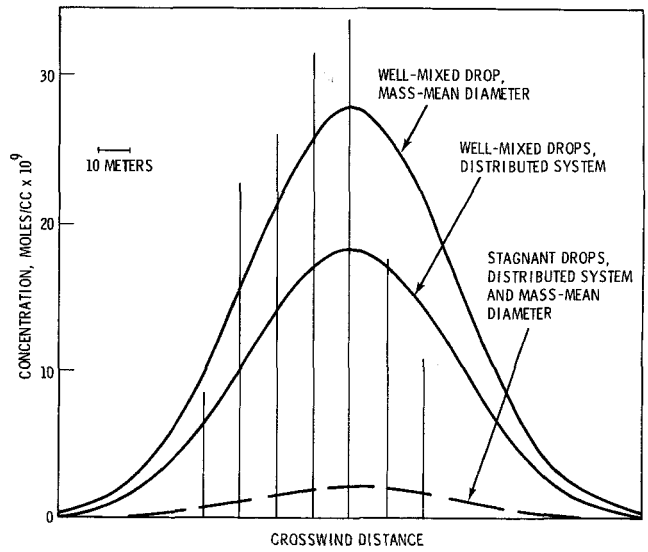

Fig. 2. Observed crosswind distribution of SO_2 concentrations in rain compared with those predicted by linear washout model. Conditions given in Tables 1 to 3.

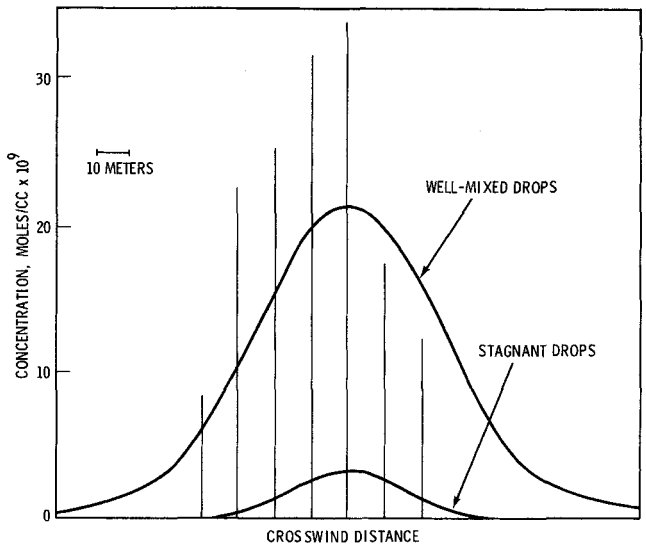

Fig. 3. Observed crosswind distribution of SO_2 concentrations in rain compared with those predicted by nonlinear washout model. Conditions given in Tables 1 to 3.

obviously provided a low estimate of true behavior in this instance.

The previously mentioned computer model, which accounts for nonvertical rain trajectories and nonlinear solubility behavior, has also been applied to the system described in Tables 1 to 3. Shown in Figure 3, the results computed from this model do not deviate markedly from those based on the applications of Equation (11) in conjunction with the more exact description of raindrop size statistics.

Figures 2 and 3 pertain to a single experiment and therefore provide only a partial test of the predictive capability of Equation (11). A more complete test is exhibited in Figure 4, which is a plot of predicted washout rates, as defined by the equation

$$M = J \int_{-\infty}^{\infty} c_{\text{avg}}(y)\, dy, \qquad (24)$$

versus those measured experimentally for SO_2 during a number of field experiments. All computations leading to the predicted rates shown in Figure 4 were based upon the use of the mass-mean diameter as the representative drop size. Limits on the horizontal lines correspond to the limits of well-mixed and stagnant drop behavior.

A similar plot based on results of the more elaborate computer model applied to measured drop-size distributions is shown in Figure 5 for comparison. Experimental conditions for these tests are summarized in Table 4, and a more complete description is given elsewhere (Dana et al., 1972).

From Figures 4 and 5 it is apparent that the predictive capabilities of this linear model, at least as applied with raindrop statistics lumped to the mass-average drop size, are somewhat inferior to those of the more exact treatment.

Evaluation of individual factors contributing to the deviations between theory and experiment shown in Figures 4 and 5 is complicated by the variety of assumptions that have been incorporated by the model. Uncertainties involved with applying limiting models of mass-transfer behavior can be evaluated rather directly* by observing the lengths of the lines on the figures; however, such factors as uncertainties in the plume dispersion model and the assmptions of a linear system composed of vertically-falling, spherical, noninteracting drops of static size distribution are more difficult to assess. Certainly any deviation between true plume behavior and that predicted by the plume Equation (10) will have a direct influence on the quality of the corresponding washout calculations. An indication of the extent of such deviations is given in Figure 6, which compares airborne SO_2 concentrations measured at various ground-level sampling points during the washout experiments with those predicted by Equation (10). Although these data are insufficient for a complete quantitative assessment of deviations arising from application of Equation (10) to the washout model, they do indicate qualitively that washout calculations of this type would benefit substantially from an improved ability to describe plume behavior.

Because of the assumptions of linearity used to derive Equation (11) it seems reasonable to expect that agreement between experiment and theory should be improved for gases that do not exhibit the strongly nonlinear solubility behavior characteristic of sulfur dioxide. From a comparison of Figures 2 and 3 it is evident, however, that the manner in which the raindrop spectrum is treated is of more importance in determining the outcome of the predicted results.

In view of these considerations the above results suggest

Table 4. Summary of Experiment Conditions for Data Exhibited in Figures 4 and 5

Variable	Range
Release height	7.6-30.5 meters
Downwind sampling distance	30.5-122 meters
SO_2 release rate	0.0063-0.0944 moles/s
Wind speed	2.2-7.6 meters/s
Rainfall rate	2.2×10^{-5}-11.1×10^{-5} cm/s

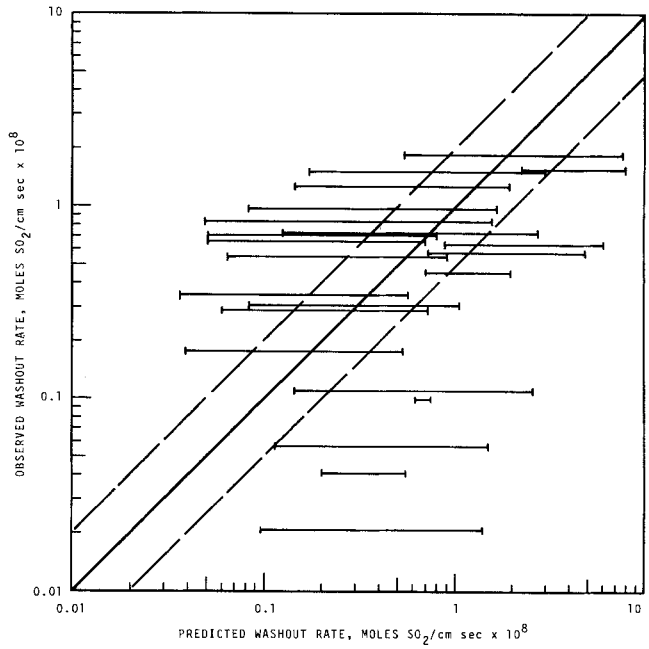

Fig. 4. Comparison of observed washout rates with those predicted using the mass-mean raindrop size in conjunction with the linear model. Ranges of conditions summarized in Table 4. Dashed lines denote deviation by factor of two.

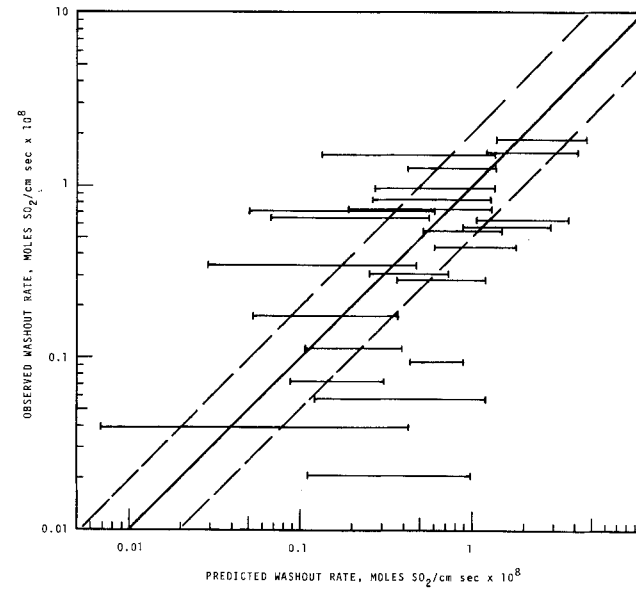

Fig. 5. Comparison of observed washout rates with those predicted using nonlinear model in conjunction with distributed raindrop statistics. Ranges of conditions summarized in Table 4. Dashed lines denote deviation by factor of two.

* It should be noted here, however, that limiting values of the mass-transfer coefficient do not necessarily define limiting values of the washout rate.

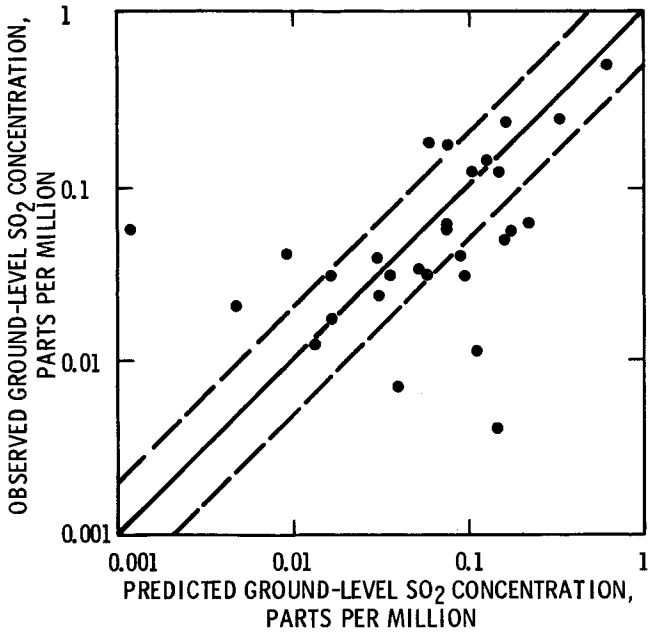

Fig. 6. Comparison of observed concentrations of SO_2 in air with those predicted on basis of Equation (10).

that the procedure described in this paper should be a reasonably valid means of estimating gas washout. It is recommended for linear systems and for nonlinear systems when preliminary estimates are required; more reliable results will be provided using more sophisticated techniques, however. It also should be noted that all data presently available for comparison pertain to relatively close distances from the source. At much greater distances significant plume modification may occur from dry deposition and other effects, necessitating creation of a modified equation based upon the altered distribution of the plume.

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NOTATION

a = drop radius, cm
c = mixed-mean concentration in drop, moles/cm^3 water
c_x = total concentration of liquid in water, moles/cm^3 water
c_y = total concentration of gas in air, moles/cm^3 air
D_{Ax} = diffusivity of component A in water, cm^2/s
D_{Ay} = diffusivity of component A in air, cm^2/s
F = fraction of plume not removed by washout
F_z = washout flux of pollutant at elevation z, moles/cm^2 s
f = probability-density function for size spectrum of raindrops, 1/cm
H' = apparent Henry's-law constant for pollutant in rain, cm^3/mole
h = effective release height of plume, cm
J = rainfall rate cm/s
K_y = overall mass-transfer coefficient, moles/cm^2 s
k_x = liquid-phase mass-transfer coefficient, moles/cm^2 s
k_y = gas-phase mass-transfer coefficient, moles/cm^2 s
N_{A0} = average flux of pollutant A from a raindrop moles/cm^2 s
N_0 = number concentration of drops in rain
Q = volumetric pollutant release rate, calculated at ambient conditions cm^3/s
r_{Ax} = generation of pollutant by liquid-phase reaction, moles/cm^3 s
r_{Ay} = generation of pollutant by gas-phase reaction, moles/cm^3 s
S_n = Heaviside step-function operator
t = time, s
$\bar{u}$ = mean wind velocity, cm/s
$\bar{v}_A$ = velocity vector for pollutant A in air cm/s
$\bar{v}_t$ = velocity vector for pollutant A in rain, cm/s
v_z = terminal velocity of raindrop, cm/s
w = washout rate, moles taken into liquid phase per unit time per unit total volume of space, moles/cm^3 s
W = total washout of pollutant, moles/s
x = downwind distance from source, cm
y = crosswind distance from plume centerline, cm
y_{AB} = mole-fraction of pollutant in gas-phase bulk
z = distance above ground, cm

Greek Letters

ζ = constant defined by Equation (12), 1/cm
ξ = constant defined by Equation (13) moles/cm^4
ν = kinematic viscosity of air, cm^2/s
ρ_{Ax} = molar density of A in liquid phase, moles/total volume of space, moles/cm^3
ρ_{Ay} = molar density of A in gas phase, moles/total volume of space, moles/cm^3
σ_y = crossplume plume-spread parameter, cm
σ_z = vertical plume-spread parameter, cm

Subscripts

A = pollutant A
b = bulk
0 = interface or ground level
x = liquid phase
y = gas phase

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