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THE CALCULATION OF PRECIPITATION SCAVENGING

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THE CALCULATION OF PRECIPITATION SCAVENGING

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ABSTRACT

THE CALCULATION OF PRECIPITATION SCAVENGING

This review is designed to give a solid and up-to-date basis for newcomers and fringe workers in precipitation scavenging. It is also intended as a guide to quantitative predictions.

Collection efficiencies, washout coefficients for particles and gases, rain spectra, and washout by snow are discussed, and values given, of which some are previously unpublished. Extensive "area within a washout isopleth" charts are shown for three atmospheric stabilities and several washout coefficients.

NOTATION

a	Particle radius
A	Cross sectional area of drop of diameter, D
d	Particle diameter (the collected)
D	Drop diameter (the collector)
e	Exponential
E	Scavenging efficiency
F	Flux density of drops (drops/area time diameter interval)
k_1	Concentration of contaminant in cloud water
K	Impaction parameter, or Stokes' number
L/ρ	Fraction of cloud space filled with liquid water
M	Mass of aerosol particle of diameter d
N	Concentration of drops in air (drops/volume diameter interval)
Q_o	Plume source strength (mass, or mass/time)
Re	Reynold's number of waterdrop
Sh	Nusselt diffusion number or Sherwood number
t	Time
T	Age of cloud
$\bar{u}$	Wind speed
U	Obstacle (raindrop) speed
x	Distance downwind
y	Distance crosswind from plume centerline
Y	Diffusivity of a vapour in air
z	Height
α	Radioactivity per mass of particle
ϵ	Rainout efficiency, $1 - (\chi/\chi_o)$
Λ	Washout coefficient (fraction/time)
μ	Dynamic viscosity of air
ν	Kinematic viscosity of air
ρ	Density of particle
σ_θ	Standard deviation of wind direction
σ_y	Standard deviation of plume concentration

- χ Concentration of contaminant in air
- χ_o Concentration at start of scavenging
- ψ Rainout coefficient (fraction/time)
- ω Deposition rate by washout (mass/area time), or total deposition (mass/area), in accordance with dimensions of Q_o

THE CALCULATION OF PRECIPITATION SCAVENGING

INTRODUCTION

This paper is the attempt to place under one cover our present knowledge of scavenging in the troposphere. Both the scientist entering the field of study and the scientific engineer with a problem of hazards analysis, are currently faced with an arduous literature search with no guarantee that the articles, theories, or numbers they intercept will be accurate, current, or even applicable.

The scavenging process has three major divisions: 1) delivery or transport to the scavenging site, 2) in-cloud scavenging by the cloud elements, herein called rainout and snowout, and 3) below-cloud scavenging by the precipitation (herein called washout). Material from low elevation sources will be transported with the low-elevation winds, diffused upwards by low-level turbulence, and once in the vicinity of clouds may be carried into those clouds by organized vertical motions. When the material is from high elevations (stratosphere), the general circulation of the earth's atmosphere and the exchange mechanism at the tropopause have control of the delivery.

There are several good starting points for the study of scavenging. Junge (1963) and Facy (1962) have looked at the in-cloud scavenging, although with differing conclusions. Fletcher (1962) and Mason (1957) have published texts on precipitation and cloud physics with comprehensive bibliographies. Chamberlain (1953) appears to have attempted the first broad application of scavenging equations, and his approach is still used. Fuchs' (1964) extensive volume on the mechanics of aerosols has been recently revised. Reiter (1964) and Danielsen (1964) will serve as introductions to the controversy on stratosphere-troposphere exchange.

At the time of this writing, there is increasing interest in the scavenging problem, and the reader should be alert for the very probable later measurements of rain spectra, washout coefficients, scavenging efficiencies, and other parameters discussed below.

SPECIFICATION OF THE PROBLEM

Quite obviously, the intensity and location of the clouds and precipitation will determine the potential for scavenging and can usually be stated explicitly. However, fine details of the clouds and precipitation, such as entrainment, electrical charge, and snow crystal shape are also important, but generally impossible to specify.

The nature of the material to be scavenged is fully as important as the nature of the precipitation, and should be specified as to size if particulate, and as to solubility if gaseous. Density is important for at least the larger particles. Characteristics of electrical charge and wettability can be quite important, and these effects are yet under debate and investigation. With increasing time and distance from the source, all of these characteristics are modified by agglomeration, and by attachment and adsorption onto natural aerosols. The material should then assume the nature of this host.

Now the "natural aerosol" is not a well defined parameter either, varying with elevation, geography, and meteorology. In his extensive treatment of natural aerosols, Junge (1963) presents model size distributions which may be used in the absence of local measurement (Figure 1). Although little is known about the density of natural aerosols, Junge concludes from chemical composition that most have densities of 1 to 2 g/cm³.

The scavenging process consists of repeated exposures of particles and gases to cloud or precipitation elements, each time with some chance of collection on the element. The scavenging is consequently an exponential decay process, obeying the equations

$$\chi = \chi_0 \exp(-\psi t) \quad (1)$$

$$\chi = \chi_0 \exp(-\Lambda t), \quad (2)$$

where ψ and Λ are called the rainout and washout coefficients, respectively. Because these coefficients are dependent upon many aerosol and scavenger characteristics, it is easier and generally as accurate to begin the solution of a problem by adjustment or modification of previously measured or predicted coefficients, rather than with the more basic variables.

FIGURE 1

**Model Size Distributions
for Atmospheric Aerosols**

Curve 1, continent. Curve 2, ocean. The part of this curve below 1μ is estimated. Curve 3, sea-spray component of the maritime aerosol. The hatched area between Curves 2 and 3 represents the non-sea-spray component over the ocean. The straight lines indicate the shift of the continental log radius-number distributions curve due to particle growth by humidity for mixed particles of about 20% soluble material (Junge, 1963, Academic Press)

THE THEORY OF WASHOUT BY RAIN

When an obstacle (such as a raindrop) falls, it sweeps out a volume of air. This volume of air must separate to allow the obstacle to pass. Particles therein will tend to follow the air. Due to their inertia, electrical attraction, and to molecular diffusion, however, some fraction of them will cross the streamlines of the air and intersect the obstacle. This fraction is the target or collision efficiency.

Contact with the obstacle does not ensure collection. Some particles may bounce off or be reentrained by the air. Of those making contact, the fraction remaining with the obstacle is the coalescence or retention efficiency.

The product of the target efficiency with the retention efficiency is the collection efficiency, E , of the obstacle for the particles considered. If 50 of the 100 particles in the path of a raindrop contact it, but only 20 are washed out, then the target efficiency is 0.5, the retention efficiency is 0.4, and the collection efficiency is 0.2.

The target efficiency is often presented as a function of the "impaction parameter", which is derived from the equation of motion of a particle with drag resistance. The impaction or inertial parameter is

$$K = \frac{4 a^2 \rho U}{9 \mu D} \quad (3)$$

where a and ρ are the particle radius and density, μ is the dynamic viscosity of the air, and U/D is the ratio of obstacle speed and diameter. In the case of rain, U and D are related, and μ is nearly constant so that, approximately,

$$E = f(a^2 \rho, D). \quad (4)$$

Langmuir and Blodgett (1945) have calculated the target efficiencies for spherical collectors in potential and viscous flow fields, and Langmuir (1949) has provided an interpolation formula between the two flow types. In the past, it has been generally assumed that this interpolation formula was applicable for raindrops. However, later measurements and theoretical predictions (e.g., Percy and Hill, 1957) question this assumption.

Langmuir and Blodgett's target efficiencies were calculated for point-sized particles. They did not allow for collection of a finite-sized particle whose center missed the drop, but whose edge made contact. It may be that if even the edge of some kinds of particles touch the drop, coalescence or collection may result. If so, the Langmuir efficiencies will be low. They may be approximately corrected by assuming a larger rain-drop cross section of $(D + 2a)^2/D^2$; this factor is to be multiplied by the Langmuir point-particle efficiency.

The Langmuir target efficiencies of raindrops at a pressure of 785 millibars collecting particles of unit density appear in Table I. Conversion for particles of another density or at another altitude is made by equating $a^2 \rho$ or K for the two particles, in compliance with equations (3) and (4).

TABLE I
THE COLLISION(TARGET) EFFICIENCY E FOR DROPS OF DIAMETER D
FALLING THROUGH A CLOUD OF SMALLER DROPS OF RADIUS a
(Langmuir, J. Met. 5, 182)

D, mm	a (microns)							
	2	3	4	6	8	10	15	20
0.03	-	-	-	-	0.092	0.269	0.500	0.643
0.05	-	-	-	0.050	0.277	0.411	0.613	0.724
0.08	-	-	-	0.205	0.394	0.510	0.690	0.782
0.14	-	-	0.035	0.340	0.500	0.608	0.750	0.834
0.2	-	-	0.133	0.418	0.564	0.660	0.793	0.862
0.3	-	0.010	0.245	0.498	0.631	0.713	0.829	0.887
0.4	-	0.085	0.326	0.564	0.684	0.756	0.859	0.908
0.6	-	0.213	0.425	0.643	0.749	0.810	0.892	0.929
0.8	0.040	0.303	0.500	0.698	0.793	0.849	0.919	0.950
1.2	0.121	0.355	0.530	0.731	0.827	0.876	0.939	0.963
2.0	0.140	0.358	0.535	0.738	0.834	0.886	0.944	0.966
2.8	0.168	0.360	0.534	0.735	0.840	0.890	0.950	0.970
3.6	0.117	0.288	0.456	0.680	0.800	0.865	0.935	0.965
4.8	0.075	0.220	0.372	0.606	0.743	0.823	0.920	0.950
6.0	0.050	0.170	0.306	0.546	0.690	0.785	0.900	0.940

Das (1950), and Fonda and Herne (Herne, 1960), have considered the finite size of the particles in later calculations of target efficiencies. The interception effect is noticeable for small raindrops and large particles. After adding to the calculations made by Fonda and Herne, Mason (1957) applied the Langmuir interpolation formula. The resulting target efficiencies for unit density particles at 900 millibars appear in Table II. Approximate conversion to other densities can still be made by equating $a^2 \rho$ for the two particles, and multiplying the appropriate tabular efficiency by $(D + 2a_2)^2 / (D + 2a_1)^2$ to allow for the change in interception area of the particle. It may be argued that because of the uncertainties introduced by unknown retention efficiencies, and by Langmuir's interpolation formula, there is little purpose in choosing Mason's efficiencies over Langmuir's, or in considering corrections to these efficiencies for interception areas.

TABLE II
THE COLLISION (TARGET) EFFICIENCIES FOR DROPS OF DIAMETER D
COLLIDING WITH DROPLETS OF RADIUS a AT 0 °C AND 900 millibars
(Mason, Oxford University Press)

D, mm	a (microns)							
	2	3	4	6	8	10	15	20
0.03	0.03	0.05	-	-	-	-	0	-
0.05	0.01	0.02	0.05	0.23	-	-	-	-
0.08	0.004	0.01	0.04	0.29	0.55	-	-	-
0.14	0.02	0.03	0.11	0.40	0.65	0.83	1.12	-
0.2	0.03	0.07	0.18	0.47	0.67	0.83	1.08	1.26
0.3	0.07	0.14	0.28	0.55	0.72	0.84	1.04	1.16
0.4	0.10	0.21	0.35	0.61	0.76	0.86	1.01	1.11
0.6	0.15	0.31	0.45	0.68	0.80	0.89	1.00	1.06
0.8	0.17	0.37	0.51	0.72	0.84	0.91	1.00	1.06
1.2	0.17	0.40	0.54	0.74	0.85	0.91	0.99	1.02
2.0	0.15	0.37	0.52	0.74	0.83	0.90	0.97	1.00
2.8	0.11	0.34	0.49	0.71	0.83	0.89	0.96	0.96
3.6	0.08	0.29	0.45	0.68	0.80	0.86	0.98	0.95
4.8	0.04	0.22	0.39	0.62	0.75	0.83	0.93	0.97
6.0	0.02	0.16	0.33	0.55	0.71	0.81	0.91	0.95

The effect on retention efficiencies of variations in the wettabilities of particles is a very unsettled question about which McDonald (1963) and Berg (1963) have published recent articles. Not only the effects of "partial" wettabilities, but the actual wettability of natural airborne particles are in need of further definition. Even the coalescence of colliding water droplets is not assured. However, in the absence of wettability information, the likely assumption is that of perfect retention or coalescence. This is equivalent to equating target and collection efficiencies.

When air flow patterns other than viscous and potential are considered to exist about falling drops, calculated efficiencies can become very different. Efficiencies greater than 1.0 are predicted (and observed) for spheres with wakes (Pearcey and Hill, 1957). Whereas, Langmuir's interpolation formula predicts a rather flat maximum in E at a drop diameter of 1.2 mm, Kinzer and Cobb and Engelmann have observed a peak in E for drops of about 0.4 mm diam (Figure 2).

$$\rho = 4$$

$$a = \text{radius}$$

$$12 = 4a^2 \quad a^2 = 3$$

$$a = 1.7$$

$$d = 3.4$$

$$d = 3.4 \mu$$

FIGURE 2

Scavenging Efficiencies Versus Drop Diameter and $a^2 \rho$ ($\mu^2 \text{ g cm}^{-3}$), where a and ρ are the radius and density of the particles (after Engelmann, submitted to J. Atmos. Sc., 1965)

When E is known as a function of drop size, the washout coefficient for a particular aerosol size and density can be computed from

$$\Lambda = \int_0^{\infty} F E A dD \quad (5)$$

where A is the cross-sectional area of drops of diameter D , and F is their flux density (drops per area per time per diameter interval). The flux density divided by the terminal velocity, U , yields the "space density," $N(D)$, which is the form most frequently used in presenting measured rain spectra. Equation (5) may, therefore, be written as

$$\Lambda = \int_0^{\infty} N U E A dD. \quad (6)$$

Russian authors call the product, UEA , the "coefficient of capture" of the drop of size D for the particle considered. Table 3 gives the terminal velocity of waterdrops near sea level (Gunn and Kinzer). Because waterdrops greater than 1.2 mm diam are flattened, the diameter of a sphere of the same volume is listed as the "equivalent diameter." In steadystate rain, the flux density is less dependent upon elevation than is the space density.

TABLE III

TERMINAL VELOCITY OF FALL FOR DISTILLED WATER DROPLETS IN STAGNANT AIR
(pressure 760 mm, temperature 20 °C, relative humidity 50%)
(Gunn and Kinzer, J. Met. 6, 246)

Equivalent Drop Diameter, mm	Terminal Velocity, cm/sec	Equivalent Drop Diameter, mm	Terminal Velocity, cm/sec
0.1	27	3.2	826
0.2	72	3.4	844
0.3	117	3.6	860
0.4	162	3.8	872
0.5	206	4.0	883
0.6	247	4.2	892
0.7	287	4.4	898
0.8	327	4.6	903
0.9	367	4.8	907
1.0	403	5.0	909
1.2	464	5.2	912
1.4	517	5.4	914
1.6	565	5.6	916
1.8	609	5.8	917
2.0	649		
2.2	690		
2.4	727		
2.6	757		
2.8	782		
3.0	806		

The washout coefficient for the entire aerosol is calculated by integrating equation (5) over particle size. If the aerosol characteristic of interest is size dependent, this relation can be inserted into the integral. For instance, for radioactivity uniformly distributed throughout the mass of aerosol,

$$\Lambda = \alpha \int_0^{\infty} \int_0^{\infty} F E A M dD dd \quad (7)$$

where α is the activity per mass and M is the mass of the aerosol particle of diameter d .

RAIN SPECTRA

Numerous measurements of rain spectra have been made, and several empirical equations have been fitted to them. While rain spectra usually have peak frequencies between 0.5 and 1.0 mm diam, most empirical equations cannot fit peak frequencies. The distributions of Marshall and Palmer (1948) and Best (1950) have been most widely used, but these give excessive numbers of small droplets.

Levin (1954) has shown that several empirical space density formulae are approximated by portions of a logarithmic-normal distribution. This distribution can fit a peak and can be argued to have a theoretical basis. The additional convenience of logarithmic-normal probability paper makes this an attractive distribution. It approximates, for example, rains sampled by Kelkar (1959) in India, and a shower sampled by Engelmann in the semiarid climate of Sunnyside, Washington (after somewhat arbitrary allowance for evaporation of the falling drops). The latter spectrum, uncorrected for evaporation, is shown in Figure 3, together with selected Kelkar spectra. Sims, et al. (1965), found that spectra from several locations were best fit with logarithmic-normal functions, but the function parameters did not seem to be ordered with rainfall rate.

FIGURE 3

Selected Rain Spectra on Logarithmic-Normal Probability Paper. The dots represent a spectrum from a semiarid region of Washington. The remaining curves were measured by Kelkar (1959) in India.

Users of measured rain spectra should consider the effects of evaporation as the rain falls between cloud and ground. Evaporation reduces the sizes of all drops and the numbers of the small ones. Splash droplets from the ground and equipment around rain samplers, and breakup of drops on the entrance edge of rain samplers will increase the apparent number of small droplets. The use of a calibration curve on one batch of water-sensitive paper that was derived by someone else using another batch of paper seems to be a frequent practice. The errors introduced in spectra by these several effects can be very large, and are frequently not adequately discussed when the spectra are published.

CALCULATED WASHOUT COEFFICIENTS FOR RAIN

Chamberlain (1953) used Langmuir's efficiencies and Best's (1950) fitted spectra to calculate washout coefficients for various rainfall rates and particle (cloud droplets) sizes. These coefficients are given in Figure 4 as a function of $a^2 \rho$.

FIGURE 4

Washout Coefficients for Unit Density Particles Versus Rainfall Rate and $a^2 \rho$ ($\mu^2 \text{ g cm}^{-3}$), where a and ρ are the radius and density of the particles. These calculations are based on Best's (1950) spectra and Langmuir's (1948) target efficiencies (after Chamberlain, 1953).

Similar calculations have been made by May (1958) using the somewhat higher efficiencies of Mason, but these give coefficients only about 10% greater than Chamberlain's. May measured washout coefficients in natural rain using spores ($a^2 \rho = 320$) and found Chamberlain's predictions substantially correct.

A different result is obtained using the efficiencies of Kinzer and Cobb with the Washington and Indian spectra, as shown in Figure 5. The two lines were fitted "by eye" to the coefficients calculated for $a^2 \rho = 42$. A slightly convex line could be used. Only the plotted points for Kinzer and Cobb efficiencies are shown, to prevent confusion; less scatter exists with the calculation using Mason's efficiencies.

FIGURE 5

Washout coefficients for $a^2 \rho = 42$ only, calculated from spectra measured in Washington and India, using theoretical efficiencies from Mason (1957) and measured efficiencies of Kinzer and Cobb (1958).

Even though the Kinzer and Cobb efficiencies are greater than Mason's for the more numerous small drops, the calculated washout coefficients are lower. This is because of the greater contribution to the cross sectional scavenging area made by the larger drops, where Mason's efficiencies are larger than Kinzer and Cobb's. The slope of the lines show that the washout coefficient for large particles varies with about the 0.75 or 0.8 power of the rainfall rate. There is a large amount of scatter within a single shower and between storms, however.

The coefficients calculated from the Langmuir or Mason efficiencies may be regarded as slight overestimates in the absence of significant electrical charges on the rain. Both, the measurement of lower coefficients

(actually collection efficiencies) by Kinzer and Cobb and by Engelmann, and the possibility of retention efficiencies less than one, support this view.

For particles less than 1μ particle diameter, the calculations based only upon inertial effects in idealized flow patterns show negligible washout. However, they cannot be considered at all reliable, owing to the presence of electrical charges (see Byutner and Gisina, 1963). Solutions for particular particle and drop charges have been made by several workers (summarized by Fuchs, 1964), but we have insufficient information to safely extrapolate these predictions to other charge values. In addition, there is insufficient information on the normal charges existent in rain and clouds.

There are also no measurements on natural washout of these smaller particles, although some current research plans do include them.* It is apparent that the problems of small particle size measurement, low collection efficiencies, and nonuniform raindrop charge will make these difficult experiments to perform.

WASHOUT COEFFICIENTS FOR PARTICLES IN SNOW

There are no published washout coefficients, theoretical or measured, for snow. The large numbers of frozen cloud droplets occasionally attached to snowflakes establish that snow is an effective scavenger for at least large particulates. This effectiveness will vary with the type of crystal, its shape, fall speed, and electrical charge.

For the same rate of precipitation, we expect a greater scavenging of smaller particles by the slower "feathery" flake (with its larger area and probable electrostatic charge) than by its equivalent waterdrop. Large numbers of Aitken particles (diameter $<0.02\mu$) have been observed on snowflakes.

* Jan Rosinski. Private Communications, and Georgii and Weber (1964).

Hinzpeter's (1958) data show a lower specific radioactivity for snow water (about 0.4 to 0.6 as great as rain water) at the same daily precipitation rates. However, the snow and rain were sampled at different times of the year, and the air activity has greater variation than this. Georgii and Weber (1964) found snow water to contain ammonia, nitrate, sulfate, and other ions at 1.7 to 2.9 times their concentration in rain water. Reasoning that both precipitation types were originally snow within the cloud, they conclude that snow has a higher washout coefficient than rain for these ions.

However, Facy (1962) suggests that the snowflake is less able to retain its scavenged materials and gases, and will even reject material in its path during evaporation. Hinzpeter's data (1958) show increasing radioactivity in air following snow, but decreasing radioactivity following rain. He interprets this as the release of adsorbed material when the flakes melt and evaporate on the ground.

In the absence of quantitative measurement in snow, the provision of specific washout coefficients for particulates is highly speculative.

WASHOUT COEFFICIENTS FOR GASES IN SNOW

We expect that snow crystals will be effective scavengers of gases due to their large area and small terminal velocity. However, the surface of the flake will reach a saturation point with some gases quicker than the raindrop, which has its entire volume available as a sink. There may well be a difference in absorption and cohesion at ice and water surfaces. Theoretical predictions of gaseous scavenging by snow require knowledge of the electrostatic capacitance of each type of snow crystal and its space density (number per volume of air), neither of which is presently known.

Engelmann and Perkins (1965) have made a few measurements of the washout of inorganic gaseous iodine, as shown in Figure 6. The values suggest that the washout depends upon the type of crystal and its fall velocity, and that, for a given crystal type, the washout varies linearly with precipitation rate. Until further measurements are available, Engelmann and Perkins assume that there is a family of parallel lines, one for each crystal type, with the

position of each line being determined by the crystal's velocity and electrostatic capacitance. The two lines in Figure 6 would bracket most snowfalls.

FIGURE 6

Washout Coefficients of Inorganic Iodine Vapor as a Function of Snowfall Rate for Columnar and Agglomerated Dendritic Crystals (after Engelmann and Perkins, submitted to Nature, 1965)

Washout coefficients for other soluble gases will compare to those for iodine as the ratio of their diffusivities in air. It should be possible to treat trace quantities of partially soluble gases as being fully soluble. However, Engelmann and Perkins found that organic iodine was scavenged with a distinctly lower coefficient than the inorganic.

SCAVENGING OF GASES BY WATERDROPS

Scavenging of gases is predicted using molecular diffusion to the drop or droplet in accordance with the vapour pressures and solubilities of the free and collected gases (Junge, 1963; Chamberlain, 1960; Griffiths, 1963).

For fully soluble gases,

$$\Lambda = \int_0^{\infty} \pi Y \text{ Sh } D N dD \quad (8)$$

where Y is the coefficient of molecular diffusion (or diffusivity) and Sh is the Sherwood (or Nusselt diffusion) number. The Sherwood number for spheres is approximately

$$Sh = 2 + 0.6 Re^{1/2} (\nu/Y)^{1/3} \quad (9)$$

where Re is the Reynold's number and ν is the kinematic viscosity of air. Because Equation (8) is nearly linear in Y , washout coefficients for soluble gases are nearly proportional to their diffusion coefficients in a rain of given flux density. Theoretically, trace quantities of insoluble gases may be treated as soluble in the scavenging process.

Predicted coefficients for gaseous iodine appear in Figure 7. Those predicted from Best's fitted spectra are considerably larger than those from the Washington and Indian spectra. Best's spectra are unrealistic in the small drop region, and although the Washington spectra may have a minimum number of small drops because of evaporation, the latter curve is recommended. Effects from the spectral difference show up more in gaseous scavenging than in the particulate case.

FIGURE 7

Predicted Washout Coefficients in Rain for I_2 , Using Best's (1950) Fitted Spectra, and Spectra from Washington and India

Recent laboratory work (Booker, 1965; Friedman, Machta, and Soller, 1962) confirms theoretical predictions of the exchange of deuteriated and tritiated water vapor with monodisperse waterdrops in saturated air.

RAINOUT (IN-CLOUD SCAVENGING)

Collection due to inertia becomes insignificant with small droplets in viscous flow and with very small particles. It is then necessary to use electrical effects, nucleation, diffusiophoresis, Brownian motion, and velocity gradients to explain rainout. These effects are also small, and calculations of rainout, such as by Greenfield (1957), Junge (1963), and Chamberlain (1960), show long residence times for aerosols in nonfalling clouds. This is especially true in the aerosol range of 0.1 to 0.4 μ , for which there seems to be no adequate scavenging mechanism.

However, cloud elements do fall, and it may be that the inertia of a fine particle is sufficient to carry it into the boundary layer of the falling cloud element where weak forces have opportunity to complete the collection. Facy considers the value of E in clouds to be at least 1.0. This yields high values for the rainout coefficient in the equations

$$\psi = \int_0^{\infty} F E A dD, \quad (10)$$

$$\chi = \chi_0 \exp (-\psi t). \quad (1)$$

Byutner and Gisina (1963) have calculated rainout coefficients in clouds composed of 100 drops/cm³, 10 μ in diameter. They assumed charges of 5×10^{-7} on the cloud droplets and 4.8×10^{-10} esu cgs on ash particles, and calculated separately coefficients for the several effects operating. The coefficients cannot be added together to obtain a total coefficient, but their values indicate that a minimum total rainout coefficient of about $3 \times 10^{-5} \text{ sec}^{-1}$ occurs near the 0.2 μ particle diameter. This corresponds to an efficiency of 0.1, which is less than 1.0, but quite significant. Smaller cloud drops will have higher efficiencies, but may have lower flux densities.

Junge introduces the equation

$$k_1 = \chi_o \epsilon \rho / L \quad (11)$$

where

k_1 = aerosol per volume of cloud water

χ_o = aerosol per volume of unwashed air

ρ = density of water

L = mass of condensed water per cloud space volume, and

ϵ = "rainout efficiency, " or $1 - (\chi/\chi_o)$.

In Equation (1), t is the time the air parcel is within the cloud. Using this equation, the cloud air would eventually be cleansed, and ϵ would approach 1.0. However, since most clouds entrain new air continuously, the aerosol contained in the cloud water increases as long as solubility allows and may be better represented by

$$k_1 = \chi_o \psi T \rho / L, \quad (12)$$

where T , the age of the cloud, is different from t .

The parameter (ψT) at the onset of precipitation is quite possibly a characteristic of the cloud type, since the cloud elements also grow by aerosol collection processes. Both ϵ and the parameter (ψT) may be estimated from measurements of χ_o/k_1 and estimates of L . The proper choice for L is debatable since it varies greatly with cloud type and precipitation amount, but reasonable values can be chosen which will keep (ψT) or ϵ in the range 0.5 to 1.0. Table 4 lists such values.

The value of k_1 must be adjusted to allow for evaporation of the precipitation element as it falls. Aerosol scavenged or lost in the air below the cloud must also be added to k_1 . These adjustments are implicit in the data of Small (1960) and Hinzpeter (1958) since their measurements were taken from air near the ground.

Because of the complexity of in-cloud scavenging, it would seem that particulate scavenging by rain and snow cloud elements are presently best estimated from data of the type given in Table 4. However, the near certainty that

TABLE IV

SELECTED MEASUREMENTS OF SURFACE AIR
TO PRECIPITATION ACTIVITY RATIOS, AND VALUES OF CLOUD WATER
CONCENTRATION CHOSEN TO PRODUCE RAINOUT EFFICIENCIES BETWEEN 0.5 AND 1.0

Data Source	χ_o / k_1	k_1 / χ_o	L/ρ	ε or ψT
Small (Norway)				
Minimum	0.25×10^{-6}	4.0×10^6	0.25×10^{-6}	1.0
Maximum	2.1	0.48	2.0	0.98
Mean	0.9	1.1	0.9	1.0
Hinzpeter (Germany)				
Rain 0.1 mm/day	0.8	1.25	0.5	0.62
1.0	1.4	0.71	1.0	0.71
10.0	2.5	0.40	2.0	0.8
Snow 0.15	0.9	1.1	0.5	0.55
1.0	1.6	0.62	1.0	0.62
10.0	3.4	0.29	2.0	0.59

χ_o varies with elevation must be considered when using such data. As yet, little is known about the height variation of entrainment of outside air into convective clouds (aufm Kampe and Weickmann, 1957) and, also, the height of the cloud water which contributes to the precipitation is uncertain.

TREATMENT OF PRECIPITATION CLIMATOLOGY

The preceding sections have provided the means of predicting scavenging when the precipitation characteristics are specified. We have considerable latitude when applying these techniques to uncertain future precipitation, and how we do this will depend upon the particular problem at hand. We will now discuss in general terms the introduction of precipitation climatology into the applied problem.

A wide variety of prediction forms may be made, according to the forms of the precipitation climatology and the contamination description. For instance, one may obtain:

- Total washout - area isopleths
- Maximum possible washout, total or rate
- Return periods for washout rates
- Joint probabilities of accidental release and specified scavenging rates
- Mean yearly and seasonal washout and rainout for continuous releases

Unfortunately, the accuracy of the estimated washout coefficient will usually be so poor as to allow the assumption that Λ is linearly related to the precipitation rate. In this case, the washout can be estimated from the total depth of rain or snow. Area-depth relationships (e.g., Court, 1961) can be superimposed on the predicted contaminant distribution to obtain total washout-area isopleths.

The reader is by no means restricted to considering area isopleths. Often a problem consists of determining whether some total deposition can be exceeded, and for this the centerline or peak exposure, and the expected total depth of precipitation is sufficient.

Special consideration must be given to the climatology of plume widths, turbulence, and wind speeds during precipitation. At many locations, the wind direction during precipitation differs noticeably from the mean direction for all weather types. The different region likely to be affected may be very significant in hazards appraisals. This is especially noticeable with winds above the boundary layer.

In the special case of horizontally uniform precipitation over a continuous plume, the washout area may be obtained mathematically (USWB, 1955). Culkowski (1963) and Guthrie and Nichols (1964) have calculated the maximum washout possible at one particular distance with a rain which starts as soon as (or earlier than) the release. This washout rate for the Gaussian plume model is

$$w_{\max} = \frac{Q_o}{\exp \sigma_y \sqrt{2\pi}} \exp \left(-y^2 / 2 \sigma_y^2 \right) \quad (13)$$

where w is the maximum washout rate ($\text{ML}^{-2} \text{T}^{-1}$) for a source strength Q_o (MT^{-1}) or total washout (ML^{-2}) for Q_o (M).

When calculating maximum possible washout, it must be remembered that the length of release and the duration of washout are often not the same. The washout rates at some distance from a source are generally greater when there is little or no intermediate precipitation.

The washout rate for precipitation which is just beginning is

$$\omega_o = \Lambda \int_0^z \chi(x, y, z, Q_o) dz. \quad (14)$$

The upper limit for this integral is the height from which the rain falls. For the Gaussian plume model, Equation (14) becomes

$$\omega_o = \frac{\Lambda Q_o}{\bar{u} \sigma_y \sqrt{2\pi}} \exp(-y^2/2\sigma_y^2). \quad (15)$$

Equation (14) assumes that the plume does not lean with elevation because of wind direction shear.

If the rain cloud is stationary while the contaminant is moving, Equation (15) will continue to apply on the upstream side of the rain cloud. Because of this, the continuous introduction of new material to an orographic precipitation cloud may result in washout rates which do not decrease in time. In this way, Fuquay (1963) has explained greater radioactivity in the snow found on western mountain slopes.

Normally, χ must be modified to account for previous scavenging upstream from x . This scavenging can easily reach 50% in 10 mi of rainfall. For instance, if the precipitation extends upstream to the source, Equation (15) is modified to read

$$\omega = \frac{\Lambda Q_o}{\bar{u} \sigma_y \sqrt{2\pi}} \exp(-y^2/2\sigma_y^2) \exp(-\Lambda x/\bar{u}), \quad (16)$$

from which Equation (13) was derived.

In Equation (16), the two washout coefficients are not necessarily the same. The first applies to the current precipitation, and the second to the earlier precipitation extending back to the source. The area within a washout isopleth, according to (16), has been obtained by numerical integration, and these results are presented in Figures 8, 9, and 10, for three values of the stability parameter, $\sigma_\theta \bar{u}$, as a function of both the current and "upstream" washout coefficients. $(\sigma_\theta \bar{u})$ is the product of the mean wind

FIGURE 8

Area Within a Current Washout Isopleth for Various Earlier Washout Rates. Based on a Gaussian plume model and plume variance as a function of $\sigma_{\theta} \bar{u}$.

FIGURE 9

Area Within a Current Washout Isopleth for Various Earlier Washout Rates. Based on a Gaussian plume model and plume variance as a function of $\sigma_{\theta} \bar{u}$.

FIGURE 10

Area Within a Current Washout Isopleth for Various Earlier Washout Rates.
Based on a Gaussian plume model and plume variance as a function of $\sigma_\theta \bar{u}$.

speed and the standard deviation of the wind direction at plume height, in radians meters/seconds, as defined by Fuquay, Simpson, and Hinds (1964).

As an example in the use of these figures, suppose that it had been snowing lightly for some time, but that the snow had now intensified considerably. The reader wishes to know the area within a particular washout isopleth for the current stability and precipitation. He chooses the figure appropriate to the existing value of $\sigma_\theta \bar{u}$, then on the left side of the graph finds the value of w/Q of interest divided by the current washout, $\Lambda/\bar{u}$. He then moves his pointer over to the curve labelled with the earlier washout, then down to the scale marked area. He multiplies this by $\bar{u}$ to obtain the area of the washout isopleth in square meters.

As a matter of interest, the calculations have also been made for the plume described by

$$\sigma_y = 0.2 x^{0.8} \quad (17)$$

(where σ_y and x are in meters) and are shown in Figure 11. Equation (17) is a kind of composite of experimental data for 30 min long releases at Hanford, Washington.

Comparison of the figures will show that the area is not only quite dependent upon the earlier washout coefficient, but also upon the stability parameter. Some study of the climatology of $\sigma_\theta \bar{u}$ versus $\Lambda/\bar{u}$ should therefore be made before the figures are used in hazards appraisals.

SUMMARY AND CONCLUSIONS

This paper contains the basic equations, the measured and predicted parameters, and the bibliography pertinent to the calculation of precipitation scavenging.

Chamberlain's predicted washout coefficients in rain for particles larger than one micron diameter (Figure 4) are considered to be slight overestimates. In the case of particles smaller than one micron, scientists generally agree that washout by rain is insignificant. The theory cannot be

FIGURE 11

Area Within a Current Washout Isopleth for Various Earlier Washout Rates.
Based on a Gaussian plume model and a plume standard deviation as a
function of distance.

considered reliable for smaller particles, however, due to uncertain electrical effects, and quantitative measurements of this washout are not yet available.

The washout of gases can be predicted using the theory of molecular diffusion to waterdrops. Newly calculated washout coefficients, based on measured rain spectra rather than on fitted spectral equations, are presented and recommended (Figure 5). The prediction of washout of gases by snow must rely upon a very few measurements for iodine gas (Figure 6), there being no theoretical predictions.

Snow apparently has about the same to twice the effectiveness as rain at the same precipitation rate for both washout and rainout of particles, but definitive measurements are greatly needed.

Research is also still inadequate on many of the complex in-cloud scavenging processes. Formal mathematical predictions of rainout due to various mechanisms are interesting, but not decisive. It is recommended that simultaneous measurements of contaminant in air and precipitation (e. g., Table IV) be used for prediction.

The manner in which the washout coefficient, Λ , and the surface air to precipitation activity ratio, χ_o/k_1 , vary with storm and cloud type is unknown. Therefore, recommended values of Λ and χ_o/k_1 may be directly combined with precipitation rates to predict scavenging rates.

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