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WASHOUT COEFFICIENTS FOR POLYDISPERSE AEROSOLS

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The concept of the washout coefficient has proved to be useful in describing the washout properties of irreversibly scavenged atmospheric pollutants. In approximate terms, the washout coefficient can be described as the fraction of the amount of pollutant removed per unit time by precipitation. The key word in this rather loose definition of course is "amount;" it is important that the "amount" or some size-dependent property of the pollutant in question be specified, especially when polydisperse aerosols are delt with. One should discriminate between *number* of particles removed per unit time, *length* of particles removed per unit time, *area* of particles removed per unit time, or *mass* of particles removed per unit time. Indeed, for some noxious pollutants one must consider that the amount of pollutant removal may not depend in any simple way on the length scale of each particle. For monodisperse aerosols, there is no ambiguity, and in fact, the extent of most theoretical developments regarding the washout of aerosols concern only monodisperse aerosols. But in nature, and most often in a consideration of artifically produced pollutant and tracer aerosols, it is necessary to deal with polydisperse aerosols; the very nature of the washout process -- if it is particle size dependent -- results in different interpretations of the washout coefficient as one deals with particle number, area, etc. In order to interpret experimental results properly, and also to make valid theoretical predictions (for environmental impact analyses, say), it is necessary to account for the polydispersity of aerosols and their size-dependent properties.

The specific problem we approach here is the gap between theoretical expectations and experimentally measured washout properties of polydisperse aerosols. In the field, measuring washout coefficients, and in the laboratory, measuring collection efficiencies (say), the result is derived from a measurement of a property of the aerosol (usually total mass removed or rainwater concentration) which is the culmination of an integral process of washout of a particle size distribution by a hydrometeor distribution. Since in its usually-arrived-at form, particle washout theory involves monodisperse aerosols -- i.e., the theoretical washout coefficient is a differential quantity -- it is difficult to relate experimental observations to theory. The objective of this paper is merely to extend the usual theoretical rain washout coefficient concept to account for the polydispersity of aerosols. There will be no new theory considered; we have attempted to utilize current state-of-the-art collection efficiencies and calculate washout coefficients (with some simplifying assumptions) for some reasonable particle and raindrop size spectra.

It is useful to begin with a basic definition of washout coefficient. If the rate of removal of the pollutant from the gas-phase by washout is k (amount*/volume of air·time), and the local gas-phase concentration is χ (amount/volume of air), the washout coefficient is

$$\Lambda \equiv \frac{k}{\chi} \cdot \qquad (\text{time})^{-1} \tag{1}$$

Considerable confusion can result from definitions of the washout coefficients in terms of measurable quantities, which invariably require some simplifying assumptions (e.g., steady state plume, vertical rainfall, etc.); we will not deal with those but propose Eq. (1) as the basic definition for theoretical purposes. If we consider monodisperse particles washed out by a raindrop size distribution, a simple geometrical argument leads to the familiar form

$$\Lambda(a,R_s) = \int_0^\infty \pi R^2 \; E(a,R) \; F(R) \; dR \tag{2}$$

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* number, length, area, mass, radioactivity, etc.

where a is the particle radius

E(a,R) is the collection efficiency, and

F(R) dR is the flux (number/unit area • time) of raindrops with radii R to R+dR.

In Eq. (2) and succeeding developments, the subscript s refers to "the spectrum of," that is, $\Lambda\ (a,R_s)$ means the washout coefficient for particle radius a acted on by the raindrop spectrum defined by F(R) dR.

The next step of this process is to derive the integral washout coefficient which results from the polydispersity of both the raindrop and particle distributions. Thus, Eq. (2) must be integrated over the particle distribution:

$$\Lambda\ (a_s, R_s) = \int_0^\infty \Lambda(a,R_s)\ f_n(a)\ da\ . \qquad (3)$$

Here, $f_n(a)$ da is the probability density function (pdf) characterizing what we will call here the <u>order</u> of the aerosol washout. For the <u>number</u> pdf, n = 0, for <u>length</u> pdf, n = 2, etc. These $f_n(a)$ da are related to the number pdf by

$$f_n\ (a)\ da = \frac{a^n f_0(a)\ da}{\int_0^\infty a^n\ f_0(a)\ da}\ . \qquad (4)$$

COLLECTION EFFICIENCIES

In order to calculate washout coefficients for a broad range of particle size, and in response to a judgment regarding the most important microphysical processes involved in washout, we have chosen three forms of E(a,R) to cover the particle size range .001<a<10μ. These are listed separately in Table 1 and are plotted as a function of a for two values of R in Figure 1.

The Brownian diffusion estimate is one suggested by Slinn[1]. The Brownian diffusion coefficient D for particles was approximated by a power-law

TABLE 1. Collection Efficiencies E(a,R)

(a in μ; R in cm)

DIFFUSION[1]	$\frac{\alpha}{a^2R^2} + \frac{\beta}{a^{\frac{4}{3}} R}$	$\alpha = 0.65 \times 10^{-11}$ $\beta = 0.14 \times 10^{-6}$
IMPACTION[2]	$(3 \times 10^{-4}) \frac{a}{R}$	
INERTIAL[3]	$\left(\frac{S - \frac{1}{12}}{S + \frac{7}{12}}\right)^{\frac{3}{2}}$	S = Stokes Parameter $\simeq .1038\ a^2\ \rho_p$

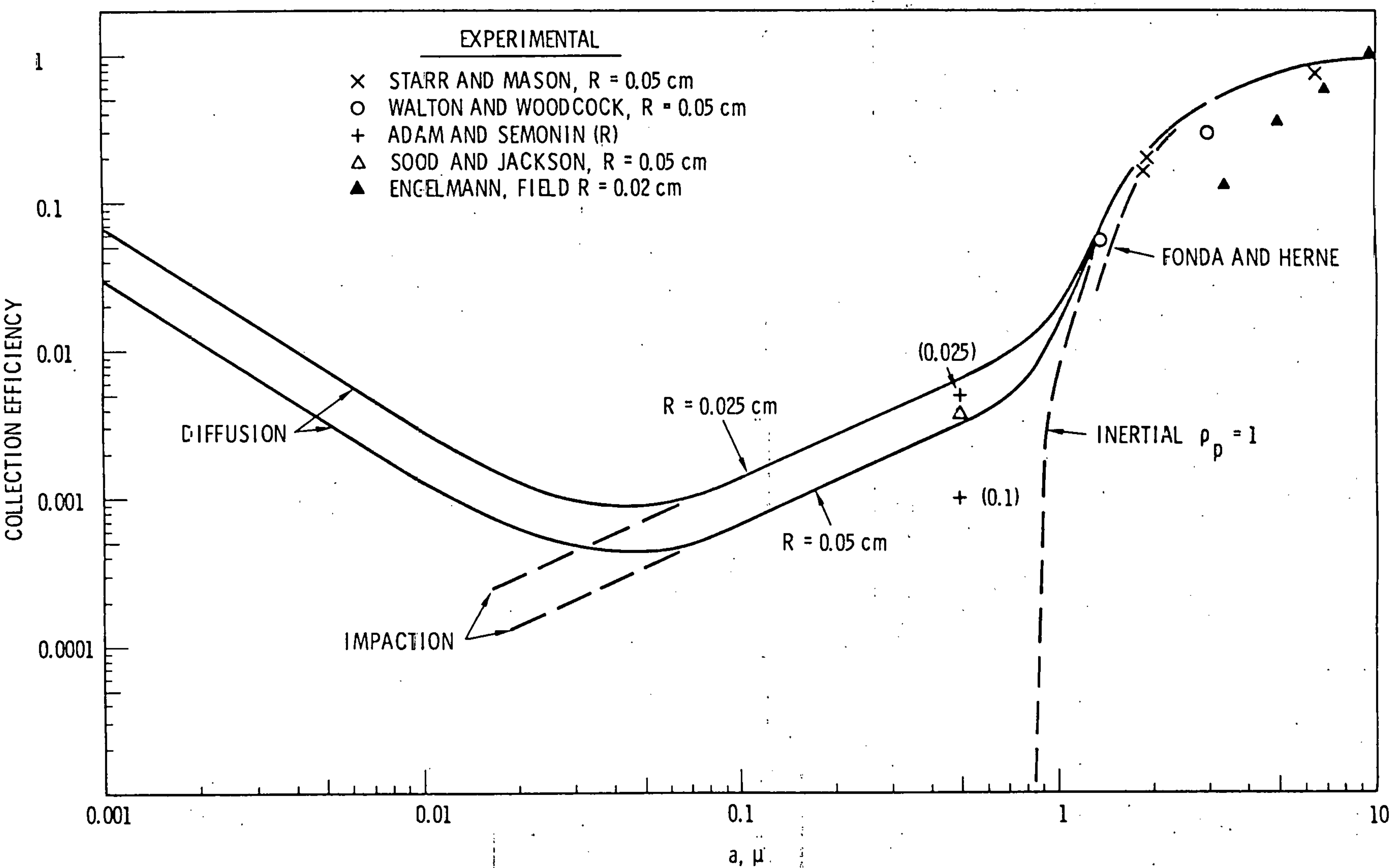

FIGURE 1. Theoretical collection efficiencies (assuming retention) used in the calculations, and selected experimental results.

$$D \simeq 1.3 \times 10^{-8}\ a^{-2} \tag{5}$$

(D in cm^2/sec, a in μ)

which is within 10% of the real value[2] for $a \lesssim .05\mu$. The impaction time is that of Fuchs[2] for the case of inertialess motion in potential flow. The internal collection efficiency is a semi-empirical expression[3] fitted to experimental results of Walton and Woodcock[4] and numerical results of Fonda and Herne[5]. The inertial collection efficiency is a function of the dimensionless Stokes parameter

$$S = \frac{2}{9}\,\frac{a^2 \rho_p}{R\,\rho_a}\,\frac{V_t}{\nu} \tag{6}$$

where ρ_p and ρ_a are the mass density of the particles and air, respectively, V_t is the raindrop terminal velocity and ν is the kinematic viscosity of air.

Since in the size range of most raindrops ($0.01 \lesssim R \lesssim 0.1$ cm) the terminal velocity is approximately proportional[6] to R, the Stokes parameter reduces for present purposes to approximately

$$S = 0.10\ a^2 \rho_p \quad , \tag{7}$$

(a in μ, ρ_p in g/cm^3). This assumption appears to be adequate for low-intensity frontal raindrops.

Also shown in Figure 1 are selected results of experimental measurements.[4,7-9] The measured values indicated are only those found in the literature which represent experiments involving reasonably monodisperse particles (adjusted to unit density) and realistic raindrop sizes falling at terminal velocity. The dashed curve represents the numerical results of Fonda and Herne.[5]

SAMPLE CALCULATIONS

The calculation of washout coefficients using Eqs. (2) and (3) and the collection efficiencies of Table 1 were performed utilizing the lognormal dis-

distribution of x with geometric mean x_g and geometric standard deviation μ is

$$f_o(x)dx = \frac{1}{(2\pi)^{1/2}\ln \mu} \exp\left(\frac{-(\ln x - \ln x_g)^2}{2(\ln \mu)^2}\right) d \ln x \qquad (8)$$

For the raindrop spectrum, the geometric mean radius is here called R_g and the geometric standard deviation Σ_g; for particles, the corresponding parameters are a_g and σ_g.

A further assumption was made that the raindrop spectrum is invariant with rainfall rate J; i.e., only the flux of raindrops varies with J. This appears to be a valid assumption when $J \lesssim 5$ mm/hr, the basis for this being our experience with Pacific Coast frontal rainfall.[10] Explicitly,

$$J = \frac{4}{3}\pi \int_o^\infty R^3\ F(R)\ dR$$

$$= \frac{4}{3}\pi\ F_o \int_o^\infty R^3\ f_o(R)\ dR, \qquad (9)$$

where F_o is the total flux of rainfall and $f_o(R)$ is the (lognormal) number pdf for the rain distribution. Thus Eq. (2) takes the fomm

$$\Lambda(a,R_s) = \frac{3J}{4} \frac{\int_o^\infty R^2\ E(a,R)\ f_o(R)\ dR}{\int_o^\infty R^3\ f_o(R)\ dR} \qquad (10)$$

Evaluation of Eq. (10) (and subsequently, Eq. (3)), involves essentially the calculation of moments of the lognormal distribution. These have been integrated[11], and are given by

$$M_i^i = \int_o^\infty x^i\ f_o(x)dx = x_g^i \exp\left(\frac{i^2}{2} (\ln \mu)^2\right) \qquad (11)$$

(In the following discussion, M_i^i will refer to the raindrop spectrum, and m_i^i to the particle spectrum.) The resulting expressions for $\Lambda(a_s,R_s)$ (where the same integration process is applied to the particle size distribution) are listed in Tables 2 and 3. The inertial $\Lambda(a_s,R_s)$, not being integrable analytically, must be calculated numerically by computer.

Sample calculations were performed using a representative frontal rain spectrum (R_g = .02 cm, Σ_g = 1.86) and numerous particle size spectra. The results for the case of n = 3 (or mass washout) are shown in Figure 2, where the curve for σ_g = 1 is the monodisperse case and the other curves show the integral washout coefficient (normalized to unit rainfall rate) for polydisperse distributions of given "spread" σ_g. The curves clearly show that as the distribution becomes broader (σ_g increases), the effect of the presence of larger particles -- more effectively washed out in the inertial size range -- affects the washout coefficient for the entire distribution.

It should be noted that the abscissa of Figure 2 is geometric mean radius a_g, which proves to be a poor choice as a size parameter with which to characterize a particle size distribution for washout purposes. The figure can be used to estimate the error involved in characterizing a particle size distribution by some other size parameter. For example, the geometric mean particle radius of nth order[11] may be computed from

$$a_{ng} = a_g \exp\left(n[\ln \sigma_g]^2\right) \qquad (12)$$

The washout coefficient for a spectrum of given σ_g and a_g may be seen (for a_g = 0.1 μ, and σ_g = 2.0, Λ/J = 0.02 mm^{-1}) and this may be compared with the corresponding washout coefficient from the monodisperse curve for a_{3g}, say (for the above example a_{3g} = 0.42 μ; the monodisperse Λ/J for which is 0.0055 mm^{-1}).

TABLE 2. Monodisperse Washout Coefficients $\Lambda(a,R_s)$

DIFFUSION	$\left(\frac{3J}{4M_3^3}\right)\left(\frac{\alpha}{a^2} + \frac{\beta}{a^{\frac{4}{3}}} M_1^1\right)$
IMPACTION	$\left(\frac{3J}{4M_3^3}\right) (3 \times 10^{-4})\ a\ M_1^1$
INERTIAL	$\left(\frac{3J}{4M_3^3}\right) M_2^2 \left(\frac{S - \frac{1}{12}}{S + \frac{7}{12}}\right)^{\frac{3}{2}}$

M_i^i = ith moment of raindrop size distribution $(cm)^i$

TABLE 3. Polydisperse Washout Coefficients $\Lambda(a_s, R_s)$

(a in μ; R in cm)

DIFFUSION	$\left(\frac{3J}{4M_3^3}\right) \frac{1}{m_i^i} \left[\alpha\, a_g^{i-2} \exp\left\{\frac{(i-2)^2}{2} (\ln \sigma_g)^2\right\} + \beta\, M_1^1\, a_g^{\left(i - \frac{4}{3}\right)} \exp\left\{\frac{\left(i - \frac{4}{3}\right)^2}{2} (\ln \sigma_g)^2\right\}\right]$
IMPACTION	$\left(\frac{3J}{4M_3^3}\right) \frac{M_1^1}{m_i^i} \left[(3 \times 10^{-4})\, a_g^{i+1} \exp\left\{\frac{(i+1)^2}{2} (\ln \sigma_g)^2\right\}\right]$
INERTIAL	$\left(\frac{3J}{4M_3^3}\right) \frac{M_2^2}{m_i^i} \int_o^\infty \left[\frac{S - \frac{1}{12}}{S + \frac{7}{12}}\right]^{\frac{3}{4}} f_o(a)\, a^i\, da$

$f_o(a)\, da$ = number pdf of particle spectrum

m_i^i = ith moment of particle spectrum, $(\mu)^i$

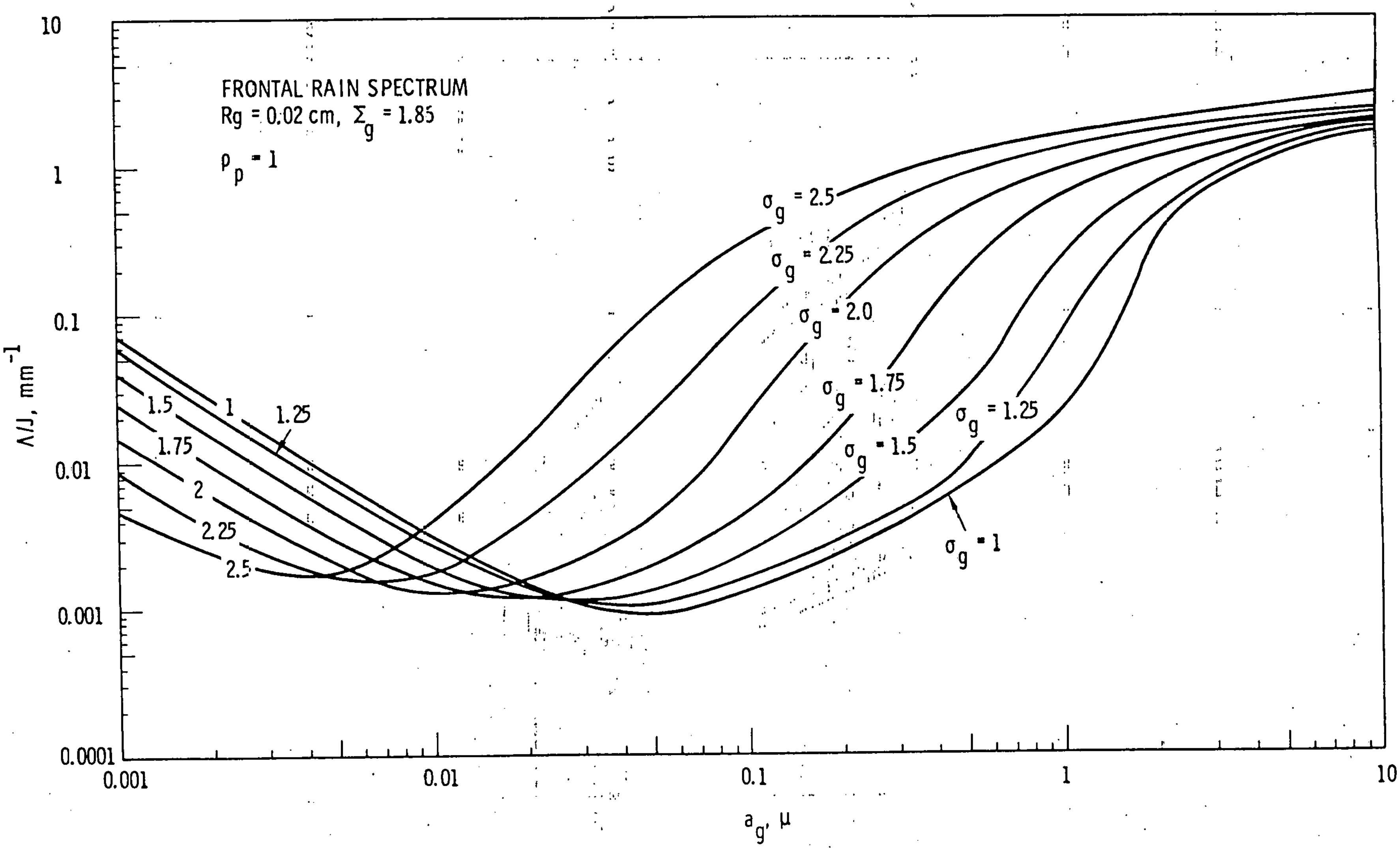

FIGURE 2. Theoretical mass washout coefficiants for unit-density particles, normalized to unit rainfall rate. Collection efficiencies from Figure 1, and a typical frontal rain size spectrum employed.

Thus, in this example it turns out that using the geometric mass mean radius to characterize the particle spectrum resulted in an error of some 73% in the expected value of the theoretical washout coefficient.

When a different rain spectrum is used in the calculations, similar curves result, except that in the region of $a<\sim 1\mu$, the washout coefficient is approximately proportional to $(M_1^1)^{-1}$. This is shown in Figure 3, which is the same as Figure 2 except that a convective shower spectrum (derived from rain spectra measured in the Caibbean Sea[12]) was used. Of course, since we have eliminated the R-dependence from the inertial term, the inertial effect is unchanged, and is overestimated here.

The effect of varying the order n of the washout is demonstrated in Figure 4, which is similar to Figure 2, except that, along with the monodisperse case (not dependent on n), there are curves with fixed $\sigma_g = 2$ for n = 0 to n = 3. (The latter curve is identical to the $\sigma_g = 2$ curve of Figure 2. It is interesting to note that even when washout of number of particles is examined, the polydispersity is effective, because the a-dependency of E is still involved in the integration.

Finally, Figure 5 shows the theoretical result when a very dense aerosol is considered. For such massive particles, it appears useful to think of the washout as uniformly high for all a, without much regard for the character of the size distribution.

DISCUSSION

Clearly, the integral washout coefficient of any order for polydisperse aerosols depends upon the characteristics of the particle size spectrum, as well as the raindrop spectrum and the collection efficiencies. The collection efficiencies, which are the basic element of the theory of aerosol washout,

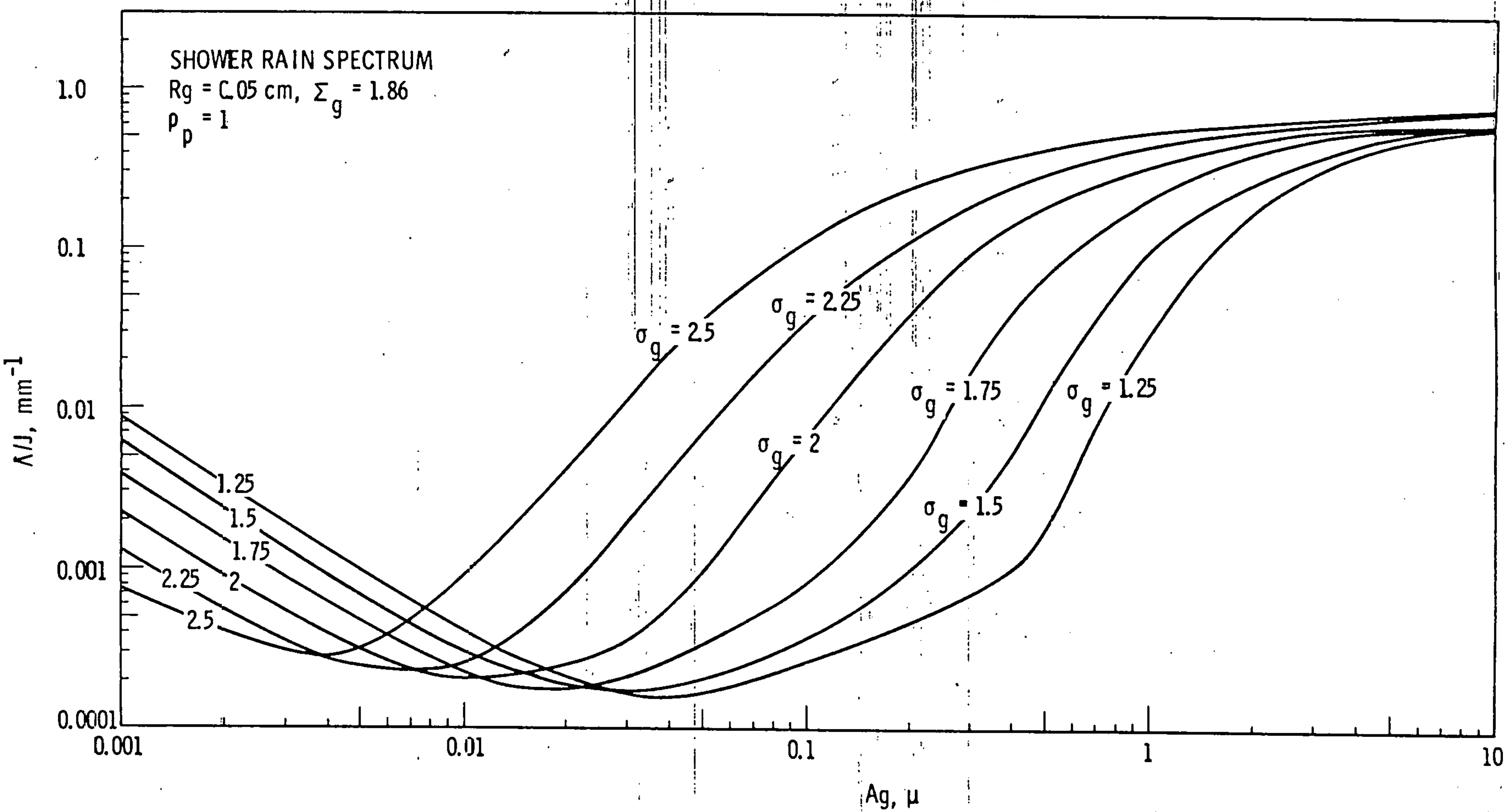

FIGURE 3. Same as Figure 2, except for shower rainfall.

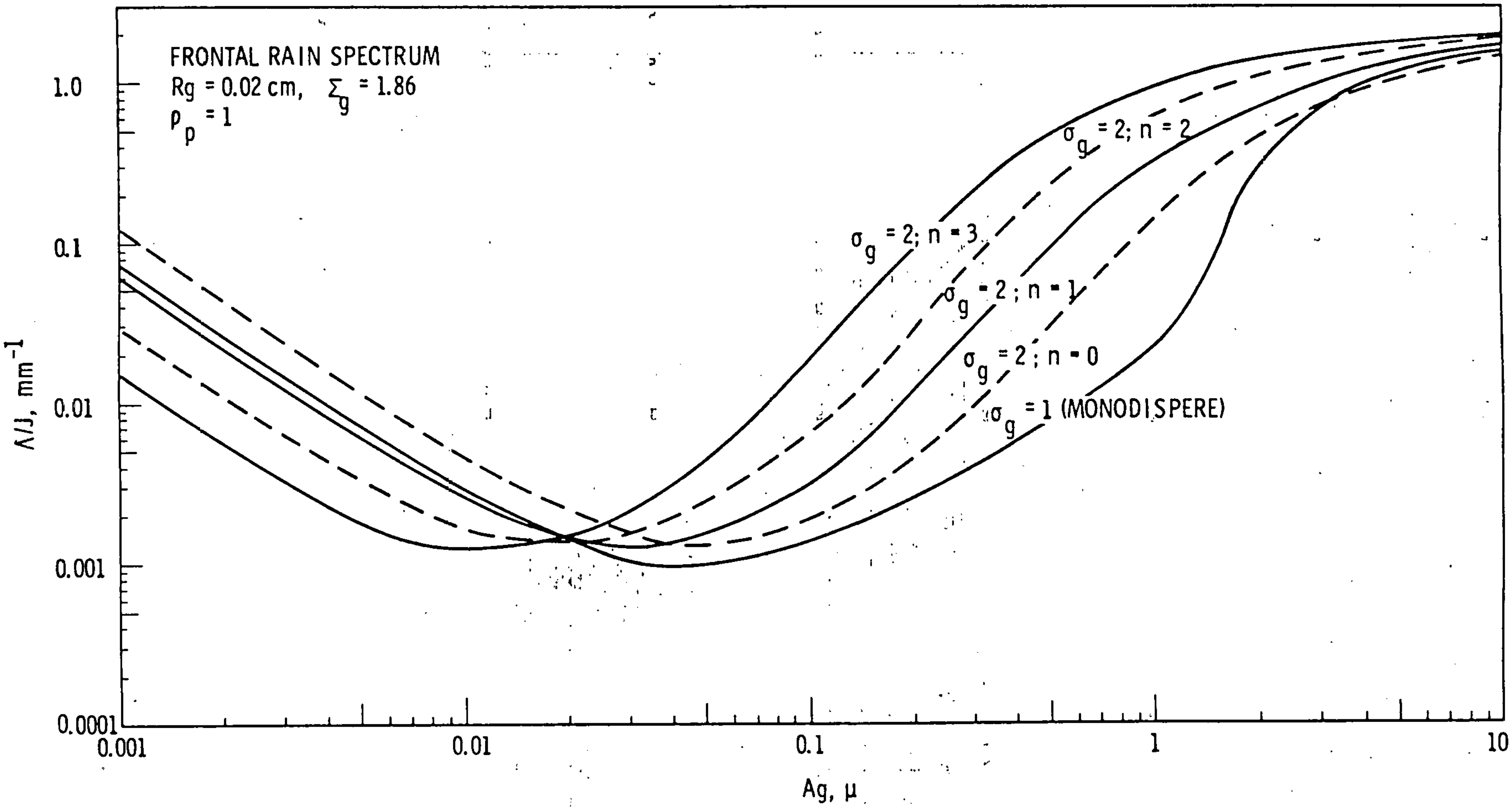

FIGURE 4. Same as Figure 2, except that $\sigma_g = 2$, and the order of the washout, n, is varied.

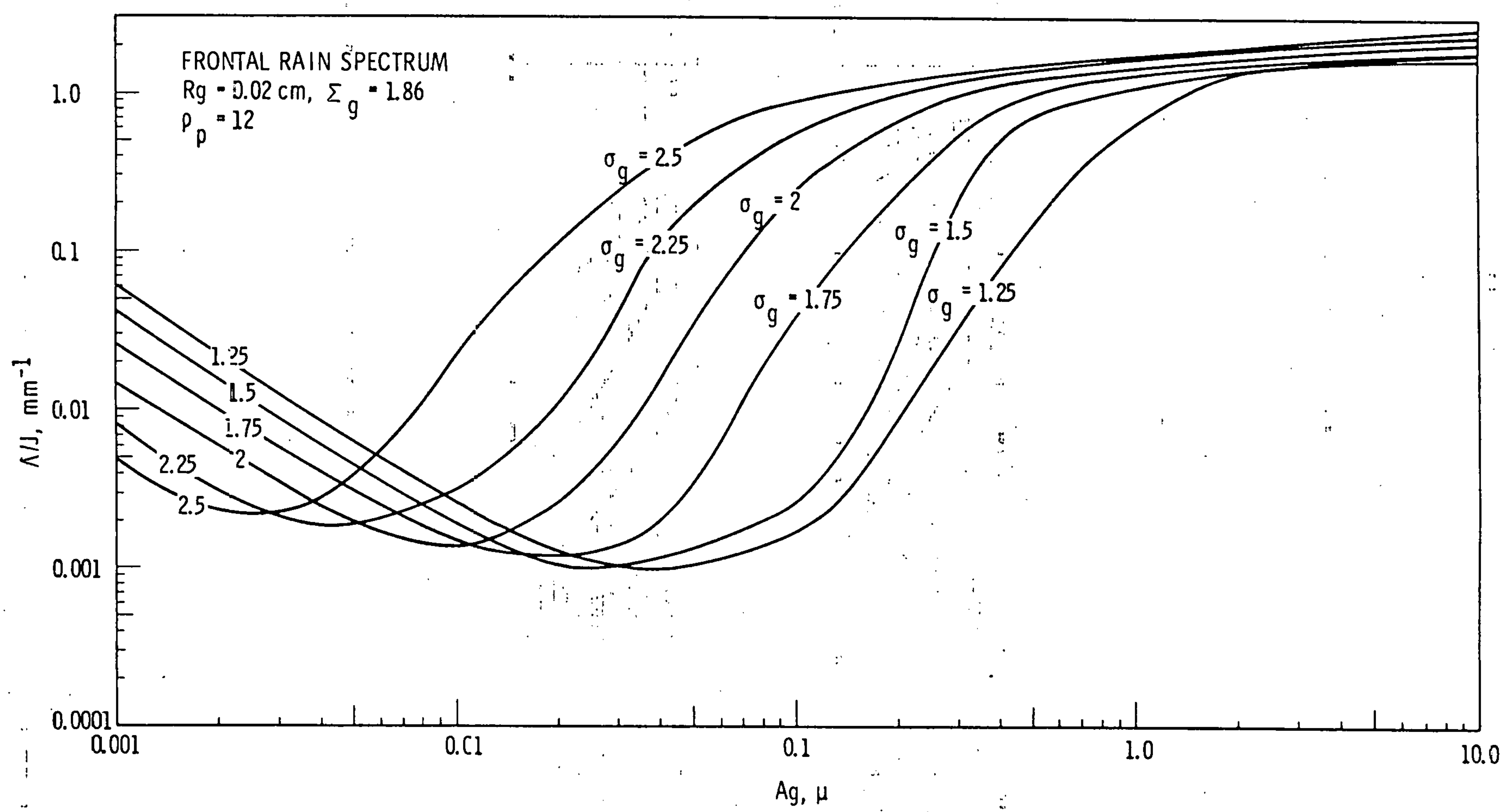

FIGURE 5. Same as Figure 2, except that $\rho_p = 12$.

were assumed to be of a certain mathematical form in this analysis. Inasmuch as the washout coefficient is a result of integration of collection efficiencies over two spectra, one must know the particle and raindrop size dependency of the collection efficiencies in order to calculate the washout coefficient (or rainwater concentration) to gain information about collection efficiencies requires very precise knowledge of the characteristics of the particle and rain spectra. This kind of experimental knowledge is very hard to obtain, at least in the field, and probably in most laboratory experiments. Thus, it appears very difficult, if not practically impossible, to use experimental data to test particle washout theory.

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