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A literature review on wet deposition

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A LITERATURE REVIEW ON WET DEPOSITION

Frits Heikel Vinther and Ole John Nielsen

Abstract. The literature on wet deposition or precipitation scavenging have been reviewed with special reference to predicting the radiological consequences of accidental contamination. The work was part of the EEC Radiation Protection Programme and done under a subcontract with Association Euratom-C.E.A. No. SC-014-BIAF-423-DK(SD).

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1. INTRODUCTION AND DEFINITIONS

Wet deposition or more precisely precipitation scavenging is defined as the removal of any material from the atmosphere to the earth's surface by various types of precipitation mechanisms: liquid or frozen atmospheric water called hydrometeorites, with a gravitational terminal velocity of about 10 cm/s. The words "material" and "matter" denote both particulates and gases. "Washout" is often used as a short form for precepitation scavenging.

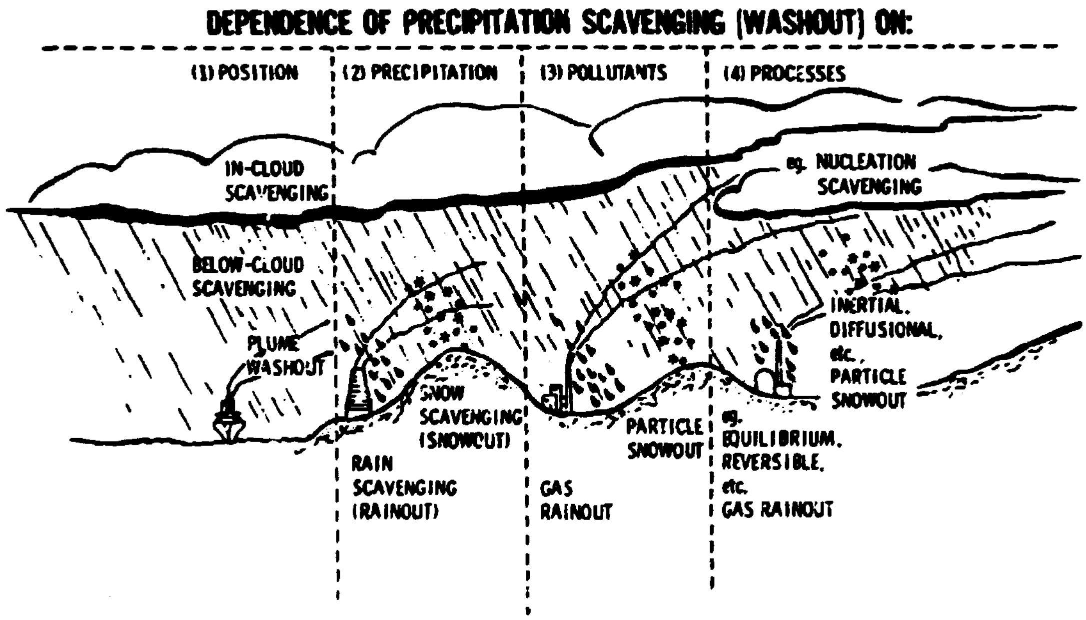

Fig. 1. A schematic illustration of some basic concepts in precipitation scavenging. After Slinn.

The three first sections of figure 1 illustrate the possible situations which determine the relevance of the various processes, as shown in the fourth section.

(1). Precipitation scavenging can be considered as a function of where the processes take place. Below-cloud scavenging denotes washout beneath the visible cloud, which is the source of the precipitation. In-cloud scavenging means scavenging of material within the visible cloud, which results in deposition on the earth's surface.

(2). The type of precipitation, rain or snow, is of course important. Snow and ice crystals are expected to be more efficient scavengers than water drops, because of their larger surface-to-volume ratio. Snow scavenging is also called snowout and rain scavenging is correspondingly denoted rainout. (Unfortunately, rainout is also often used to mean in-cloud scavenging).

(3). The nature of the material to be scavenged has to be taken into account. If it is particulate the size distribution should be specified, and if gaseous the diffusivity and solubility should be specified. With increasing time and distance from a source these characteristics are modified by agglomeration, fragmentation and by attachment and adsorption onto "natural" aerosols. The "natural aerosol" does not have well-defined parameters, the size distribution varies with elevation, source and meteorology. Junge (1963) presents model size distributions that can be used in the absence of measurements.

(4). The scavenging is a sum of a bewildering number of processes: e.g. condensation, Brownian motion, thermophoresis, diffusiophoresis, turbulent inertial interception, and gravitational capture (see e.g. Pruppacher and Klett (1978)).

The particles in the volume of air swept out by a raindrop or snowflake will tend to follow the air. Inertia will cause some fraction of the particles to collide with the raindrop or snowflake. This fraction is called the target or collision efficiency.

Some of these particles may collide elastically and are not collected. The fraction of those remaining with the raindrop or snowflake is called the retention efficiency.

The product of target efficiency and retention efficiency is the collection efficiency, E. The collection efficiency can as a first approximation be regarded as a function of particle diameter, s_p and hydrometeor diameter, s_h:

$$E = f(s_p, s_h)$$

Some information regarding target efficiencies is available in the literature (Langmuir 1948). Very little is known about retention efficiencies. A likely assumption is that of perfect retention.

The below-cloud scavenging processes may be considered as involving material being exposed to moving precipitation elements with some chance of collection. Consequently, scavenging can be described by an exponential function:

$$X(t) = X(0) \cdot \exp(-\Lambda t)$$

where X(t) is the atmospheric concentration of material at time t, X(0) the concentration at time zero, and Λ in units of $time^{-1}$ is called the washout or scavenging coefficient. Λ denotes the fraction of material removed per unit time. The basic assumption is implicitly that the fraction of material removed per unit time is independent of the amount of material present. Usually the exponential description will be true, but if there are too many particles which act as cloud-droplet or ice-crystal nuclei then their presence could influence the precipitation rate, and some gases can saturate the cloud droplets; in that case, only a certain amount, not a specific fraction, of the gas will be removed.

In some references "Λ" is expressed as

$$\Lambda = c\ p^a$$

where c is a constant in s^{-1}, p the precipitation rate in mm/h and a is a number between 0 and 1. This type of relationship is a consequence of the observation that drop size distribution is dependent on the rainfall intensity; a higher intensity gives larger drops.

The interesting result of adding the effects of various scavenging mechanisms was first obtained by Greenfield (1957). His model gave scavenging coefficients showing a strong broad minimum for aerosol particles between about 0.1 and 1.0 µm radius. This minimum is often referred to as the "Greenfield gap" or the "scavenging gap".

A simplificated and alternative formulation for precipitation scavenging for both gases and particles uses the scavenging ratio, w, commonly called the washout ratio. This is usually defined as the ratio of the material concentration in the precipitation at surface level, k_o (curies per volume of precipitation), to its average concentration in the air at surface level, X_o (curies per volume of air). Washout ratios are normally reported for a particular element or compound.

The wet flux to the ground, W, can be written:

$$W = w\ p\ X_o$$

and a wet deposition velocity can be defined:

$$V_w = W/X_o = w\ p.$$

It should be noted that washout ratios represent averages over many parameters, over the precipitation element sizes, the material's vertical distribution, the particle sizes, different chemical forms of the element, different rainfall amounts, different wind directions, different types of storm, different sources, etc. Therefore, washout ratios can be used for predicting precipitation scavenging of routine releases, where long-term averages are of interest. On the other hand, this ratio normally cannot be used in dose calculations for hypothetical reactor accidents.

In most European countries, it is raining or snowing less than 10% of the time. From precipitation statistics (Gylander and Widemo 1980) covering a 5- and 7-year period in Sweden and Denmark, respectively, it is seen that there is precipitation (at a rate greater than 0.1 mm/h) 7.9% and 7.2% of the total time. The same statistics show that in 92.5% and 93.5% of the total precipitation time, the precipitation rate is less than 2 mm/h. Precipitation occurs 85.7% of the time during weather situations characterized by Pasquill categories D and E. It should also be pointed out that precipitation at a rate greater than 1 mm/h during a very stable weather situation, Pasquill category F, has occurred only 0.0076% of the total time.

Numerous measurements of raindrop size distributions have been made, and several empirical equations have been fitted to them. Rain spectra have peak frequencies between drop diameters of 0.5 and 1.0 mm. The distributions given by Marshall and Palmer (1948) and Best (1950) have been most widely accepted and used. Rain spectra sampled by Kelkar (1959) show that for a rainfall rate of 0.2 mm/h 30% of the drops have diameters greater than 0.4 mm; however, for a rainfall rate of 8.6 mm/h 30% of the drops have diameters greater than 2 mm. It is also seen that when the rainfall rate is high, 8 mm/h or greater, the raindrop size distributions do not vary very much. One should bear in mind the effects of evaporation on drop size as it falls to the ground.

Many papers review the general outline of precipitation scavenging. One of the most comprehensive is that of Slinn (1980). Other recent works are by Bonka and Horn (1983), Hales (1983), Nielsen (1981), Brenk and Vogt (1981), and Semonin and Beadle (1977).

2. IN-CLOUD SCAVENGING

By in-cloud scavenging, the upward dispersed or transported radionuclides by atmospheric convection become the condensation nuclei of the atmospheric water vapour and get absorbed into the growing raindrops in the clouds.

The substantial difference between below-cloud and in-cloud scavenging, for the purpose of calculating wet deposition, is determined by the different outcomes of the particles. In the first process, we find the wet deposition on the ground just below the diffusing cloud, whereas in the latter process the radionuclides absorbed by cloud raindrops can be deposited also at distances or in places very far from those interested by the diffusing cloud. In effect, such radionuclides follow the cloud during its atmospheric motion which is basically governed by synoptic flows and winds (Ferrara et al. 1983).

It is extremely difficult to compute the rate of in-cloud scavenging, and further, it is difficult to distinguish between in- and below-cloud scavenging at ground level. The general approach to the problem is not to distinguish between the two processes. This approach was justified by the only known direct experimental test of the relative importance of below- versus in-cloud scavenging performed by Slinn et al. (1979). This study gave evidence of in-cloud scavenging being as efficient as below-cloud scavenging.

3. RAIN SCAVENGING OF PARTICLES

Chamberlain (1953) has calculated washout scavenging coefficients for various rainfall rates and particle sizes. The calculated washout coefficients are shown in figure 2 for different values of $a^2\rho$ where a is the radius and ρ is the density of the particle. It is seen that Λ is almost proportional to both rainfall rate and particle diameter.

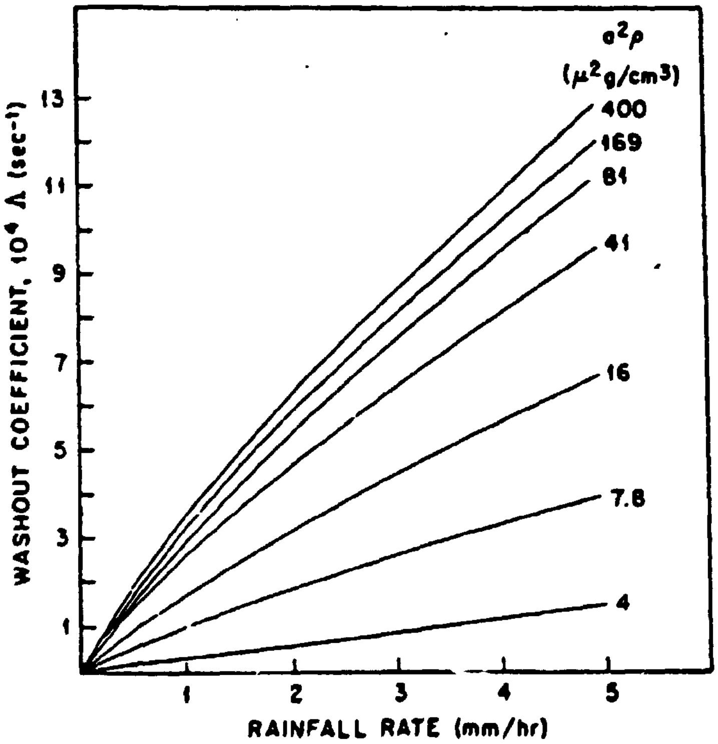

Fig. 2. Washout coefficients for unit density particles vs. rainfall rate and $a^2\rho$, where a and ρ are the radius and density of the particles. (Chamberlain 1953).

Similarly, Slinn (1980) has suggested the following approximation for the rain scavenging rate:

$$\Lambda(a;\bar{r},t) = 0.5\ p(\bar{r},t)\ \bar{E}(a,R_m)/R_m$$

where a is the aerosol particle radius, r the position vector, p the rainfall rate, E the particle/drop collision efficiency and R_m the mean drop radius, which for fairly steady rains can be set to

$$R_m = 0.35\ \text{mm}\ p^{1/4}$$

where p is measured in mm/h. In fig. 3, this approximation is compared with results from experiments in which radioactivity-tagged aerosol particles were released into cumulonimbus storms.

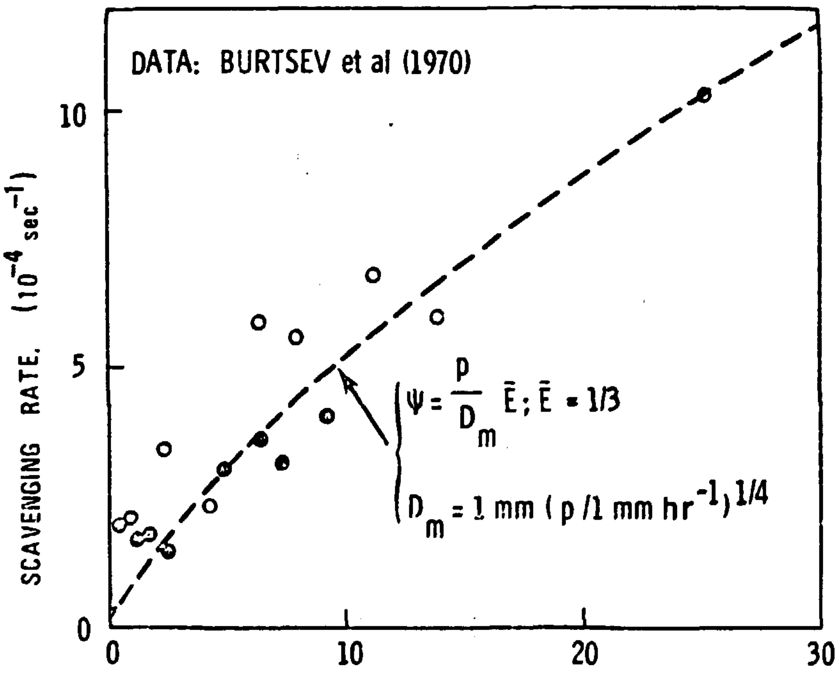

Fig. 3. A comparison of the theoretical prediction of Slinn (dashed curve) against experimental data obtained by Burtsev, Burtseva, and Malakov (1970) for the in-cloud scavenging of tracer mass released into the top of the rain shaft (solid circles) and into the region of "cloud drops" (open circles) of a cumulonimbus cloud. (Slinn 1980).

Brenk and Vogt (1981) recommend a formula for the scavenging coefficient of the type mentioned in chapter 1:

$$\Lambda_p = 1.2 \cdot 10^{-4} \cdot p^{0.5}$$

In the following two figures, they compare this formula with some experimental results:

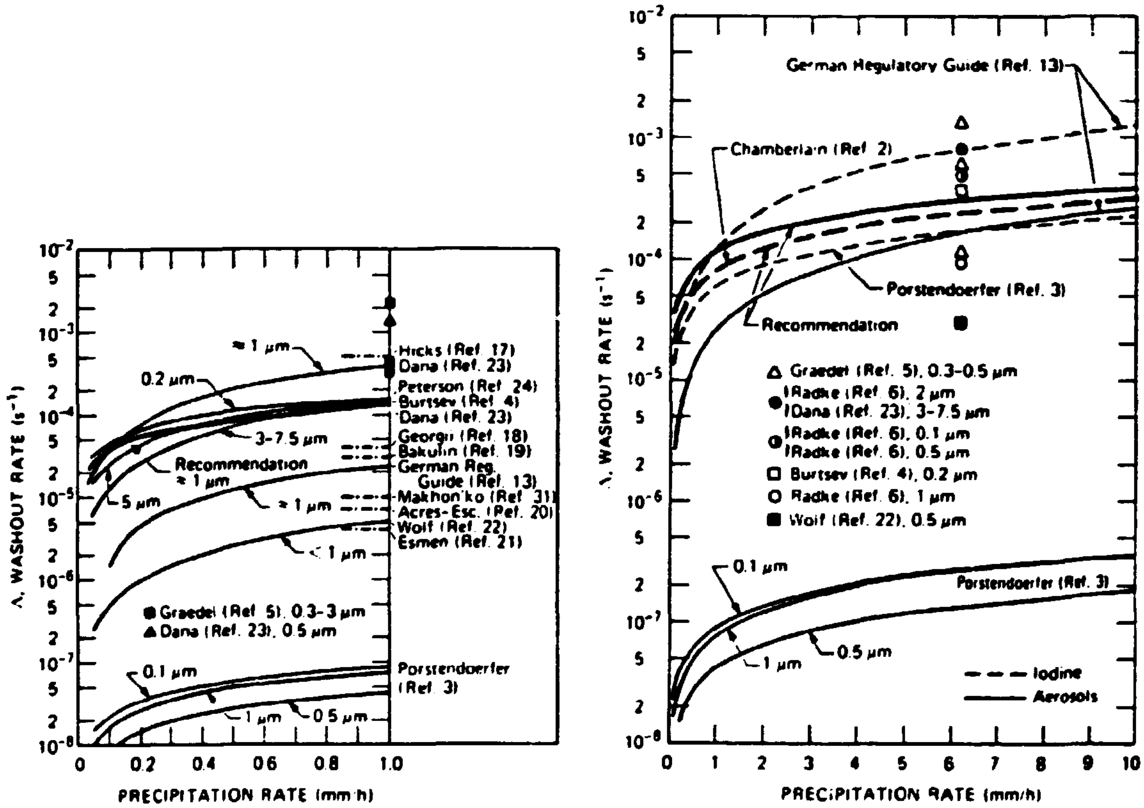

Fig. 4a and b. Washout rates for aerosols and iodine (I_2 vapor) as functions of the precipitation rate. All values have been derived from field experiments except those of Porstendoerfer (1978) and Chamberlain (1953), which have semiempirical character (Brenk and Vogt 1981).

.___.___. Values are constant with respect to the precipitation rate.

The correlation between scavenging rate and the size of aerosol particles (which is of minor interest in relation to accident consequences) has been studied by Radke et al. (1980). They studied plumes from coal power plants and a large paper mill and emission from a volcano. The average rain rate varied from 7-19 mm/h.

The measurements agree well with theoretical calculations for aerosol particles > 1 μm, but for the submicron aerosol particles the scavenging collection efficiencies are generally much higher, and the region of very low scavenging efficiencies (the "scavenging gap") much narrower than current theories predict.

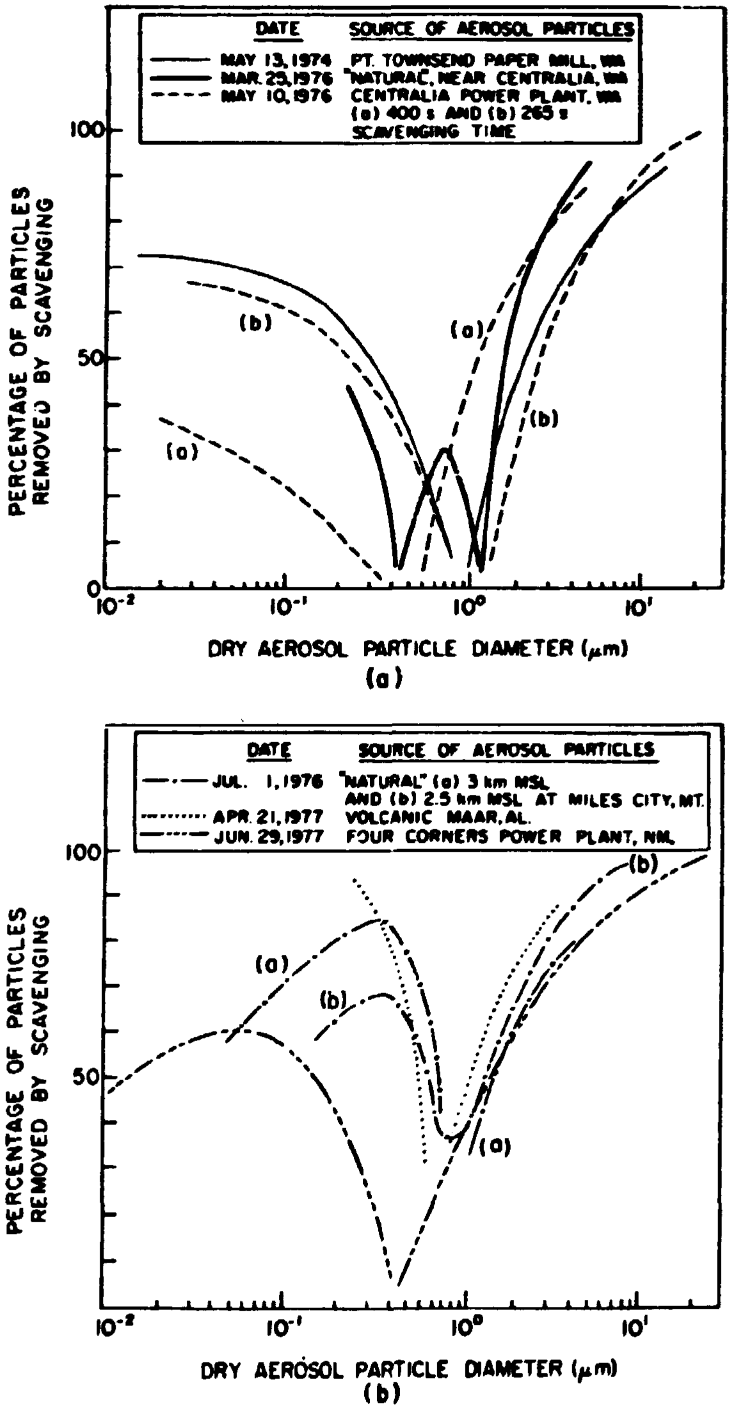

Fig. 5. Percentages of aerosol particles of various sizes removed by precipitation scavenging. (Radke et al. 1980).

4. RAIN SCAVENGING OF GASES

When dealing with precipitation scavenging of gases, the gases should be divided into two categories: those that are highly reactive towards water, e.g. bromine and iodine, and those less reactive that form simple solutions in water, e.g. CH_3I and CO_2. In relation to rain scavenging after reactor accidents the only gas of any importance is iodine.

Experiments performed in connection with spray systems installed inside reactor containments for safety reasons have given a lot of data on iodine scavenging (ANS 1971). However, these experiments were carried out using much higher iodine concentrations than would be relevant to atmospheric situations. Moreover, the sprays often contained chemical additives. Therefore, data from these experiments cannot be used for evaluation of wet atmospheric deposition.

Engelmann et al. (1966) measured the washout coefficient for iodine. The measured values spread over three orders of magnitude. Engelmann and Perkins (1966) used iodine released from a process plant in their study. The washout coefficients from this study are high compared with those of other investigations. This can be explained by the presence of an amount of water vapour in the plant exhausts, sufficient to produce a cloud of water droplets. If the released iodine were inside the water drops before scavenging, the washout rate would be equal to that of the droplets. This can explain why the values of measured washout coefficients are high. Hence, release conditions can play a very important role when evaluating washout.

Brenk and Vogt recommend the following formula for rain scavenging of iodine (cf. Fig. 4b):

$$\Lambda = 8 \cdot 10^{-5} p^{0.6}$$

which is a slight modification of a formula proposed by Chamberlain (1953) and similar to another formula suggested by Porstendoerfer (1978).

Obviously, much more work needs to be done in this field.

5. SNOW SCAVENGING

Washout by snow could be expected to be more effective than for the water equivalent of rain, because slow "feathery" snowflakes have larger surface areas than the equivalent water-drops. Results from the study by Graedel and Franey (1975) indicate snow scavenging to be 28-50 times more effective than water equivalent rain scavenging.

Few studies have been devoted to snow scavenging. One reason may be the large variety of snow types; another the lack of even approximate descriptions of flow fields about snow crystals.

W.G.N. Slinn (1980) has suggested an approximation for the snow scavenging coefficient similar to his approximation for rain scavenging:

$$\Lambda(a;\bar{r},t) = \gamma p(\bar{r},t)\ E(a,\lambda)/D_m$$

where γ is a dimensionless constant of order unity, λ the characteristic capture length scale of hydrometeor, and D_m a characteristic length.

Fig. 6 shows Slinns formula adapted to the experimental data of Engelmann et al. (1966), and Fig. 7 the same formula together with data from Wolf and Dana (1969).

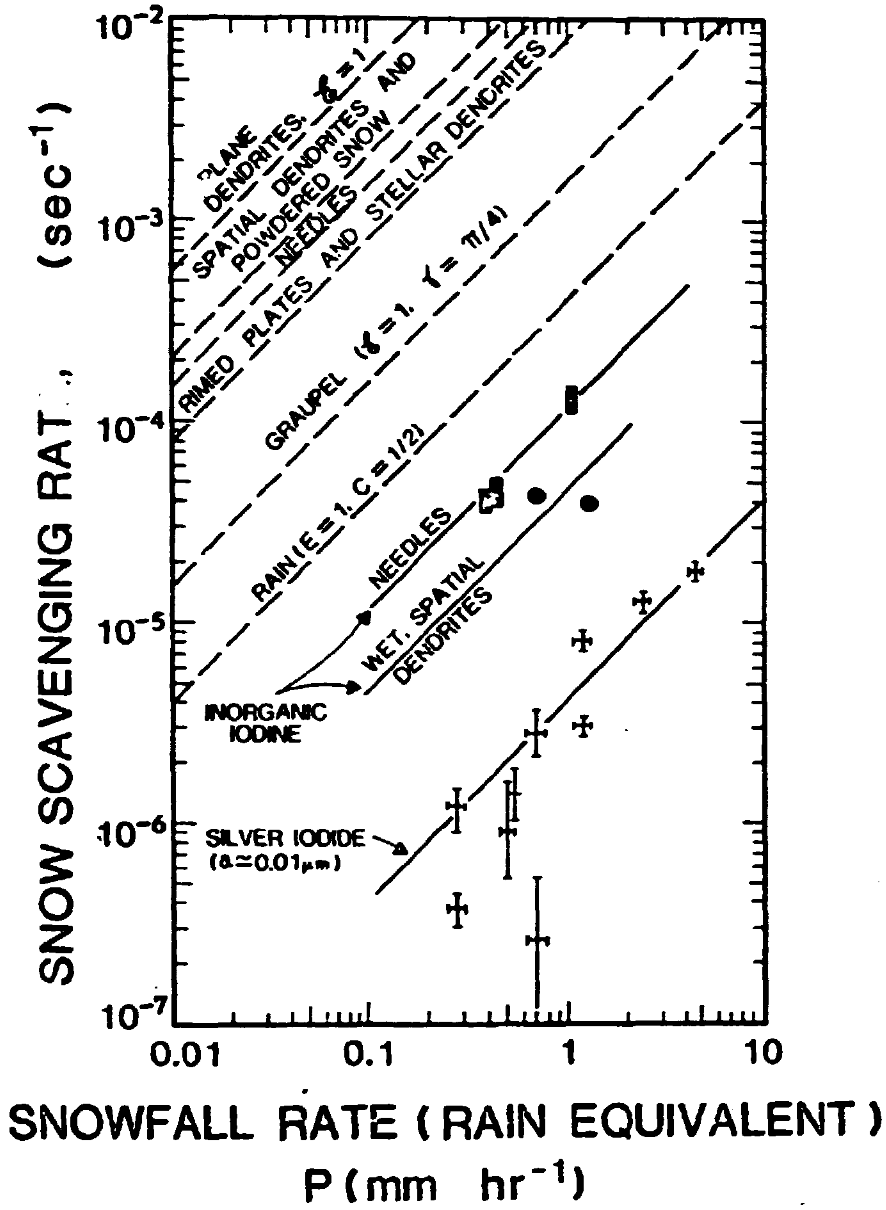

Fig. 6. Estimates for the snow scavenging rates as functions of precipitation intensity. The dashed curves, with E = 1 and $\gamma = \pi/4$, suggest the dependence on crystal type. Different values for γE were chosen to fit the experimental data of Engelmann et al. (1966): $7.3 \cdot 10^{-3}$ for process-plant iodine (possibly dissolved in plume droplets) scavenged by needles; $1.8 \cdot 10^{-4}$ for silver iodide particles scavenged by various crystal types, usually powdered snow or spatial dendrites. (From Slinn 1980).

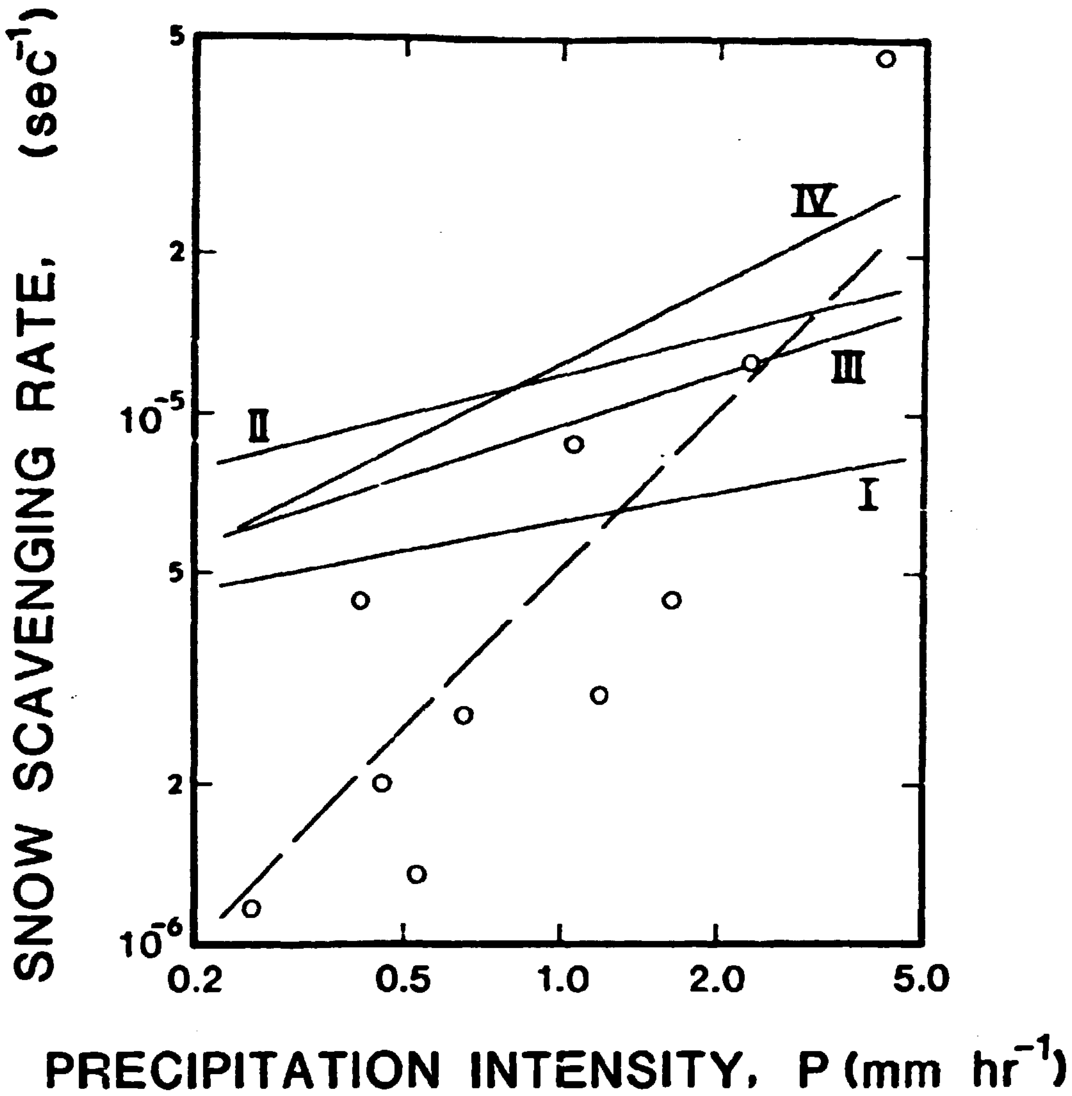

Fig. 7. Comparisons of empirical expressions for the rate of scavenging of particles by snow against data from Wolf and Dana (1969). Curves I-IV are the Knutson and Stockham formulae using D_p = 0.5 μm. The dry mass mean diameter of the AgI particles was actually about 0.012 μm, ranging in size from about 0.002 to 0.05 μm. The dashed curve "through" the data points (circles) is given by Slinn. Adapted from Knutson and Stockham by Slinn (1980).

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