

NATURAL REMOVAL PROCESSES IN THE ATMOSPHERE (1)

by

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INTRODUCTION

All too often, atmospheric removal processes are considered to result in aggravating consequences such as depositions of fly ash, soiling of clothing and painted surfaces, and creating a general nuisance. However, these are the processes by which impurities are removed from the air such that the pollutant concentrations do not continually increase. Without such mechanisms, the atmosphere would ultimately become untenable.

The removal of material from the atmosphere may occur in a number of different ways. Particles large enough and heavy enough will settle to the ground due to the action of gravity. Some gaseous material may be removed by absorption upon particulate matter in the atmosphere or by chemical reactions changing the material into a new compound. Adsorption may also take place at the ground by the earth's surface or by vegetation. Impaction of particles onto buildings and vegetation and turbulent impaction upon the earth's surface are very significant removal processes. Precipitation also removes material from the atmosphere by interception of particulates by falling raindrops (washout) or by raindrop formation within clouds and subsequent falling as precipitation (rainout).

Gravitational Settling

A freely falling particle within the size range found in dusts, smokes, and mists rapidly attains a constant or terminal velocity when the aerodynamic drag on the particle is equal to the weight of the particle. When the particle is of a size comparable with the mean free path of the gas molecules, bombardment by the molecules results in a random or Brownian motion which is superimposed on its downward motion. In considering falling speed, it is desirable to take the simplest case of a rigid spherical particle falling independently of other particles and not so large as to cause inertia effects from the displaced gas to arise, then Stokes' law applies. A spherical particle of density D falling through a medium of density ρ is accelerated under the action of gravity with a force

$$f = \frac{4}{3} \pi (D - \rho) r^3 g. \quad (1)$$

Equation (1) represents the difference between the unimpeded fall of the particle in a vacuum ($\frac{4}{3}\pi r^3 g$), and the buoyant force of the air ($\frac{4}{3}\pi r^3 \rho g$). r is the radius of the particle and g the acceleration of gravity. The accelerating force is opposed by a frictional force arising from the viscosity of the air and the turbulence induced in the air by the passage of the drop. A measure of the turbulence is given by a dimensionless parameter, the Reynold's number, that enters in the theory of the flow of fluids. The Reynold's number is defined as

$$Re = \frac{2r\rho v}{\mu} \quad (2)$$

where v is the relative velocity of the particle to the air of density ρ and viscosity μ . $2r$ is the principal cross-sectional dimension of the particle. The ratio

$$\eta = \frac{\mu}{\rho}$$

is called the kinematic viscosity and has dimensions of $\text{cm}^2 \text{sec}^{-1}$.

The generalized resisting force on spheres has been found to be of the form

$$f_r = 6\pi\mu r v \left(\frac{C_D Re}{24} \right). \quad (3)$$

C_D is called the drag coefficient and is the constant of proportionality between the measured force f_r and the physical quantities in the equation. For low Reynold's numbers, $Re < 1$, $C_D Re/24 = 1$, and equation 3 becomes

$$f_r = 6\pi\mu r v \quad (4)$$

Equation 4 is called Stokes' law.

C_D is not a constant but is a function of the Reynold's number. However, since C_D varies only slowly with r and v , it is useful to use equation 2 to write equation 3.

$$f_r = \frac{\pi}{2} C_D \rho r^2 v^2. \quad (5)$$

Thus, over a limited range of r and v , an average value of C_D may be selected.

Freely falling particles accelerated by a force given by equation 1 are opposed by a constantly resisting force given by equation 3. A point is reached where the two forces balance one another and equilibrium exists. At this point, the velocity reaches a maximum, called the terminal velocity. By equating equation 1 and 3, the terminal velocity v_T becomes

$$v_T = \frac{2}{9} \frac{(D-\rho)}{\mu} g \left(\frac{24}{C_D Re} \right) r^2. \quad (6)$$

For most of our applications, $D \gg \rho$ so that $D - \rho = D$ to all practical limits. When Stokes' law is followed (valid for unit density particles in air for $r \leq 40$ microns, equation 6 becomes

$$v_T = \frac{2}{9} \frac{D_g}{\mu} r^2 = \frac{1}{18} \frac{D_g}{\mu} a^2. \quad (7)$$

Calculation of v_T by equation 7 is straightforward. Once Stokes' law has become invalid, the calculation of v_T from equation 6 becomes tedious because v_T is explicit in the definition of Re and implicit in the definition of C_D . Table I gives values of a , Re , and v_T for unit density spheres at sea level pressure.

TABLE I*

A Short Table of Terminal Velocities, Reynold's Number, and Correction Terms to Stokes' Law for Unit Density Spheres.

Diameter (microns)	$\frac{C_D Re}{24}$	v_T (cm/sec)	Re
20	1.00	1.24	0.017
50	1.00	7.72	0.268
80	1.00	20.	1.11
200	1.68	72	9.61
400	2.98	162	43.2
800	5.94	327	175
1200	9.41	464	372
2000	18.7	649	866

*Physical Meteorology - J. C. Johnson, Wiley and Sons 1954. page 230.

Thus, particles large enough or dense enough to have appreciable terminal velocities will fall to the ground within rather short travel distances, depending on their height of release. This process constitutes a significant removal process for very large particles, of fly ash or dusts from cement plants. Because of the inertia of these particles, their reaction to the turbulence in the atmosphere is retarded. Also, because of their settling, they are in motion relative to specific eddies. Therefore, the action of settling actually changes the diffusion of these particles relative to what would occur with gases.

The concentrations in air of an effluent released from a single source can be expressed in the Gaussian form

$$X = \frac{Q}{\pi \sigma_y \sigma_z \bar{u}} \exp \left[-\frac{1}{2} \left(\frac{y^2}{\sigma_y^2} + \frac{h^2}{\sigma_z^2} \right) \right] \quad (8)$$

where X represents the concentration in gm/m³
Q represents the source strength in gm/sec
u represents the mean wind speed in m/sec
y represents the distance crosswind from the plume axis
in meters
h represents the source height in meters

σ_y^2, σ_z^2 represents dispersion coefficients in m²

This equation describes the distribution of concentration only when there are no processes depleting the cloud. One method used for correcting this equation for deposition amounts to permitting the diffusing plume to settle at the average terminal velocity of the particles as the material travels downwind (See p. 93, Meteorology and Atomic Energy.). Thus, the height of emission is decreased with distance according to the settling velocity. The corrected emission height is then substituted into the diffusion equation. The corrected height of emission, H', is given by the following equation

$$H' (\text{meters}) = H (\text{meters}) - \frac{V (\text{m/sec}) X (\text{m})}{\bar{u} (\text{m/sec})} \quad (9)$$

where H is the effective stack height of the source, v is the settling velocity determined by Stokes' law, x is the distance downwind, and u is the mean wind speed. Van der Hoven (1962) used a deposition model of the tilted plume type to estimate the deposition from ground tested

nuclear engines. As explained by Csanady (1957), the tilted plume model should have a further correction for the source strength term for loss by deposition.

Diffusion to the Surface and Impaction

Generally, only particles in the size range greater than 10 microns have appreciable gravitational settling velocity. However, recent studies (Simpson 1961; Islitzer and Dumbauld, 1962) indicate deposition of micron and submicron particles upon the ground and vegetation may be considerable. Accounting for deposition of these smaller particles requires extension beyond the simple gravitational settling model.

Using the concepts presented by Chamberlain (1953) along with extensions reported by Healy (1957), a reasonable model can be developed. For these small particulates, deposition from the cloud to the ground is conceived as a process limited by meteorological diffusion and processes of impaction and sticking to objects on the ground rather than the settling of particles through the influence of gravity. It is visualized that the particles are brought through the boundary layer to the ground by the turbulent diffusion process. They then deposit on vegetation or other objects by inertial impaction and diffusion and stick by electrostatic forces, chemical attraction or other means. Investigations of this process in the neutral condition assuming that the transfer coefficients for mass and momentum are equal indicates that the velocity of deposition should vary with wind speed. There is also evidence that the velocity of deposition changes with atmospheric stability. Data on the velocity of deposition of Iodine-131 at Hanford and from experiments conducted by the English have indicated a value of about 2.7 cm/sec. Calculations based on the equivalence of mass and momentum transfer coefficients during neutral conditions yield about the same value. Measurements of the velocity of deposition of fission products from arc burned uranium have indicated that the velocity of deposition of these particles is considerably lower, on the order of 0.1 cm/sec.

Gifford and Pack (1962) recently published an evaluation of most of the experimental data obtained to date on deposition velocities of interest in nuclear safety studies. Major conclusions were that the deposition velocities for active materials such as Iodine-131, sulfur dioxide, and ruthenium on flat plates or bare soil is less than 1 cm/sec and is between 1 and 3 cm/sec for deposition on vegetation. Also, it was concluded that the average deposition velocity of inert materials such as Cesium-137 and Strontium-90 on flat plates and vegetation is less than 0.1 and 0.1-0.2 cm/sec, respectively. These results are quite consistent, indicating that for particles of diameter less than 10-15 microns, the relative effects of impaction, diffusion, and absorption are more important than the widely varying gravitational settling velocities.

It is postulated that the particles are brought to the ground by turbulent diffusion where the iodine reacts with the vegetation to absorb strongly on the surface. The small smoke particles must depend on impaction or some other process to stick. Thus, although the rate of transfer of both materials to the ground was the same, the velocity of deposition was different because of the difference in retention.

Chamberlain (1953) bypasses these effects, and simplifies the problem by defining a deposition velocity V_g , as

$$V_g = \frac{\text{amount deposited/cm}^2 \text{ per sec}}{\text{volumetric concentration/cm}^3 \text{ above the surface}} \quad (10)$$

This definition may be expressed in the integral form as

$$V_g = \frac{\text{total deposition/cm}^2}{\text{dosage/cm}^3} \quad (11)$$

Thus, the deposition becomes

$$w(x,y) = X \cdot V_g. \quad (12)$$

To account for depletion of material from the cloud, the equation must satisfy mass continuity principles such that

$$\int_0^{\infty} \int_0^{\infty} X V_g dy dx = Q \quad (13)$$

This equation is analytic in the first integration, but the resulting equation from combining (13) and (8)

$$I = \int_0^{\infty} \frac{V_g}{\sqrt{\pi \sigma_z^2} \bar{u}} \exp - \frac{1}{2} \left(\frac{h^2}{\sigma_z^2} \right) dx \quad (14)$$

can be solved analytically if σ_z can be expressed in terms of x .

To account for cloud depletion resulting from the deposition, the constant source strength, Q , is replaced by a quantity Q_x , depending on x , which is defined so that

$$\frac{\partial Q_x}{\partial x} = - \int_{-\infty}^{\infty} w(x,y) dy. \quad (15)$$

Q_x is the total depletion to a distance x . Again, it is necessary to employ explicit relationships for the dispersion in order to derive specific deposition formulae.

Removal by Rainfall - Rainout

The cleansing of the air by rain is an everyday experience and is one of the most important scavenging mechanisms in the atmosphere. In an analysis of rain scavenging of radioactive particulate matter from the atmosphere, Greenfield (1957) found that the direct interaction of raindrops and particles does not account for the efficient removal of material whose diameter is below approximately one micron. However, he was able to explain the removal of smaller particles by allowing them to mix with the water cloud before the rain starts. The small particles that are scavenged by coagulation are then placed in a position to be more efficiently removed by the rain.

Our present knowledge is still inadequate to provide anything beyond a suggestion of the scavenging processes in the atmosphere for such finely divided material as radioactive debris. Evidence from world-wide fall-out studies suggests that the very small fission products become attached to the natural aerosol particles and then have a history in the atmosphere similar to that of the host. The time required for such a coalition to near completion is not known, but one would expect it to be as rapid as the coagulation with larger water droplets because of the relative differences in mean free paths. Junge (1958) suggests that the predominant cause of the modification in size-distribution of the stratospheric aerosols on the way down through the troposphere is the repeated cycle of condensation and evaporation of clouds, a process requiring considerable time. Thus, the scavenging mechanism for small particles is envisioned as cloud-droplet-scavenging coupled with later scavenging of the cloud droplets by larger raindrops. The amount of activity that will fall out as rain will depend upon the time of mixing of the contaminant and the cloud elements and the rate at which the cloud elements are swept from the cloud by larger raindrops.

Barad, Haugen, and Fuquay (1960) made estimates of some of these scavenging parameters. The effective scavenging constants, that is, the time required for the number of particles to decrease to $1/e$ of the initial value, for the contaminant activity in the presence of cloud droplets

characteristic of stratus and cumulus cloud conditions were computed from Greenfield's curves, assuming that the activity was proportional to the volume of the particle. The cloud characteristics used by Greenfield compared favorably with later data reported by aufm Kampe and Weickman (1957) and were not altered. The calculated times required for one-half the activity from the air to enter the cloud droplets, that is, the scavenging constant Ψ , is shown in Table II.

The amount of activity that will fall out as rain will depend upon the time of mixing of the contaminant cloud elements and the rate at which the cloud droplets are swept from the cloud by larger raindrops. This latter aspect was studied by Chamberlain using Langmuir's theory (1948) of the formation of raindrops by coalescence with smaller raindrops and Best's (1950) relationship between raindrop size and rainfall rate. Results of Chamberlain's study applicable to this type of scavenging are also summarized in Table II. The scavenging of materials by clouds can only occur at heights at which clouds form. The heights of the various types of clouds vary within wide limits. However, scavenging by stratus clouds should be limited to 5000 feet and below, whereas scavenging by cumuliform clouds can occur at any height between 2000 feet and the tropopause.

TABLE II. Parameters for cloud-droplet scavenging of particulate cloud and precipitation scavenging of cloud droplets.

Height Interval	Cloud Type	Rainfall Rate (mm/hr)	Scavenging Constant, Ψ (sec ⁻¹)	Elimination Constant, (sec ⁻¹)
500-5,000 feet	Stratus	0.5	5×10^{-6}	2×10^{-4} ⁶⁰ $\frac{59 \text{ min}}{3465 \text{ sec}}$ _{0002/690}
5,000-35,000 feet	Cumulus	3.5	6×10^{-5}	1×10^{-3}

Removal by Rainfall - Washout

Calculations for precipitation scavenging of materials released into the lower atmosphere usually consider only the interaction between the falling raindrop and the contaminant particles. However, the computed values correspond only approximately to the actual conditions, as the size of raindrops always show a spectral distribution. Best's curves contain mean values. Large deviations from this curve are found in individual rains with the extreme range in rainfall rate for a given mean drop size about a factor of two. In addition, the Langmuir theory is applicable to coalescence of waterdrops, with certain restrictions, and particle interception could be quite different, depending on the physical and chemical nature of the particle.

In an effort to clarify the scavenging processes in the lower layers of

the atmosphere, a part of the total scavenging problem, Hanford meteorologists have been dispersing zinc sulfide into the air and measuring the amounts scavenged by both natural and artificial rain. Results thus far indicate a peak scavenging efficiency for raindrops of about 0.4 mm diameter and suggest a minimum scavenging efficiency for raindrops of 0.7-1.0 mm diameter for this material. This deviation from currently used theoretical calculations which predict a rather flat peak in efficiency for drops larger than 1.0 mm suggests that rainfall intensity as now related to rainfall type may not have as great an effect on washout values as previously thought.

In order to account for washout in the dispersion equations, we can write

$$Q_N = Q \exp\left(-\frac{\lambda x}{u}\right), \quad (16)$$

where Q_N is the source strength correction for the scavenged material. Since rain removes material from the whole cloud depth, the process can be likened to radioactive decay in that the entire cloud is affected uniformly rather than preferentially near the ground, and the shape of the cloud distribution function is not altered. In this case, the concentration from a ground level source becomes:

$$X = \frac{Q}{\pi \bar{u} \sigma_y \sigma_z} \exp\left(-\frac{\lambda x}{u}\right) \exp\left[-\left(\frac{y^2}{2\sigma_y^2} + \frac{z^2}{2\sigma_z^2}\right)\right] \quad (17)$$

The amount of material deposited per second by rainout, w_r , is obtained by multiplying equation (17) by λ and integrating with respect to z .

$$W_N(x, y) = \frac{\lambda \epsilon}{\sqrt{2\pi} \bar{u} \sigma_y} \exp\left(-\frac{\lambda x}{u}\right) \exp\left[-\frac{y^2}{2\sigma_y^2}\right] \quad (18)$$

Values of λ , the washout per second, are the subject of considerable speculation, and observational data are only now becoming available.

Scavenging in a Pollution Episode

Meethan (1956) has discussed some interesting aspects of the London smog of 5-9 December, 1952, in which about 4000 people died above the

normal death rate in Greater London. You have covered most of the details in an earlier session. Perhaps, you did not cover some of the speculations regarding scavenging mechanisms in this episode.

Much can be learned by studying the physics of fogs themselves. City fogs of this episode magnitude have an estimated chance of recurrence of about 2% in any given winter. They are persistent, of limited area and volume, and relatively homogeneous; but not too well understood. You will recall that daily measurements of smoke and sulfur dioxide were being made at this time, but little if anything was known regarding concentrations of trace constituents, such as vanadium compounds, coal distillates, or aldehydes. Other materials, no doubt, present in the smog include sulphuric acid, carbon monoxide, hydrochloric acid, fluorides, and excess carbon dioxide. Consequently, the smog was greatly different in chemical composition from a relatively clear fog.

Meethan considered an area of 450 square miles in the London Basin containing about 8 million people. The fog covered the entire area to a height of about 500 feet. The temperature was near freezing so that the mass of air in this volume was 226 million tons. Material could diffuse from this volume upward at only a very slow rate because of the capping inversion above the fog. He estimated that the light and variable winds could have produced no more than one air change in about four days. Thus, he considered the region as a closed system in which to make estimates.

The air contained about 2,000,000 tons of liquid water as fog and 750,000 tons as vapor. The film of water on the ground, vegetation, and other objects was estimated to weigh 500,000 tons. The air also contained 380 tons of free smoke and 370 tons of free sulfur dioxide, not including any that was attached to or dissolved in fog droplets, or any other sulfur dioxide attached to smoke. The other impurities were not accounted for in the estimates.

About 70,000 tons of coal were burned each day in the region; releasing 1000 tons of smoke particles, 2000 tons of carbon dioxide, 140 tons of hydrochloric acid, and about 14 tons of fluorine compounds. Imperfect combustion of coal and motor vehicle exhaust contributed 8000 tons per day of carbon monoxide. In addition, 200,000 tons of carbon dioxide were emitted per day to be added to the 90,000 tons originally present.

Now, let us look at the heat balance. Essentially all of the heat from burning 70,000 tons of coal went into the fog volume and the heat equivalent of another 1400 tons of coal was added from the earth and surrounding air, which was warmer than the fog. This incoming heat was sufficient to warm the whole volume by 10° C per day, yet the temperature

1.2 $\frac{g}{m^3}$

over the five days remained essentially constant. Fog behaves in the opposite way to a greenhouse so that radiation permitted this heat loss from the fog. If it had warmed up, the fog would have dissipated.

Another paradox is in the water balance. As in most persistent fogs, droplets of water are continually falling to the ground, and yet there is no reduction in the number of droplets in the air. In the London fog, the droplets had to fall through saturated air an average distance of 250 feet to reach the ground, a process would take about 6 hours. Thus, an estimated 800,000 tons of water per day left the fog and some-
way an equal amount replaced it.

Combustion of coal added some 35,000 tons per day and humans an additional 2000 tons. The remaining 763,000 tons per day, must have been evolved from evaporation from the ground. The heat coming up from the ground was sufficient to evaporate 18 million tons of water per day, so that it appears reasonable to assume that 4.3% of this heat was used for the evaporation of water, which ultimately replenished the fog.

Thus, we have a somewhat plausible explanation of how fogs can persist in spite of apparently losing water and gaining heat.

Now let us examine the smoke balance. Smoke particles entered the fog at the rate of 1000 tons per day, and must have left at the same rate because a fairly steady equilibrium was maintained at 2.2 mg/m^3 , or 380 tons in the whole region. The average smoke particles must have remained in the air for $380/1000$ of a day or about 10 hours. Smoke particles are far too small to fall of their own weight as much as 250 feet in 10 hours. Meethan calculated that any smoke particle must collide with a fog droplet every couple of minutes or so and could have stuck to the droplet after some 4-10 hours. There was considerable dirt on the pavement which may have been smoke originally.

In the sulfur balance calculation, he assumed a rate of 2000 tons of sulfur dioxide per day entering the fog, and left at the same rate, maintaining a fairly steady equilibrium of 370 tons in the air. The average free life of a sulfur dioxide molecule would, therefore, be $370/2000$ of a day or 4.5 hours. He considers that a few sulfur dioxide molecules were dissolved in the water on ground, vegetation, etc., but most of them were removed by the fog droplets, whose total surface area was about $170,000 \text{ km}^2$ compared with only 1160 km^2 of ground. The average sulfur dioxide molecule spent, perhaps, 0.05% of its time dissolved in fog droplets, moving freely in and out of them. Eventually, after a free life of 4.5 hours, during which about 8 seconds were spent within droplets, it became oxidized within a droplet, and remained fixed there

as sulphuric acid. Six hours later, on the average, the droplet fell to the ground. If these calculations are correct, the mass of sulphuric acid in equilibrium in the fog must have been 800 tons. Its concentration was 4.5 mg/m^3 in the air, and, on the average, the fog droplets were a 0.4% solution of sulphuric acid.

Chlorine from coal entered the air in the form of hydrochloric acid at a rate of 140 tons per day. It was quickly dissolved in the fog droplets, remaining in the air an average of 6 hours. Its concentration was 0.2 mg/m^3 in the air or 0.02% in the fog droplets. Fluorine was present in about one-tenth of these amounts.

Oxides of carbon are not scavenged effectively and probably increased steadily throughout the period of fog. It was calculated that carbon dioxide reached a maximum concentration in the air of 0.4% by weight, or ten times its natural concentration. The concentration of carbon monoxide was calculated to be 180 mg/m^3 or 0.07% by weight.

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