On the giant sea-salt particles in the atmosphere

I. *General Features of the Distribution*[1]

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ABSTRACT

Some observational data of giant sea-salt particles in the atmosphere, now available from many references, were examined to find mechanisms that would explain their distribution.

The first part concerns the vertical distribution over the sea. It is shown that the average decrease of the concentration with height, as a function of particle weight, is quantitatively explained by a combination of sedimentation, diffusion, and convection processes. It is also suggested that the frequent observation of a maximum concentration at some level above the sea surface may be explained by the presence of a boundary layer of a thickness of several meters over the sea surface, where relative humidity steeply changes, in a nonsteady state.

In the second part, the distribution over the continent is discussed. The vertical distribution inland is characterized by a low concentration near the ground which may be ascribed mainly to impaction on ground obstacles, an effect which is much greater than dry fallout. This type of distribution is analytically expressed, and some relations have been formulated; for example, the ratio between the maximum and the ground concentrations as related to an efficiency of the impaction by ground obstacles, also the height where the maximum concentrations appear as related to the factor representing the exponential decrease of the concentration with height in an equilibrium distribution at sea. These expressions are used with available data of the actual distribution in order to examine values of the included factors. Transition from the distribution over the sea to that over the continent is also discussed.

1. Introduction

Giant sea-salt particles in the atmosphere act in producing rain in non-freezing clouds. Even in those clouds whose tops extend above freezing level in middle latitudes, the warm rain mechanism seems to play some important role in rain initiation (BRAHAM, 1963).[3] Hence the manner in which they are distributed over the world is an interesting meteorological problem. The particles are, in general, produced on the sea surface, representing a phase of direct mutual reaction between sea and atmosphere. The circulation of the particles from the sea to the continent is also an interesting geochemical problem. This author and collaborators have obtained some knowledge of the process of production of sea-salt particles on the sea surface by model experiments and other techniques (HAYAMI & TOBA, 1958; TOBA, 1959, 1961; TOBA & TANAKA, 1963*a*). In this report an attempt is made to examine observational data, now available from many references, of giant sea-salt particles from various altitudes and locations, and to find mechanisms that will explain their distribution as far as possible.

In the consideration of the distribution over the sea, the present study is based on the observational data above an air-sea boundary layer of several meters. Through an understanding of the complicated processes that must exist within the boundary layer, it should be possible to relate the process of production at the actual air-sea boundary to the distribution treated here, but this problem is left for the future.

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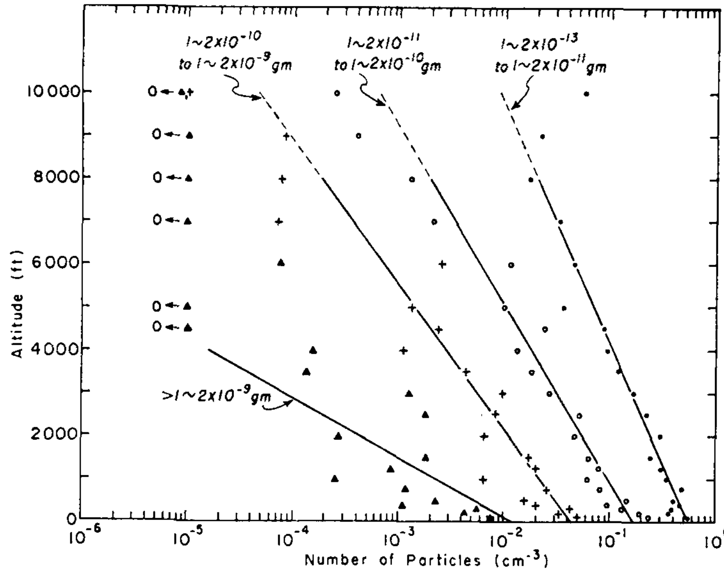


FIG. 1. An example of averaged vertical distribution of giant sea-salt particles over the sea, which was obtained by taking an average from all data of DURBIN & WHITE (1961).

The data on which the present discussion of the distribution over the continent is based, are by no means sufficient. As more data become available, it may be desirable to reconsider some of the conclusions.

2. Distribution of giant sea-salt particles over the sea

2.1. MAIN FEATURE OF THE DISTRIBUTION OVER THE SEA

It has been recognized that there are two characteristic features in the distribution of giant sea-salt particles over the sea. One is that, although the particle concentration falls off with increasing height in an approximately exponential manner, there is little size effect, that is, the size distribution is roughly the same with height. Another is that a maximum in the concentration is sometimes found at some level above the sea surface, 1500 ft or so, especially with weaker wind forces. The former characteristic is studied in detail in this section and an explanation is given; the latter will be discussed in the next section.

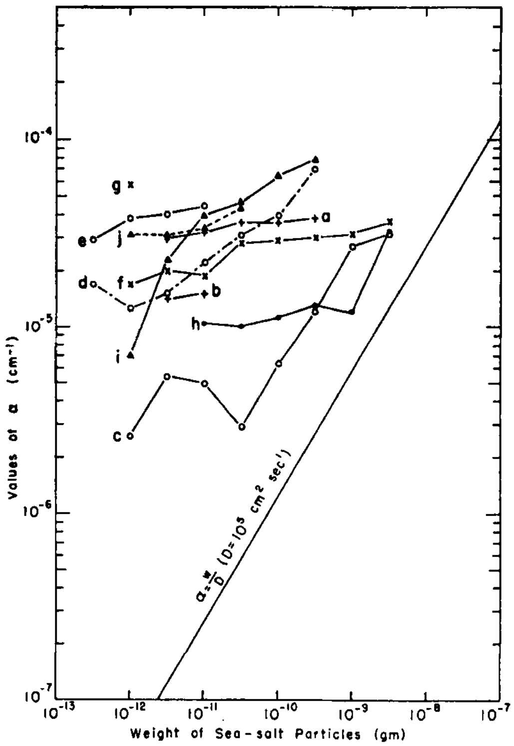
As to the absence of an appreciable size effect, JUNGE (1957*b*) assumed that there was no gravitational separation, and attributed the cause to a nonsteady state. On the other hand, Woodcock and Eriksson (ERIKSSON, 1959), and MORDY (1959) were inclined to explain this fact as due to coalescence of droplets within a cloud and subsequent evaporation which would lead to an excess of larger particles at cloud levels.

Much data of giant sea-salt particle distribution, available from various references, were examined and it was found that the distribution over the sea is close to an exponential distribution, if we take an average from a certain number of data, at least below several thousand feet, although individual data fluctuate widely. Fig. 1 shows an example of such averaged distributions. Points were obtained by taking an average from all data of DURBIN & WHITE (1961) for each height shown in the figure, and straight lines were determined by the method of least squares, using the points below 4000 ft level for the particle class of the largest size, and those below 8000 ft level for the other three classes.

Moreover, if we assume the distribution of number concentration of particles (θ) of a certain class of weight (not cumulative number) expressed by

$$\theta = \theta_0 \exp(-\alpha z), \qquad (1)$$

where θ_0 is the value of θ at $z = 0$, and z the



Curve	Wood-cock's Fig. No.	Altitude (m)	Wind Force
a	3	561–1245	3
b	3	1245–2285	3
c	4	518–1070	4–5
d	4	1070–1550	4–5
e	4	1550–2740	4–5
f	5	732–1553	5
g	5	1553–2440	5
h	6	790–1370	3–4
i	7	640–1220	6
j	7	1220–1980	6

FIG. 2. Change in α with particle weight, obtained from individual data of WOODCOCK (1953).

vertical coordinate positive upward, then the value of α takes a characteristic form; and if we calculate the value from the averaged distribution as in Fig. 1, the value falls in a narrow range, and it is a function of particle weight. Fig. 2. shows the values of α calculated from individual data of WOODCOCK (1953), Fig. 3 shows the values from Fig. 1 and from averaged data from WOODCOCK (1953 and 1957). Woodcock's 1953 data were averaged in the form of particle concentration for various weight classes as is shown in the figure, for three classes of height, of which the average heights were 616 m, 1335 m and 2361 m, respectively. The α's for two height ranges (616–1335 m and 1335–2361 m) were calculated. From Woodcock's 1957 data, concentrations of particles for four weight classes: 1.7×10^{-13} to 4.4×10^{-12} gm, 4.4×10^{-12} to 5.6×10^{-11} gm, 5.6×10^{-11} to 3.6×10^{-10} gm and 3.6×10^{-10} to 2.0×10^{-9} gm were averaged for each of seven 1000-ft height classes from 500 to 7500 ft, and α was calculated from these seven points, again by the method of least squares, for each weight class. The magnitude of the standard deviation is shown in Fig. 3 as the length of a vertical line at each point. Fig. 4 shows the values of α from Toba and Tanaka's 1963*b* data which include observation over land and sea in Japan, and from Woodcock's 1953 data which were taken from over sea to a lee side of a mountain in Hawaii.

The curves in Figs. 2 and 3 are nearly horizontal on the left edges and seem to approach asymptotically a line of $\alpha = w/D$, where w is the terminal velocity of particles, and D the eddy diffusivity. The effect of gravity apparently exists. This type of a curve may be expressed by

$$\theta = \theta_0 \exp\left\{-\left(\frac{w}{D} + \beta\right)z\right\}, \qquad (2)$$

where $$\beta = \alpha - \frac{w}{D} = \text{constant}.$$

From the horizontal part of the curves in Fig. 3, the value of β is estimated to be about 10^{-5} cm^{-1}.

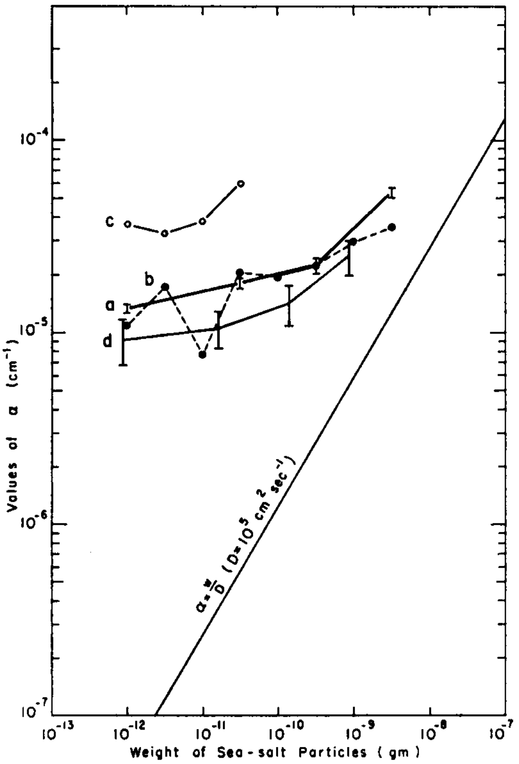


FIG. 3. Values of α obtained from averaged distribution of sea-salt particles.

Curves: a, From all data of DURBIN & WHITE (1961). *b*, From the 616 m and 1335 m data of WOODCOCK (1953). *c*, From the 1335 m and 2361 m data of WOODCOCK (1953). *d*, From all data of WOODCOCK (1957).

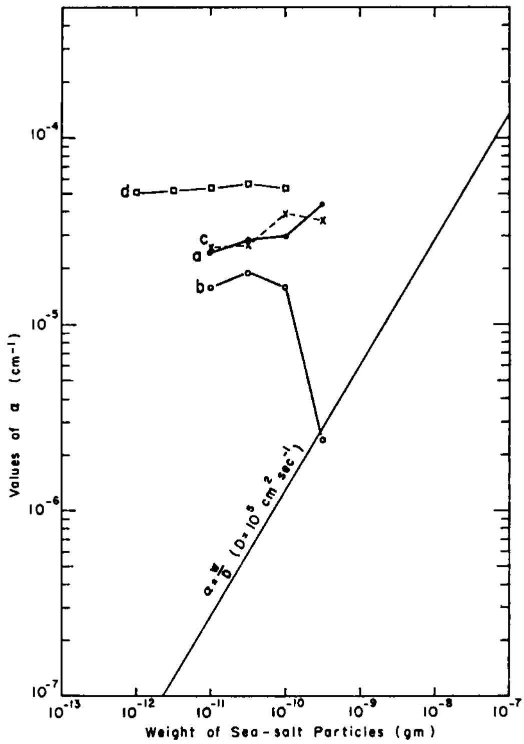


FIG. 4. Values of α obtained from data over land.

Curves: a, From 1000 m (over land)—2000 m (over land) data of TOBA & TANAKA (1963*b*). *b*, From 2000 m (over land)—3000 m (over sea) data of TOBA & TANAKA (1963*b*). *c*, From 300 m (over sea) —2100 m (over land) data of TOBA & TANAKA (1963*b*). *d*, From 457 m (over sea)—1370 m (lee side of Mt.) data of WOODCOCK (1953, Fig. 8).

It should be noted that the reciprocal of α has dimensions of length, being actually the height at which $\theta = \theta_0/e$. It is approximately the same concept of a coefficient which was introduced by BYERS (1957) in respect to the vertical distribution of water vapor, and which has a connotation of a characteristic mixing length or height for vertical distribution by eddy diffusion.

First we consider a case where sedimentation-diffusion equilibrium holds in the atmosphere, pressure of which is a function of z. The differential equation has a form of

$$w\theta = wn\nu = -Dn\frac{d\nu}{dz},$$

where n is the number of air molecules per unit volume and $\nu = \theta/n$ is the mixing ratio of particles with air molecules. The solution for the condition of $\nu = \nu_0$ at $z = 0$ is that

$$\nu = \nu_0 \exp\left(-\int_0^z \frac{w}{D}dz\right)$$

or

$$\theta = \theta_0 \frac{n}{n_0} \exp\left(-\int_0^z \frac{w}{D}dz\right). \tag{3}$$

Since the number of air molecules per unit volume is expressed by

$$n = \frac{pN}{mRT},$$

where p is the atmospheric pressure, m the molecular weight of the air, N Avogadro's number, R the gas constant for the air, and T the absolute temperature, it follows that

$$\frac{n}{n_0} = \frac{p}{p_0}\frac{T_0}{T}$$

$$= \frac{T_0}{T} \exp\left(-\frac{g}{R}\int_0^z \frac{dz}{T}\right),$$

where g is the acceleration of gravity, and equation (3) is to have a form of

$$\theta = \theta_0 \frac{T_0}{T} \exp\left\{-\int_0^z \left(\frac{w}{D} + \frac{g}{RT}\right) dz\right\}. \quad (4)$$

If we assume constant values of w and D, and also an isothermal atmosphere, equation (4) is reduced to equation (2), where

$$\beta = \frac{g}{RT}.$$

The numerical values of the factors: $g = 980$ cm sec^{-2}, $R = 2.87 \times 10^6$ ergs gm^{-1} °K^{-1} and $T_0 =$ 263°K to 298°K, give a value of $\beta = (1.30$ to $1.15) \times 10^{-6}$ cm^{-1}, which is smaller than the observed value of β by about one order of magnitude. If the air temperature decreases with height as in the usual case, the β is smaller than the above values, and it will diminish for a homogeneous atmosphere. It is concluded that the effect of the density decrease with height is not sufficient in explaining the actual distribution.

The larger values of β may be interpreted as factors in the exponential distribution by which the sedimentation-diffusion equilibrium must be multiplied, producing a reduction factor which becomes stronger exponentially with height. From the value of β of 10^{-5} cm^{-1}, it follows that the reduction becomes nearly complete at 3 km level because $\exp(\beta z) \approx \exp(3) \approx$ 0.05 there. The factor might be considered as expressing a sink, such as cloud formation, coalescence and removal by rain, and in this case, it is more efficient in higher levels by an exponential manner.

There is a possibility, however, that the curves in Figs. 2, 3 and 4 may be derived and the β may be well explained, by introducing vertical motion of the air. We consider a system including the vertical air velocity W taken positive downward. The diffusion equation with no horizontal gradient will be written as

$$\frac{\partial}{\partial t}(n\nu) = \frac{\partial}{\partial z}\{(w+W)n\nu\} + \frac{\partial}{\partial z}\left(Dn\frac{\partial \nu}{\partial z}\right), \quad (5)$$

where t is time. Equation (5) may be expressed by the equation of continuity:

$$\frac{\partial(n\nu)}{\partial t} = \frac{\partial \gamma}{\partial z} \quad (6)$$

and $$\gamma = (w+W)n\nu + Dn\frac{\partial \nu}{\partial z}, \quad (7)$$

where γ is the flux, positive downward, of particles. The solution of equation (7) for the condition of $\nu = \nu_0$ at $z = 0$ becomes

$$\nu = \left\{\nu_0 + \int_0^z \frac{\gamma}{nD} \exp\left(\int_0^z \frac{w+W}{D} dz\right) dz\right\} \cdot \exp\left(-\int_0^z \frac{w+W}{D} dz\right).$$

As a first approximation, assume a homogeneous atmosphere, $n =$ constant (this approximation is reasonable because the effect of the change in n was small as was already shown), and regard the factors $w + W$ and D also as constant, to obtain

$$\theta = \left\{\theta_0 + \frac{1}{D}\int_0^z \gamma \exp\left(\frac{w+W}{D}z\right) dz\right\} \cdot \exp\left(-\frac{w+W}{D}z\right). \quad (8)$$

We now consider a steady state:

$$\frac{\partial \theta}{\partial t} = 0 \quad \text{or} \quad \gamma = \gamma_0 = \text{constant},$$

to obtain

$$\theta = \theta_0 \exp\left(-\frac{w+W}{D}z\right) + \frac{\gamma_0}{w+W} \cdot \left\{1 - \exp\left(-\frac{w+W}{D}z\right)\right\}. \quad (9)$$

The actual phenomena, for example, convection in the tradewind region, are very complicated, and, for simplicity, we consider that an equi-

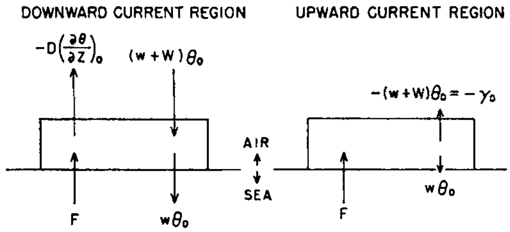


FIG. 5. Schematic representation of boundary conditions.

librium obtains in each of the upward and downward air-current regions as illustrated in Fig. 5. Although it might take too much time and distance for the smaller particle of the giant class to assume the sedimentation-diffusion equilibrium, and hence the state might be always transient, we may regard the summation of these, or the average distributions, as a kind of steady state.

We consider a case where there is neither source nor sink of particles in the air in the downward air-current region, and there is a complete sink of particles at the top of the upward air-current region, where there are usually clouds. This condition is satisfied only when rainout removes particles in the clouds. However, in a case where there is no precipitation, coalescence may still proceed in the clouds, and there will remain a small number of larger giant particles after the clouds evaporate. Consequently, in general, the above condition will be acceptable. In fact, if we assumed no sink at the top of the upward air-current region and particles circulating to the downward air-current region at its top, we would have to consider just a large scale mixing. Thus we obtain $w\theta_0 = F$ (production rate of particles per unit area at the sea surface), $\gamma_0 = 0$ and

$$\theta = \frac{F}{w}\exp\left(-\frac{w+W}{D}z\right) = \theta_2$$

for the downward air-current region, and

$$-\gamma_0 = -(w+W)\theta_0 = F - w\theta_0,$$

and $$\theta = \frac{\gamma_0}{w+W} = -\frac{F}{W} = \text{constant} = \theta_1$$

for the upward air-current region, where it is assumed that $-W > w$. Now we rewrite the velocity of the upward and the downward air currents in terms of W_1 (positive upward) and W_2 (positive downward), and the proportion of these areas is A and B, respectively ($A + B = 1$), and consider a case where an airplane flies horizontally. Thus we obtain an average concentration of particles as

$$\theta = A\theta_1 + B\theta_2$$

$$= \frac{AF}{W_1} + \frac{BF}{w}\exp\left(-\frac{w+W_2}{D}z\right). \qquad (10)$$

When we fix the value W_2 and when A is sufficiently small compared to B, it follows

$$A < B \approx 1, \quad W_1 \gg W_2$$

and $$\theta \approx \frac{F}{w}\exp(-\alpha z), \quad \alpha = \frac{w+W_2}{D}. \qquad (11)$$

The solid curves in Fig. 6 represent the change in α for some values of D and W_2, where w is taken for dry particles. If we assign the value of D, we obtain the asymptotes, and if we assign

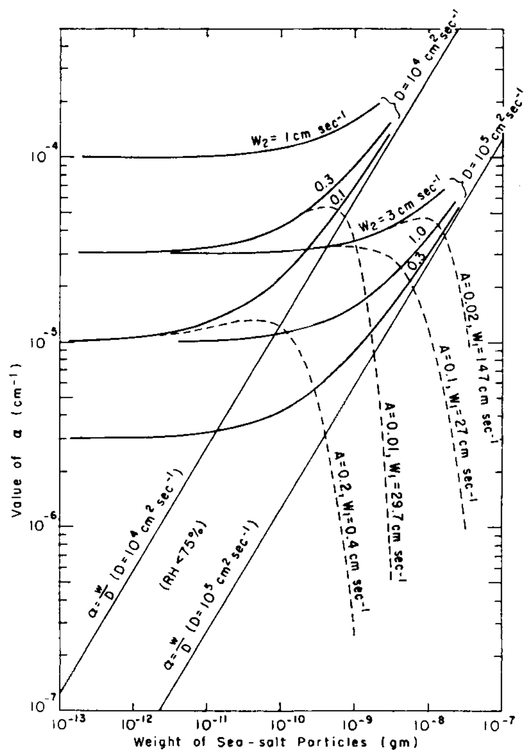


FIG. 6. Some examples of change in α calculated by equation (11) (solid curves) and equation (12) (broken curves).

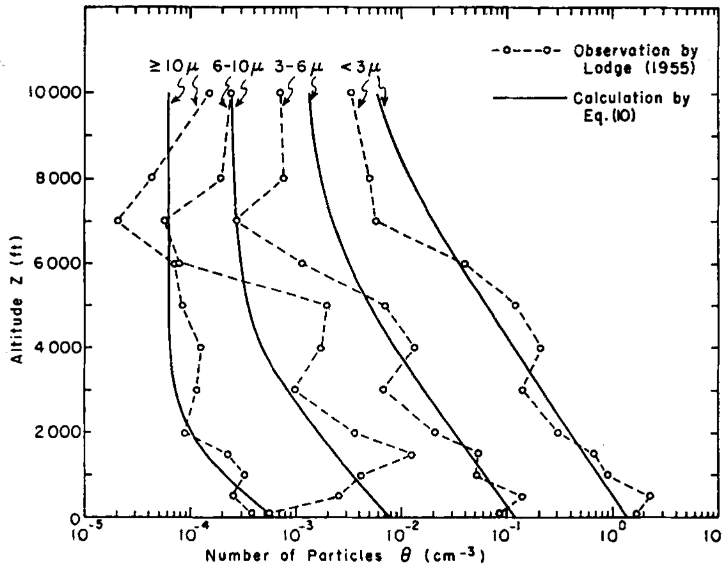


FIG. 7. An example of vertical distribution of giant sea-salt particles calculated by equation (10) and observed by LODGE (1955). In the calculation, $D = 5 \times 10^4$ cm² sec⁻¹, $A = 0.25$, $W_2 = 1$ cm sec⁻¹ (consequently, $B = 0.75$ and $W_1 = 4$ cm sec⁻¹) were assumed.

values of W_2, we obtain the position of the level part of the curves. We may see that the curves in Figs. 2 and 3 seem to fit these curves, for example, curve d in Fig. 3 closely fits a case where $D = 5 \times 10^4$ cm² sec⁻¹ and $W_2 = 0.5$ cm sec⁻¹ in equation (11); and we may consider that in many cases the distribution of giant sea-salt particles over the sea, especially the average distribution, may be approximately expressed by the equation (11). Curve c in Fig. 3 was obtained from data around or above clouds, where the equation (11) does not seem to hold any longer.

For equation (11) to hold, the value of A must be small. That the value of A is small enough means that upward air currents are confined to some very small area. Such small value of A will be also easily attained by a system of convection without roots.

The effect of an appreciable A in the equation (10), however, seems to appear sometimes over the sea. When the value of A is large enough, the distribution of larger particles deviates from the exponential type and approaches a state where the concentration is constant with height in higher levels. An example of the calculated distribution by the equation (10), where the values $D = 5 \times 10^4$ cm² sec⁻¹, $A = 0.25$ and $W_2 = 1$ cm sec⁻¹ (consequently, $B = 0.75$ and $W_1 = 4$ cm sec⁻¹) were assumed, is shown in the solid curves in Fig. 7. The circles in the figure are observed data by LODGE (1955) (from Fig. 10 of his article). The values of F in this calculation, which are entered in Fig. 12 by triangles, were determined so as to fit with the observation. As may be implied from Fig. 7, the value of α, at levels of a few thousand feet, first increases and then decreases with increasing particle weight. Another example of this effect can be seen in curves a and b in Fig. 4. A few examples of the change of α, when the value of A has some effect, were calculated by the following equation:

$$\alpha = \frac{2.3}{z_b - z_a} \log \frac{\dfrac{A}{W_1} + \dfrac{B}{w} \exp\left(-\dfrac{w+W_2}{D} z_a\right)}{\dfrac{A}{W_1} + \dfrac{B}{w} \exp\left(-\dfrac{w+W_2}{D} z_b\right)}, \quad (12)$$

where the calculation was made for $z_a = 700$ m and $z_b = 1400$ m, and are shown by dashed lines in Fig. 6.

It should be mentioned here that if we use a model where the air flowing into the upward-current region from the downward-current region carries sea-salt particles in the concentration of θ_{02} as illustrated by Fig. 8, the equation (10) must only be replaced by

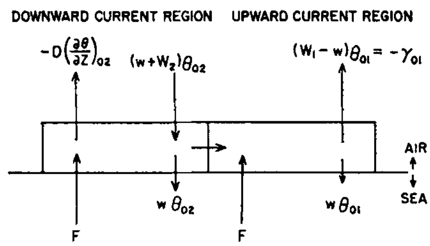


FIG. 8. Schematic representation of a boundary condition.

$$\theta=\left(\frac{1}{w+W_2}+\frac{1}{W_1}\right)AF + \frac{BF}{w+W_2}\exp\left(-\frac{w+W_2}{D}z\right).$$

In this case, the effect of A tends to appear more clearly.

2.2. Phenomenon of surface sink

The next problem concerns the observation that the concentration sometimes increases with height in the lowest levels, even over the sea. LODGE (1955) obtained a mean vertical distribution which could be approximately expressed in an exponential form, but still had an inverted gradient in the lowest several hundred feet. ERIKSSON (1959) reported Woodcock's data showing that the concentration decreases with height in higher wind forces, but increases with height in lower levels in lower wind forces, having a maximum concentration at about the 500-m level. They both attributed the distribution to variation of wind speed from time to time and from place to place. If the reason is, however, that there is little or no production in the region of weak winds, one would expect zero vertical gradient near the sea surface or $(\partial\theta/\partial z)_0=0$. So Eriksson introduced the removal by coagulation, proportional to the concentration, even in the lowest levels, and horizontal transport with vertical shear, to get a maximum concentration at some level.

We may consider, instead, a boundary layer of several meters in thickness on the sea surface, where relative humidity changes sharply with height, with much higher values very near the sea surface, than in the overlying main body of the air. Eddy diffusivity also sharply changes. The actual process of diffusion and evaporation

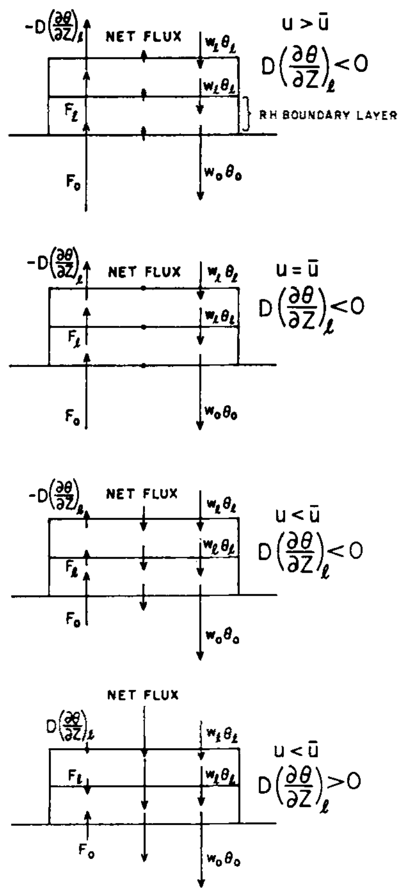


FIG. 9. Schematic representation of the effect of the boundary layer.

of particles in this layer is very complicated, but in a rough consideration, the removal rate of the sea-salt particles at the sea surface may be written as $w_0\theta_0$ where w_0 is terminal velocity of particles which have an equilibrium salt concentration in the air very near the sea surface. Usually $w_0\theta_0>w_l\theta_l$ where the subscript l refers to the top of the boundary layer. If we consider F_l as the effective production rate at the top of the boundary layer, the actual production rate F_0 differs from it according to

$$F_l=F_0-(w_0\theta_0-w_l\theta_l)$$

and as illustrated in Fig. 9. If wind speed fluctuates, F_l, and consequently, $-D(\partial\theta/\partial z)_0$ may become negative in a region of weak wind where $F_0<w_0\theta_0-w_l\theta_l$, even though F_0 is still positive.

TABLE 1. *Reasonable mean values of α determined from Fig. 3.*
Last column in parentheses uncertain.

Weight of dry particles (gm)	10^{-12}	3.2×10^{-12}	10^{-11}	3.2×10^{-11}	10^{-10}	3.2×10^{-10}	10^{-9}	3.2×10^{-9}	10^{-8}
$\alpha(10^{-5}\ cm^{-1})$	1.1	1.1	1.2	1.3	1.5	2.0	3.0	4.0	(4.0)

In other words, in these regions, number concentration near the sea surface increases with height. Since it results from a nonsteady state, an inversion of vertical gradient of the particle concentration is most likely to be found in small particles which have longer residence times.

It should be noted that this type of distribution may be expressed by a form similar to that described in section 3.1, and according to which the maximum concentration should appear at a level of $0.69/\alpha$ (cf. equation 19), which gives a height of several hundred meters, in good agreement with the actual observational data.

Another process which may be considered as a cause of the sea-surface sink is capture of the sea-salt particles by sea waves in their relative motion to the air.

2.3. Estimate of effective production rate of giant sea-salt particles at the top of the boundary layer

In this section an estimate of the effective production rate of giant sea-salt particles at the top of the boundary layer of several meters in thickness on the sea surface for various wind forces is obtained. Some efforts to obtain the change in α in the equation (1) or (11) with wind force or some other factors were attempted but no definite information was obtained. So the concentration of particles several meters above the surface θ_l was calculated by equation (1) with the reasonable mean values of α listed in Table 1, which were determined from Fig. 3, from Woodcock's 1953 sea-salt particle data for various wind forces (Fig. 1 of his article), Woodcock's data being assumed to have been taken at the 700 m level. The results are shown in Fig. 10 as a function of both particle weight and of wind speed. A comparison of θ_l in Fig. 10 with data by MOORE (1952) and by FOURNIER D'ALBE (1951), which were obtained from an observation on board a ship in the Atlantic Ocean and in the Bay of Monaco respectively, is shown in Fig. 11 as a reference. In view of the complexity in the actual concentration in the boundary layer, especially for lower wind speeds, the agreement is not bad. The effective production rate at the top of the boundary layer, $F_l = w_l\theta_l$, was calculated from Fig. 10 with values of w_l for 80 % RH, because the relative humidity at 10 m level is about 80 % on the average for those tradewind areas where Woodcock's 1953 measurements were mainly made. The results are shown in Fig. 12. This

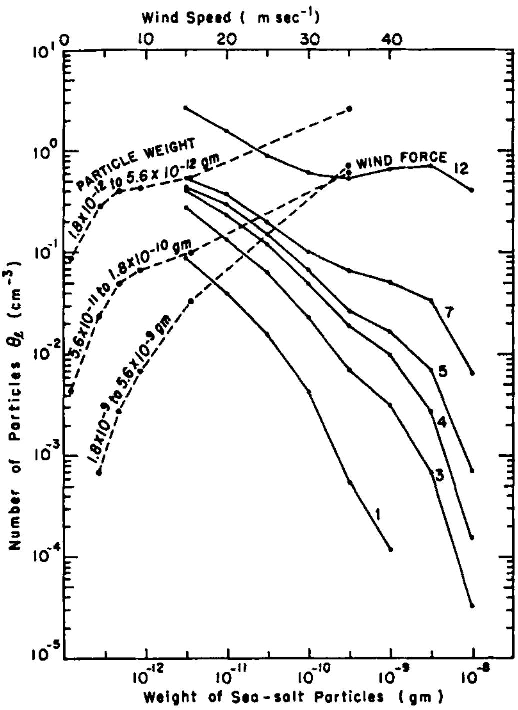


FIG. 10. An estimate of effective production rate of sea-salt particles at the top of the boundary layer, 10 m level (see the context). Triangles entered are values used in the calculation of Fig. 7 (see the context in 2.1). All values are entered for particle weight range of log (particle weight) = 0.5.

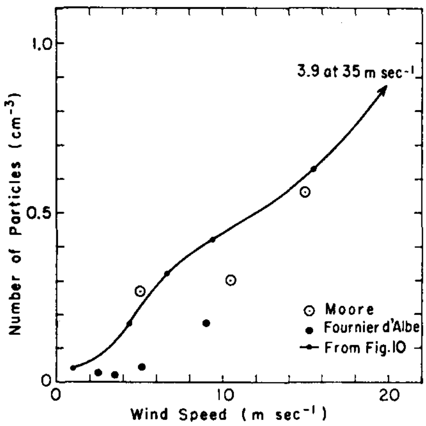


FIG. 11. Comparison of concentration of sea-salt particles larger than 10^{-11} gm weight at 10 m level obtained from Fig. 10, with data by MOORE (1952) and FOURNIER D'ALBE (1951).

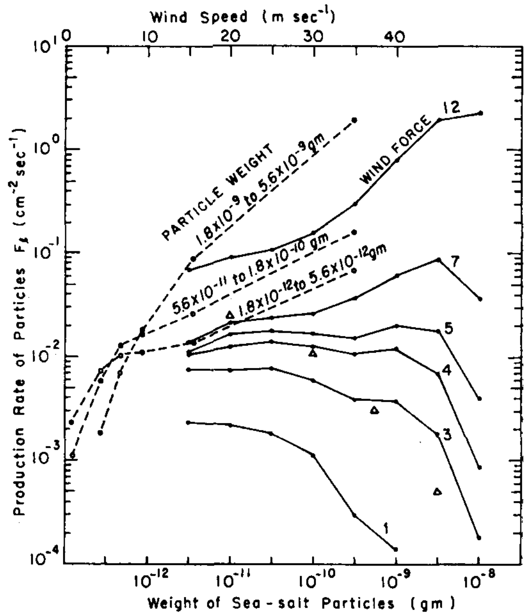


FIG. 12. An extrapolated value of concentration of sea-salt particles at the top of the boundary layer, 10 m level, by use of α, from Woodcock's 1953 data. All values are entered for particle weight range of log (particle weight) = 0.5.

estimate will be valid for ocean areas where the structure of the boundary layer (humidity distribution, etc.) is similar to that of the tradewind region.

It is noted that the curves in heavy lines in Figs. 10 and 12 have a trough in the vicinity of the weight of particles of 10^{-10} to 3.2×10^{-10} gm. The existence of the trough in the curve of the production rate was pointed out by TOBA (1961) as a result of his wind-flume experiment, and it seems to indicate that particles larger than 10^{-10} to 3.2×10^{-10} gm in weight (around 20 μ in original drop diameter) are mainly produced through the disintegration of a water jet formed upon collapse of a bubble cavity, and smaller particles are dominantly produced from shattering of the bubble film, as suggested by TOBA (1961) although the location of the trough is displaced compared with that found by the wind-flume experiment. One reason for the discrepancy might be the fact that Toba's experiment was carried out by using tap water instead of sea water, and another will be that Figs. 10 and 12 were reduced from data taken at a level of several hundred meters and are valid for the top of the boundary layer. In other words, a complicated process in the boundary layer seems to present a gap between the present estimate and the real production of particles at the immediate air-sea boundary.

It should also be mentioned that the wind dependence of the production rate or the concentration of giant sea-salt particles, appearing in Figs. 10 and 12, have different slopes with the particle weight. The slope in the present estimate is smaller than TOBA's (1961) especially for smaller particle weights. One reason will again be attributed to the process in the boundary layer, but another reason may be inferred: that the equilibrium state for each wind speed indicated was not attained for smaller particles when the observation was carried out. A further study on these points is left for the future.

2.4. RESIDENCE TIME OF GIANT SEA-SALT PARTICLES OVER THE OCEAN

Since the distribution of giant sea-salt particles over the ocean is expressed as a mean, in an exponential form, the residence time, τ, of the particles above the boundary layer of several meters may be estimated by using the value of α in Table 1 by

$$\tau = \frac{1}{w_l \theta_l} \int_0^\infty \theta_l \exp(-\alpha z) dz = \frac{1}{\alpha w_l}.$$

The values were calculated by using two tentative values of w_l for 80 % RH, and for 91.4 %

TABLE 2. *Residence time of giant sea-salt particles over the oceans calculated by the α in Table 1.* The lower part contains the estimate of the residence time by ERIKSSON (1959).

Weight of dry particles (gm)	10^{-12}	10^{-11}	10^{-10}	10^{-9}
τ (days) (w_l for 80 % RH)	86	17	2.9	.32
τ (days) (w_l for 91.4 % RH)	59	11	2.1	.23
Eriksson's estimate (from fallout)	82	16	2.6	.4
Eriksson's estimate (from production)	3.5	1.0	.6	.5

RH. They are tabulated in the upper half of Table 2. From the table it is apparent that the number of salt particles larger than about 10^{-10} gm must be subject to meteorological control or production rate at the time, but smaller ones have very long lifetimes and represent a long-period geophysical balance.

ERIKSSON (1959) estimated the residence time as shown in the lower part of Table 2 from an estimate of both fallout rate and production rate. The first estimate has a very similar trend with ours. But he stated that the values were extremely large for the small-sized particles, and that this was incompatible with the observed variability of particle numbers of these particles, and consequently, that his estimate from fallout had something wrong with it. We think that our estimate is correct; it seems to show that air parcels do not mix completely for some extended time, and that in the atmosphere there are many such salt-laden or salt-free small sections of air wandering or drifting about for a long time.

3. Distribution of giant sea-salt particles over the continent

3.1. MAIN FEATURES OF THE DISTRIBUTION AND ESTIMATE OF THE MAGNITUDE OF THE GROUND SINK

The striking characteristic of the distribution of giant sea-salt particles over the continent is, according to BYERS *et al.* (1957) that the particle concentration is fairly constant with altitude, with a sharp reduction in the lowest several hundred feet. Byers *et al.* speculated that the impaction by trees and other vegetation produced an important ground sink. The horizontal distribution also did not show a distinct decrease with distance inland at levels of 2000 to 5000 ft. JUNGE & GUSTAFSON (1957) interpreted the fairly uniform vertical distribution and the decrease of chloride concentration in rain water within 500 miles from coast, while roughly uniform chloride concentration persisted in the main part of the continent, as representing a transition from the distribution over a source region at sea to a nonsource region of the continent, emphasized by stronger convection over the continent. In the course of the discussion, they stated that washout by rain gave a half life of giant sea-salt particles over the continent of 3 or 4 days, and dry fallout was about 25 % of the washout, but an estimate of the effect of impaction by trees and other ground obstacles was not given. A further study of the distribution of the particles far inland, including the ground sink, is undertaken in this section.

In equation (8) we now consider a case where γ (the vertical flux, positive downward, of the particles) varies with height, that is, a nonsteady state. We use a symbol w' instead of $w + W$ and consider that it may contain some quantities in addition to fall velocity of the particles, but we assume for the sake of simplicity that at $z = 0$, w' has the same value as the sedimentation velocity. For boundary conditions, we assume that at $z = 0$

$$\gamma = \gamma_0 > w'\theta_0$$

or
$$D\frac{\partial\theta}{\partial z} = \gamma_0 - w'\theta_0,$$

where γ_0 is the total flux of particles into the sink at the ground, and at $z = \infty$,

$$\theta = 0$$

or
$$\gamma = 0.$$

Then the solution, containing γ, of the equation is as follows:

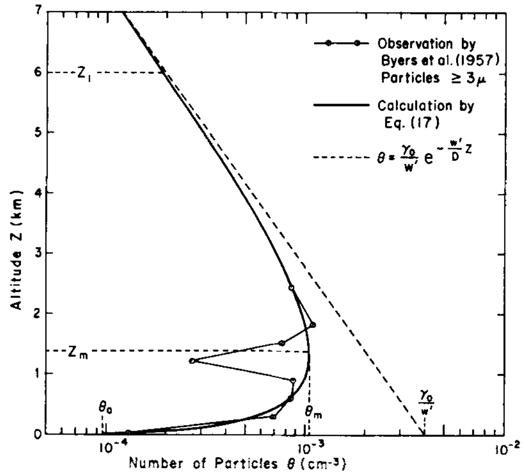


FIG. 13. An example of vertical distribution of giant sea-salt particles inland, calculated by equation (17), and observed by BYERS *et al.* (1957). In the calculation values $w' = 1.0$ cm sec^{-1}, $D = 2 \times 10^5$ cm^2 sec^{-1}, $\lambda = 0.08$ and $u = 5$ m sec^{-1} were assumed.

$$\theta = \frac{\gamma_0}{w'} \exp\left(-\frac{w'}{D}z\right) - \left\{\frac{\gamma_0}{w'} \exp\left(-\frac{w'}{D}z\right) - \theta_0\right\} \exp\left(-\frac{w'}{D}z\right)$$

and
$$\gamma = \gamma_0 \exp\left(-\frac{2w'}{D}z\right). \tag{13}$$

Equation (13) expresses the shape of the vertical distribution curve, as shown in Fig. 13, for the condition far inland.

The value of γ_0 changes with time because the total number of particles contained in the air column decreases. The change of γ_0 with time may be obtained after writing the integral

$$\gamma_0(t)\Delta t = \int_0^\infty \{\theta(z,t) - \theta(z, t+\Delta t)\} dz = -\frac{\Delta}{\Delta t}\left\{\int_0^\infty \theta(z,t)\, dz\right\} \Delta t.$$

Now we introduce a somewhat artificial concept of the efficiency of impaction by ground obstacles, λ, defined by

$$\lambda u \theta_0 = \gamma_0 - w'\theta_0, \tag{14}$$

where u is the uniform horizontal wind velocity. Taking into account equation (14), one notes that the above expression for $\gamma_0(t)$ leads to the following differential equation:

$$\frac{d\gamma_0}{dt} = -\frac{w_*^2}{D}\gamma_0,$$

where w_* is such that

$$\frac{1}{w_*^2} = \frac{1}{2w'^2} + \frac{1}{w'(\lambda u + w')}$$

or
$$w_* \approx \sqrt{2}\, w'. \tag{15}$$

The solution is

$$\gamma_0(t) = \gamma_{00} \exp\left(-\frac{w_*^2}{D}t\right), \tag{16}$$

where γ_{00} is the value of γ_0 at $t = 0$.

From equations (13), (14) and (16) we finally obtain the following solution

$$\theta = \gamma_{00} \exp\left(-\frac{w_*^2}{D}t\right)\left[\frac{1}{w'} \exp\left(-\frac{w'}{D}z\right) - \left\{\frac{1}{w'} \exp\left(-\frac{w'}{D}z\right) - \frac{1}{\lambda u + w'}\right\} \exp\left(-\frac{w'}{D}z\right)\right]. \tag{17}$$

An example of this type of distribution was provided by BYERS *et al.* (1957) in Fig. 22 of their article. One of the observed curves is reproduced here in Fig. 13.

We now examine some characteristics of the distribution curve of equation (17). The maximum concentration appears at z_m where $\partial\theta/\partial z = 0$, that is

$$z_m = -\frac{D}{w'} \ln\left(\frac{1}{2} + \frac{w'}{2(\lambda u + w')}\right) \approx -\frac{D}{w'} \ln \frac{1}{2} \approx 0.69 \frac{D}{w'}. \tag{18}$$

If we put $w'/D = \alpha$, which is the factor representing the exponential decrease of the concentration with height in an equilibrium distribution at sea, then it follows that

$$\alpha \approx \frac{-\ln \frac{1}{2}}{z_m} \approx \frac{0.69}{z_m}. \tag{19}$$

In the equilibrium distribution at sea z_m would be at the surface. Let us take a height z_1 where the concentration is less than 5 per cent of this value. The distribution places it at $z_1 = 3/\alpha$. Then, from (19) the height of the maximum over the continent is at

$$z_m \approx 0.23\, z_1. \tag{20}$$

The maximum concentration θ_m is obtained by

$$\theta_m = \frac{\gamma_0}{2}\left(\frac{1}{2w'} + \frac{1}{\lambda u + w'} + \frac{w'}{2(\lambda u + w')^2}\right) \approx \frac{\gamma_0}{4w'}. \tag{21}$$

The rario of the maximum concentration to that near the ground is

$$\frac{\theta_m}{\theta_0} = \frac{\lambda u}{4w'} + \frac{3}{4} + \frac{w'}{4(\lambda u + w')} \approx \frac{\lambda u}{4w'}. \tag{22}$$

Although the data by BYERS *et al.* (1957) do not clearly show the difference in the ratio with particle size, and some soundings in other days largely deviate from the distribution curve represented by equation (17), it seems general that the ratio θ_m/θ_0 is of the order of 10 to 100, if we compare the data by Byers *et al.* with ground data by REITAN & BRAHAM (1954). Consequently, the approximations in equations (15) and (18) through (22) are now substantiated. From equation (22) it is also seen that the ratio between the terms $\lambda u \theta_0$ and $w'\theta_0$ is $4\theta_m/\theta_0$, that is, about 40 to 400. So the effect of dry fallout in the total ground sink is very small. It should be noted, however, that the term $\lambda u \theta_0$ may possibly include, besides the impaction by ground obstacles, the effect of humidity change near the ground, which could not be expressed in the term $w'\theta_0$ in the above equations, but may cause a large amount of removal of particles by fallout. If we assume values $u = 500$ cm sec^{-1} and $w' = 1$ cm sec^{-1}, the λ will become 0.08 to 0.8 and the magnitude of the ground sink will be $\gamma_0 \approx \lambda u \theta_0 = (40 \text{ to } 400)\theta_0$.

From the above observation data, z_m seems to lie between 1 and 2 km; consequently, z_1 from equation (20) becomes several km. This means that sea-salt particles are expected to exist up to several km over the continent. From equation (19) the α turns out to be approximately 5×10^{-6} cm^{-1}, which is nearly 1/4 of the mean value over the sea, although the change in α with particle weight does not appear clearly in the data. This seems to imply that the eddy diffusivity D becomes larger as the air flows into the continent. This point will be treated in the next section.

An example of the calculated curve by equation (17), where values of $w' = 1.0$ cm sec^{-1}, $u = 500$ cm sec^{-1}, $\lambda = 0.08$ and $D = 2 \times 10^5$ cm^2 sec^{-1} were used, is shown in Fig. 13.

The vertical distributions obtained by PODZIMEK & ČERNOCH (1961) over Czechoslovakia, however, have z_m of 300 to 600 m, and z_1 of about 2 km. Although they regard the distribution as mainly that of salt of land origin, if we consider this still as an example of our model of the distribution, the α is rather large in this case. The difference seems to imply that we must take into account meteorological conditions for particular areas.

Recently published observations made by MÉSZAROS (1964) of chloride particles over Hungary showed maximum concentration at from several hundred to two thousand meters, and the ratio θ_m/θ_0 seemed also 10 to 100 when there was no thermal inversion layer. Because of the fact that vertical profiles of atmospheric aerosols including various substances, taken in the same flight with the chloride particle observations, had a maximum concentration at the ground, he concluded that the chloride particles were not of local origin.

3.2. TRANSITION FROM THE DISTRIBUTION OVER THE SEA TO THAT OVER THE CONTINENT, AND THE RESIDENCE DISTANCE OVER THE CONTINENT

In the example treated in the last section, the value of α seems to be smaller than that over the sea by a factor of four or so. JUNGE & GUSTAFSON (1957) dealt with this effect by simply calculating the effect of vertical mixing while neglecting the fall velocity of the particles. If we take into account the w' with no ground sink, the decrease in w'/D by a factor of 4 results in the decrease in the ground concentration by a factor of 4. At the same time, however, the change in the value of w'/D must be accompanied by a change in the shape of the distribution curve brought about by the ground sink. It is not easy to deal analytically with the combined effects. In this section, the effect of the ground sink only is treated for a simpler case.

We consider a case where, for simplicity, γ is again constant and use an equation similar to equation (9):

$$\theta = \frac{\gamma_0}{w'} - \left(\frac{\gamma_0}{w'} - \theta_0\right) \exp\left(-\frac{w'}{D}z\right). \tag{23}$$

In this case the sum of the eddy diffusion of the particles, $D\,(d\theta/dz)$ and the transport by w', $w'\theta$ is always equal to γ_0. It is also assumed that a quasi-steady state always holds, consequently, the distribution at any time is expressed by equation (23) with θ_0 changing with time. Then the excess of the removal by the ground sink to the downward flux γ_0 makes the distribution curve shift to the direction of a smaller θ_0, and it follows that

$$\{(\lambda u + w')\theta_0(t) - \gamma_0\}\Delta t$$

$$= \int_0^\infty \{\theta(z,t) - \theta(z, t+\Delta t)\}\,dz$$

$$= -\frac{\Delta\theta_0(t)}{\Delta t}\Delta t \int_0^\infty \exp\left(-\frac{w'}{D}z\right) dz$$

to give the following differential equation:

$$\frac{d\theta_0(t)}{dt} = -\frac{w'}{D}(\lambda u + w')\theta_0(t) + \frac{w'\gamma_0}{D}.$$

The solution is

$$\theta_0(t) = \gamma_0\left[\left(\frac{1}{w'} - \frac{1}{\lambda u + w'}\right) \cdot \exp\left\{-\frac{w'}{D}(\lambda u + w')t\right\} + \frac{1}{\lambda u + w'}\right]. \tag{24}$$

Replace t with $x = ut$, then the distance x_1, where $\theta_0(t)$ nearly assumes its final value, $\gamma_0/(\lambda u + w')$, is obtained as

$$x_1 = \frac{3Du}{w'(\lambda u + w')} \approx \frac{3D}{w'\lambda}.$$

If we use the values of $w' = 1$ cm sec^{-1}, $\lambda = 0.08$ to 0.8 and $D = 2 \times 10^5$ cm^2 sec^{-1} as before, we obtain $x_1 = 75$ to 7.5 km.

The decrease of γ_0 treated in the last section, equation (16), gives the distance x_2 where $\gamma_0(t)$ is reduced to a few per cent of the original value, γ_{00}, of

$$x_2 \approx \frac{3Du}{2w'^2}.$$

With the same values of D and w, and $u = 500$ cm sec^{-1}, we obtain $x_2 = 1500$ km. Consequently our treatment, where the changes in γ_0 in the last section and in θ_0 in this section were separated, is now reasonable.

Some data of the horizontal distribution of giant sea-salt particles over the continent at the 2500 ft level were taken from the Gulf of Mexico to the north by BYERS *et al.* (1957), while the air flowed into the continent from the Gulf (Fig. 10 of their article). Since there is a map of the air trajectories showing very simple patterns for 54 hours prior to the flight (Fig. 8 of their article), we may consider that the data well represents the change in γ_0 as expressed by equation (17).

The value of the exponential factor w_*^2/D of approximately (2 to 3) $\times 10^{-5}$ sec^{-1} was obtained from the data, while the above values of w' and D give 10^{-5} sec^{-1}. The former value must contain also the effect of rainout. The latter has nearly the same value as a factor obtained by JUNGE & GUSTAFSON (1957) for rainout alone, so the combining of the two 10^{-5} sec^{-1} factors brings us to the value of w_*^2/D.

It may be concluded that as the air flows into the continent, the eddy diffusivity increases in some cases by a factor of four or so, and the giant sea-salt particles are transported upward to several km; on the other hand the concentration near the ground decreases because of the strong ground sink which will mainly be from the impaction on ground obstacles. So the distribution type appears which has a maximum concentration somewhere between several hundred meters and about 2 km and shows little difference in the concentration for a few km in the vertical direction, provided there is no rainout which may locally reduce the concentration. This type of distribution is attained within some tens of km from the coast, after which the concentration slowly decreases for a few thousand km, if the removal mechanism is restricted to the ground sink. The effect of rainout may be of a magnitude similar to the ground sink. Because of the small amount of data available, however, the numerical details have yet to be determined.

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