# On the Distribution of Sea Salt over the United States and its Removal by Precipitation

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## *Abstract*

Average maps for summer and winter of the distribution of chloride concentration in rain water over the United States are presented, and an attempt is made to explain them quantitatively. From various considerations, it must be concluded that the essential features of these maps – the drop in the $Cl^-$ concentration along the coast and a constant level inland – are determined by large scale vertical mixing in the troposphere rather than by washout. Large scale washout appears to be rather inefficient, so that even large and hygroscopic particles, such as sea spray, can move across continents without being effectively removed from the atmosphere.

## Introduction

This paper is based on information derived from chemical analyses of rain water samples routinely collected in a network covering the United States and part of the Atlantic Ocean (JUNGE and GUSTAFSON, 1956). Of the eight components quantitatively determined, the variation of the concentration of $Cl^-$ has been selected for discussion here because of its specific interest to the fields of air chemistry and cloud physics. In contrast to other chemical components in rain water, $Cl^-$ has but one important source, the ocean, with sharp and well-known borderlines. Because of this fact, some conclusions about the large scale distribution of aerosols in the troposphere and their removal by precipitation have been deduced and are presented.

## The Sampling Network

At each of sixty-two stations in the United States and six others on islands and weather ships in the Atlantic, the rain samples are collected in a plexiglass funnel which drains into a polyethylene bottle. A wooden box totally encloses these parts except during periods of precipitation, when a cover is removed. This procedure is certainly necessary in dry, dusty locations such as in the Southwestern United States, to study the rain water content separately from the dry fallout; but even in areas with more regular rainfall throughout the year, the dry fallout is not negligible compared to the amount brought down by rain. This was shown by tests near Boston, Massachusetts, where two collectors were exposed side by side; one open in precipitation only, and the other open continuously. On the average, the open gauge collected twenty-five per cent more $Cl^-$ as a result of the dry fallout.

The disadvantage of the "cover normally on" procedure is that some rainfalls may be missed by the observers, particularly in the case of brief, heavy showers. However, errors introduced in this way are essentially random and may lead to higher scattering of the values, but likely not to systematic errors.

The sampling sites were selected by avoiding local influences of large cities and industrial areas so as to obtain representative samples. The results obtained indicate that this was achieved to a high degree because even the $SO_4^{--}$ component, which is known to be a major local industrial pollutant, shows only consistent *regional* variations.

With the exception of the protection of the funnel in dry weather, the sampling procedure was cooperatively planned to be identical with that of the Scandinavian and European network, so as to make the results comparable (ERIKSSON, 1954). According to this plan, the samples from each station each month are batched and subsequently analyzed. Thus, the values reported are the monthly averages of the $Cl^-$ concentrations in the present case. The $Cl^-$ content was turbidimetrically determined after the addition of silver nitrate to an acidic solution. An accuracy of about ±15 per cent was achieved for concentrations less than 1 mg/l.

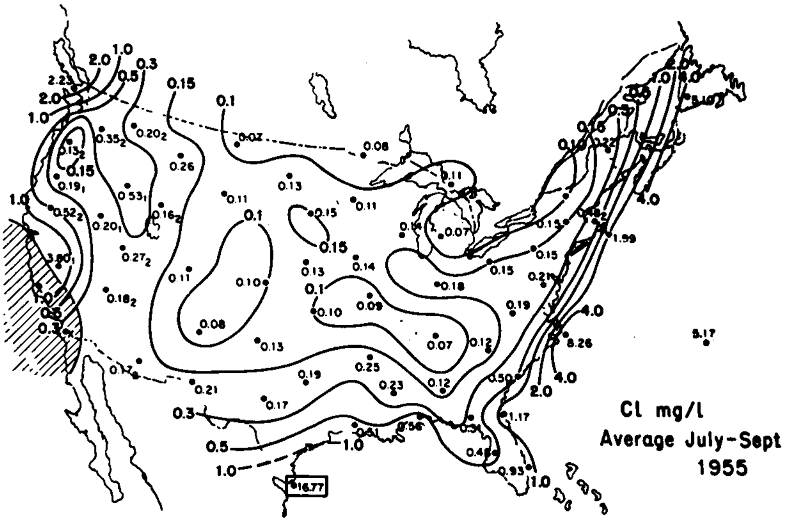

Fig. 1. Average $Cl^-$ concentration (mg/l) in rain water, July—September, 1955. The subscripts indicate the number of months upon which the average was based. No subscript indicates three months. The hatched area indicates no rainfall or insufficient sample.

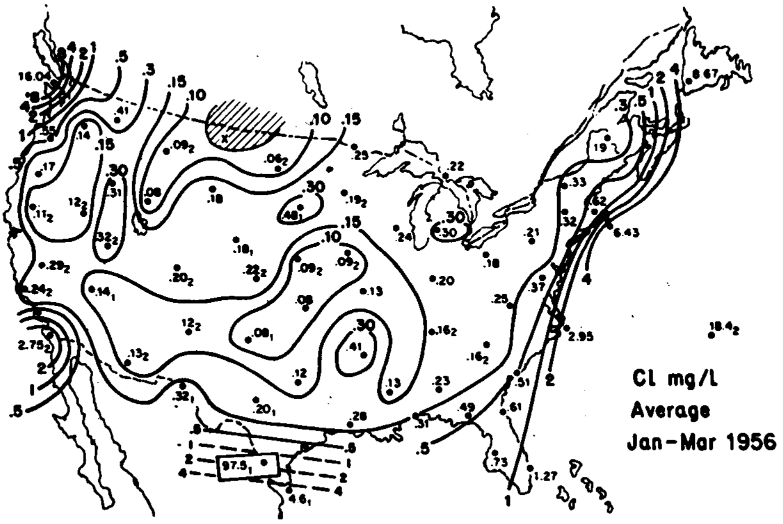

Fig. 2. Same as Fig. 1, but for the months January—March, 1956.

## Results

As this is written, twelve monthly maps are available. The pattern of the $Cl^-$ concentration is strikingly similar in this series and the essential features are well shown on the summer and winter average maps (Figs. 1 and 2). The concentration decreases rapidly with increasing distance from the coast, tending toward a constant level inland. This level is slightly higher in the winter than in the summer season, but otherwise the two maps show little difference. In summertime, the fairly constant level inland is well exhibited on the monthly maps and is not due to the averaging process. In the winter months, a few tongues of high salt concentration protrude inland from the coasts and these are smoothed somewhat in the three-month average.

Since $Cl^-$ is a minor industrial pollutant, no influence of industrial areas on the $Cl^-$ distribution was expected or found. One local *natural* source of $Cl^-$ over land was evident in the Brownsville, Texas region. Dust from coastal salt flats south of the station raises the $Cl^-$ level to such an extent as to make the values worthless. When this was first determined, an additional site at Laredo, Texas, 170 miles NW of Brownsville, was inaugurated, but even this station was influenced some months. High values at such places as Tatoosh Island, Wash., Cape Hatteras, N.C., and the ocean stations are, of course, accounted for by heavy production of sea spray. For this study, such abnormalities are of minor importance.

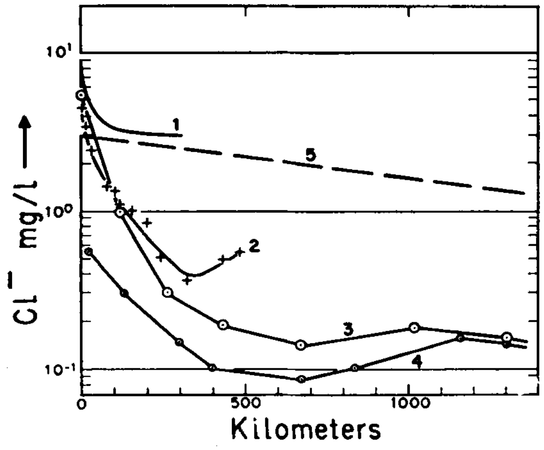

Fig. 3. $Cl^-$ concentration in rain water as a function of the distance from the coast.
Curve 1, according to Leeflang, for the Netherlands,
Curve 2, according to Emanuelsson et al. for Sweden,
Curves 3 and 4, cross sections from Cape Hatteras, N.C., to northwest and from Mobile, Ala., to north (United States). Average July—September 1955.
Curve 5, calculated decrease due to washout only, under conditions similar to those for curves 3 and 4. The initial concentration was arbitrarily assumed to be 3 mg/l.

Due to insufficient amounts of rainfall and the widely varying topography, the concentration distributions found in the western part of the U.S. do not give as consistent a picture there as in the eastern and southern portion. Our conclusions will be concerned mainly, therefore, with the latter part of the country.

The summer average $Cl^-$ concentrations as a function of distance from the coast along a line northward from Mobile, Alabama, and along a line northwestward from Cape Hatteras, N.C., are plotted in Fig. 3. The rapid decrease of concentration ceases at about 500 km from the coast. It should be noted, however, that the pattern of this drop is not very precise, because the network is not dense enough. The winter values are similar. For comparison, data available for other parts of the world are included in the figure. The profile along a line northeasterly from Göteborg, Sweden, paralleling the steepest gradient, was obtained from the annual average $Cl^-$ concentration map given by EMANUELSSON ET AL. (1954). There is agreement with the U.S. data up to about 300 km but beyond there the Swedish values rise again, due to the influence of the Baltic Sea. LEEFLANG (1938) gives values showing a rapid coastal drop which soon levels off to about 3 mg/l. Such high values are also indicated for places farther inland in continental Europe (e.g., LIESEGANG, 1934) and are incorporated in a tentative map by ROSSBY and EGNÉR (1955) of $Cl^-$ concentration in the rainfall of Europe. This chart shows high values of about 3 mg/l over Central Europe which drop over Sweden to a level comparable to the inland concentration of the U.S. It is probable that the high Central European values are due to the extreme density of population and industry, but this has to be confirmed by further measurements. Data for the coast of Brittany by FACY (1931) are not included in Fig. 3 because the shape of the coastline makes it difficult to interpret the data as a function of distance from the coast and to compare them with the other results.

The quasi-constant level of $Cl^-$ concentration inland implies that the concentration is independent of the total amount of rainfall. This was checked by plotting a scatter diagram of the $Cl^-$ concentration vs. the monthly total rainfall for all inland stations. No relationship between these two variables could be detected. On the basis of experience pertaining to radioactive matter in rainfall, this single fact indicates that, over the continents: the sea salt particles must be uniformly distributed in the troposphere; the salinity of rain water reflects the actual $Cl^-$ concentration in the troposphere; and the airborne salt particle concentration is not greatly depleted by rainfall.

## General Explanation of Results

It is very tempting to explain the decrease of $Cl^-$ concentration inland as an effect of the washout by rain. But this interpretation fails to explain why the $Cl^-$ drop levels off at a low value inland. It may be argued that the observed constant level is produced by additional continental $Cl^-$ sources. Large cities and industrial areas are the only known continental sources, and their output of $Cl^-$ is small compared to other pollutants, $SO_2$ for instance. But the maps indicate no higher values in the industrial areas of the Northeastern U.S., which would be expected especially during wintertime when more coal is burned and the accumulation of pollutants is higher, due to more stable weather conditions. With the higher density of population and industry in Europe it might be different there, as suggested above. A fairly widespread gaseous $Cl^-$ component was recently found in the air by JUNGE (1956), but quantitative considerations make it very unlikely that the continental $Cl^-$ level in rain is the result of a fixation of this component.

Thus it appears that human activities or natural sources cannot explain the observed constant $Cl^-$ level. The most probable explanation results from consideration of Fig. 4, wherein are compiled reliable data on the vertical distribution of $Cl^-$ particles. Over the ocean in the trade wind regions of Florida, Australia, and Hawaii, WOODCOCK (1953) has made careful measurements. Although most of his data were obtained in Hawaii, there is strong support for the extension of his findings to cover the Gulf of Mexico region in the summertime. His counts, labeled "W" on Fig. 4, show a rapid decrease in salt particles above the inversion layer. Below 0.5 km where he made no measurements, the values are doubled to cover the slight increase normally found in these layers.

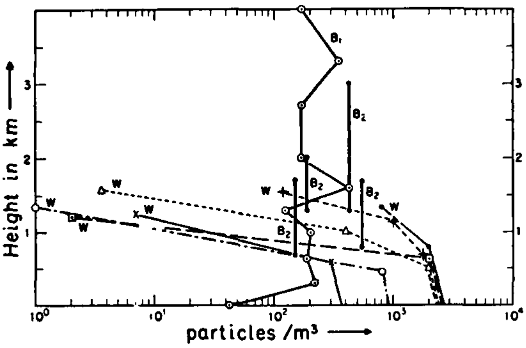

Fig. 4. Vertical distribution of the number of sea salt particles having a dry radius $\geq 3\mu$ (equivalent to a radius of $7\ \mu$ at 80 per cent relative humidity and a mass of $5 \times 10^{-10}$ gr). The curves marked W were measured by Woodcock (1953) in regions of trade winds; the portion of the curve between the ocean surface and the measured values around 0.5 km is assumed. The values show a rapid decrease at the level of the trade wind inversion. The curves marked B are given by Byers (1955). $B_1$ is an average of three soundings made in Illinois. $B_2$ are average concentrations for four overland flights southward from Chicago. The length of the bar indicates the altitude range during the flight.

BYERS (1955) gives data on the vertical distribution of giant sea salt particles from measurements during several flights near Chicago and from there to the South. The average of three vertical soundings ($B_1$ of Fig. 4) and the values along the individual overland flights ($B_2$) indicate a fairly uniform vertical distribution of salt particles up to 4 km which may well extend to higher levels. The character of the maritime and continental distributions suggests that convective activity over land is the cause of the transition. This is substantiated by the comparison of the total number of particles over a unit area. If Byers' distribution is assumed to be constant to 7.5 km (3/4 of the troposphere) there is a total number of $2.3 \times 10^6$ particles per $m^2$, whereas Woodcock's values give an average of $1.8 \times 10^6$. Evidently, the transition from the vertical distribution of particles at the maritime source to that of the continental distribution is not accompanied by substantial washout. *We postulate that the rapid decrease of $Cl^-$ content in rain water near the coast reflects primarily the decrease in particulate matter in the lower layers due to vertical mixing, and that the uniform level throughout the country reflects the resulting uniform horizontal and vertical distribution of particles.* We will try to verify this by quantitative considerations.

## Removal of Aerosol Particles and Washout

Three major processes contribute to the removal of sea salt from the atmosphere: (1) fallout due to gravity; (2) impaction of particulate matter on obstacles; and (3) washout by precipitation.

*Fallout* under the force of gravity is most important for airborne particles larger than 10—20 $\mu$. However, these particles are deposited, for the most part, near their sources. Smaller particles remain airborne for rather long times (JUNGE, 1955). Thus, the total dry deposit is not very important when compared with washout by precipitation, about 25 per cent of the latter, as noted before.

*Impaction* seems to be a more efficient process. ERIKSSON (1955) concludes that the $Cl^-$ content of river water in Sweden can be supplied only by the atmosphere. However, the total amount of Cl brought down by *rain water* is less than the runoff, and there is some evidence that a considerable amount of aerosols is combed from the air by trees and bushes. Eriksson refers to some measurements of rain water composition under trees which show much higher inorganic ion concentrations than the rain which did not touch the trees, especially at the onset of rain. The decrease of $Cl^-$ particles in the surface layer as shown in Fig. 4 was attributed by Byers to the same process. Although more reliable information is needed, it appears that this collection by obstacles is quite efficient. However, the world-wide geochemical significance of this effect is hard to estimate, and is seriously minimized by the fact that the greater portion of the earth's surface has no vegetation.

*Washout* by precipitation is probably the most important process whereby aerosols are removed from the atmosphere. The term "washout" as used here embraces three mechanisms: (1) removal of those particles which act as nuclei in the *condensation* process; (2) the removal by *agglomeration* of particles with cloud droplets; and (3) the removal by *collection* of particles by falling raindrops.

Before we consider these processes in more detail, a few words might be necessary about the size distribution of sea spray particles. About 98 per cent of the *total mass* of the sea spray particles in marine air is found in the particle sizes above 0.8 $\mu$ radius, while the *most numerous* salt particles have radii between 0.1 and 0.8 $\mu$. Insofar as we know now, the concentration of sea spray particles smaller than 0.1 $\mu$ is negligible in terms of mass *and* number.

In general, sea salt nuclei are used up rather completely by the *condensation* process during the formation of clouds. This is surely true over the oceans where the total number of condensation nuclei is of the same order as the number of cloud droplets (a few hundred per $cm^3$). Therefore, almost no nuclei are left unused in the cloud volume, a fact which indeed has been observed (WOODCOCK, private communication). It is noted that under normal conditions the total number of *sea spray* particles does not exceed about $10/cm^3$, so that, even over the oceans, the majority of nuclei must be assumed to be of continental or other origin (JUNGE, 1956). These relatively few but very large hygroscopic sea spray particles are preferentially used in cloud and raindrop formations over the oceans, as shown by WOODCOCK and BLANCHARD (1955). Over the continents, a large number of other hygroscopic nuclei are present in all size ranges. Since here the total number of nuclei larger than 0.1 $\mu$, which are preferentially used as "active" condensation nuclei in cloud formation, is of the same order as the number of cloud droplets, and since almost all the sea spray particles are larger than this size, it is very likely that a high percentage of them is used even over the continents.

The *agglomeration* of aerosol particles with cloud droplets is of importance only for those size ranges below 0.1 $\mu$ in which Brownian movement is important. Since the sea spray particles are larger, this process is of little importance for their removal.

The third removal process, *collection* of aerosol particles by falling raindrops, applies only to particles larger than about 1 $\mu$. Since most of the sea salt is found in particles larger than this size, it is of considerable importance to our problem. Small amounts of rainfall are sufficient to remove the major portion of sea spray particles below the clouds (JUNGE, 1953, TURNER, 1955). Since over the oceans and in coastal areas most of the sea spray particles are concentrated in the layers below cloud base (Fig. 4), the collection by falling raindrops is decisive for the salinity of rain water in these areas. Actually, the observed concentrations of $Cl^-$ in rain water there can be explained quantitatively by this process alone. If the aerosols are more uniformly distributed throughout the whole troposphere, this process becomes of less importance, however, than that of condensation, as will be shown later.

## Calculation of the Rate of Washout

The calculation of the removal rate by washout is attempted by determination of the average fraction of volume of the troposphere from which the aerosol is removed daily. Consider a vertical column of 1 m$^2$ cross section of height, $H$, containing precipitating clouds of liquid water content, $L$, from which $R$ inches of rain falls in one day. The equivalent thickness, $h$, of cloud from which this rain falls and from which the aerosol can coincidentally be removed is

$$h = \frac{.0254\,R}{L}$$

which corresponds to a volume fraction of the tropopause of

$$\frac{h}{H} = \frac{.0254\,R}{LH} = \alpha\,.$$

For aerosols other than sea salt, the efficiency for condensation may be less than unity, necessitating a correction factor, $\varepsilon \leq 1$, representing the fraction of particles removed:

$$\alpha = \frac{.0254\,R\varepsilon}{LH}\,.$$

Since this is a linear relation, the average fraction removed over a wide area is obtained by using an average value of $\overline{R}$,

$$\bar{\alpha} = \frac{.0254\,\overline{R}\varepsilon}{HL}\,.$$

This, however, is correct only if the rain falls simultaneously over the whole area considered. Especially in summer, most rain falls from showers or cloud systems small compared to the eastern half of the U.S. and is fairly independent in time. This means that considerable mixing will occur between rainfalls. If the average rainfall, $\overline{R}$, results from $n$ independent showers, the average fraction of aerosol remaining in the atmosphere is $(1 - \bar{\alpha}/n)^n$. With increasing $n$, the expression rapidly approaches $e^{-\bar{\alpha}}$. The average fraction of the aerosol removed, $a$, is therefore approximated by

$$a = 1 - e^{-\frac{0.0254\,\overline{R}\varepsilon}{L \cdot H}}\,.$$

For sea salt particles $\varepsilon = 1$ and with the assumption of a constant value of $H = 10^4$ m, $a$ is dependent on $\overline{R}$ and $L$ only.

The fraction of particles removed from below the cloud by collection by falling raindrops can be estimated in the following way. Consider the average base of the clouds to be at 2 km. In the worst possible case all of the aerosols from the ground to the cloud base will be removed, which amounts to 2/10 of the total tropospheric column content. Since in summer in Eastern U.S. it rains an average of eight days during the month, the daily average fraction of aerosol washed out by this process, $b$, is

$$b \leqslant 0.2 \times 8/31 = 0.05.$$

This value is an upper limit, but since even small amounts of rain remove a considerable fraction of sea salt particles and since $b$ is small compared to a, the error introduced will be unimportant.

The daily total average fraction of the aerosol removed, $c$, is

$$c = a + b.$$

If the same air mass is subjected to this washout for $m$ consecutive days, the fraction of the aerosol remaining airborne on the $m$th day, $d$, is

$$d = (1 - c)^m.$$

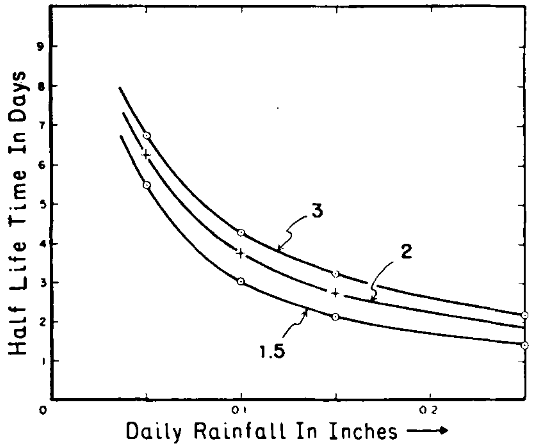

Fig. 5. Calculated half lifetime of sea spray particles as a function of the daily average rainfall at three values of the liquid water content of precipitating clouds, assuming that the height of tropopause is 10 km, the cloud base is at 2 km, and mixing occurs between showers.

From this relation, the time required for an aerosol *uniformly distributed throughout the troposphere* to be depleted to one-half its original concentration, the half lifetime, is easily computed. The half life as a function of $R$ and $L$ is plotted in Fig. 5.

An alternative computational method, using four day trajectories, the actual rainfalls of August 1955, and a more elaborate estimation of the collection by falling drops gave very similar results, justifying the simpler technique given here.

The essential assumption which enters this computation is that of continuous mixing in the troposphere between rainfalls. Other than this assumption, the results depend only on values of $\bar{R}$ and $L$, which are fairly well known. For the summertime U.S. air masses, a value of $L$ of 1.5—2.0 g/m$^3$ is given by several investigators (AUF'M KAMPE and WEICKMANN, 1953, and HOUGHTON, 1950). $\bar{R}$ fluctuates around the value of 0.1 inch per day. The corresponding half life is 3—4 days. If southerly winds in the lower levels of 10—15 km/hr are assumed, the decrease of sea salt particles with distance due to washout is indicated by curve 5, Fig. 3. This rate is not sufficient to account for the observed change in concentration with distance near the coast. The values of $\bar{R}$ and $L$ vary somewhat with season, but the variation is not large, so that the half lifetime for most of the Eastern U.S. will remain about the same throughout the year. Because of increased average wind speeds, the half life *distance* will be somewhat larger in winter.

Another check of the half lifetime of sea salt particles is given by a budget consideration over the Southeastern U.S. during summertime. A wind speed of Beaufort 3 for the Gulf of Mexico area yields $3.5 \times 10^{-6}$ g/m$^3$ of sea salt, according to WOODCOCK (1953). If the inversion level is estimated to be at 1 km, a total salt mass of $3.5 \times 10^{-3}$ g is contained in a unit area column. According to Fig. 4, this value should also be representative for air over the continent. The average Cl$^-$ content of rain water inland, Fig. 1, is $0.15 \times 10^{-3}$ g/l and the average daily rainfall is 0.1 in. or 2.5 l/m$^2$. The average amount of NaCl brought down daily is $2.5 \times 0.15 \times 10^{-3} \times 1.65 = 0.62 \times 10^{-3}$ g/m$^2$. At this rate the original salt content in a unit area column, $3.5 \times 10^{-3}$ g/m$^2$, will be decreased to one-half in 3.5 days. This value is in close agreement with the washout calculation.

It is interesting to compare these results with measurements of radioactive fallout which have recently been made by STEWART, CROOKS and FISHER (1955). These investigators, using aircraft over the East Atlantic, followed for several months the radioactive content in the troposphere resulting from the Nevada test series in the U.S. In these tests, virtually no material penetrated the tropopause, and no appreciable mixing with air masses south of the horse latitudes was observed. The radioactive material, confined in the west wind belt, circled the earth several times and the observed decrease of particulate matter must have been due to natural removal processes. Measurements on the surface indicated that dry fallout amounted only to 25 per cent of the washout, so that their figures of the removal rate are essentially those for washout. Their results indicate a half lifetime for aerosols of about twenty-two days. At the average rainfall throughout the west wind belt of 0.05 inch per day, Fig. 5 gives a value of six days. A comparison of these figures must, however, take $\varepsilon$ into account. For particles smaller than 1 $\mu$, especially if they are not hygroscopic, $\varepsilon$ is expected to be much smaller than 1. Unfortunately, Stewart et al.

do not give data on the particle sizes. However, there is strong evidence from measurements on natural radioactive particles (Wilkening, 1952) that the major part of the radioactivity is associated with particles less than 0.1 $\mu$ diameter. The discrepancy between our calculation and the results of Stewart et al. is in the expected direction, but more complete statements have to be postponed until reliable data on $\varepsilon$ become available.

In a completely different way Blifford, Lockhart and Rosenstock (1952) and also Haxel and Schumann (1955) determined values for the half life of natural aerosols. In both cases, the investigators measured the composition of the natural aerosols with respect to the short lived and long lived decay products of natural radon. If no aerosols are removed, the concentration of these products should agree with the equilibrium of decay. From the observed low level of long lived products, they obtained half lifetimes of the aerosol of twelve days (Blifford et al.) and four days (Haxel et al.). However, two facts must be considered before comparing these values with the previous ones. First, the long and short lived products may have originated at different parts of the earth and may be related to different initial concentration levels of radon. Because of this difficulty, the values can only be regarded as approximate ones. Secondly, it is possible that this half lifetime refers only to the case where the major portion of the aerosols is confined to the lowest levels of the troposphere (as is normal for most natural aerosols) and not to uniformly distributed aerosols, as in our case of sea spray over continents and in the data of Stewart et al. It is expected that this "short cycle" of removal results also in shorter half lifetimes. Despite these facts, the agreement is not bad and we can assume that the average half lifetime of large hygroscopic aerosols is about three to four days with U.S. average rainfall, increasing to about twenty days for small hygrophobic particles and smaller amounts of rain.

### The Change of Vertical Distribution of Sea Salt Particles by Convective Mixing over Land

It has been shown that the washout process alone is not sufficient to account for the observed decrease in the Cl$^-$ content of rain water with increasing distance from the coast. We now have to show that vertical mixing works fast enough to convert the vertical distribution of sea salt over the ocean, indicated by Woodcock's curves in Fig. 4, into a uniform tropospheric distribution.

Let us consider the summer, when maritime air masses from the Gulf of Mexico or the Atlantic move inland with south to southeasterly winds. The convective mixing over land may be assumed constant with height and characterized by the eddy diffusion coefficient, $D$. The distribution of salt particles $n(z, t)$ with height, $z$, and time, $t$, is to be calculated. In order to avoid extensive numerical calculations, the actual distributions are approximated by an analytical function. This must satisfy three conditions:

1. $\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial t^2}$ for $t > 0$ and $z > 0$.

This is the well-known diffusion equation.

2. $\frac{\partial n}{\partial z} = 0$ for $z = 0$ and $t > 0$

since there is no Cl source over land.

3. $\int\limits_{z=0}^{\infty} n dz = \text{const}$

which follows from 2 and indicates that the total amount of sea spray particles stays constant. A function fulfilling these requirements was used by Junge (1952) in a similar context:

$$n\,(z, t) = \sqrt{\frac{\pi}{t_0}}\,[P\,(z, t + t_0) - P\,(z, t)]$$

where

$$P\,(z, t) = \frac{1}{\sqrt{\pi}} e^{-\frac{z^2}{4\,Dt}} - \frac{z}{2}\sqrt{\frac{1}{Dt}}\left[1 + \Phi\left(\frac{z}{2}\sqrt{\frac{1}{Dt}}\right)\right]$$

and $P\,(z, t + t_0)$ is the same when $t$ is replaced by $t + t_0$. $\Phi$ is the error integral.

Actually, this function applies to the case where the atmosphere becomes contaminated from a uniformly distributed source of constant aerosol production rate to time $t_0$, at which time the source is cut off and the particle number remains constant during the subsequent change of the vertical distribution

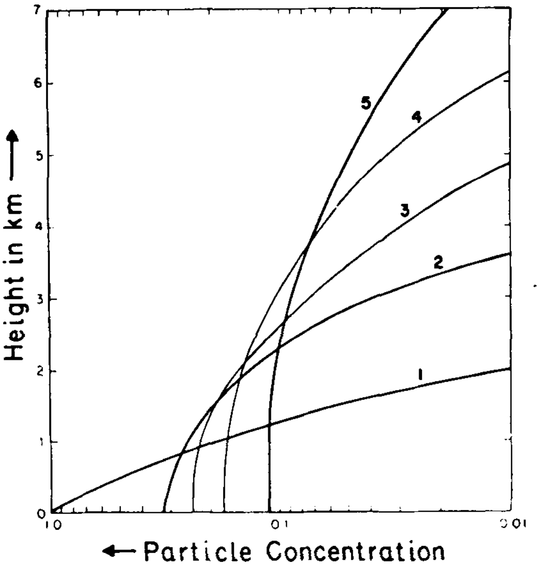

Fig. 6. The change of the vertical distribution of particles by mixing. Curve 1 gives the distribution over the Gulf at time zero. Curves 2 to 5 are calculated distributions at later times under the assumption of a constant eddy diffusion coefficient with height $D$. The time in hours which corresponds to these curves depends on the assumed value of $D$:

| | curve = | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|---|
| $D = 8 \times 10^5$ cm$^2$/sec | $t =$ | 0 | 3 | 6 | 12 | 24 hours |
| $D = 4 \times 10^5$ » | $t =$ | 0 | 6 | 12 | 24 | 48 » |
| $D = 2 \times 10^5$ » | $t =$ | 0 | 12 | 24 | 48 | 96 » |

by mixing. For the present purpose, the distribution at $z = 0$, $t = 0$ has been normalized to $n(z, t) = 1$ so that the production rate does not enter the relation. The results of these calculations are plotted in Fig. 6. Curve 1 represents the analytical approximation of Woodcock's distributions given in Fig. 4. As time increases, the concentration of particles in the ground layer decreases rapidly. The distributions at later times, as given by the other curves, pivot about a point at about 2 km altitude with rapidly increasing concentrations above 3 km.

The actual values of $D$ are not well known. From LETTAU (1951) we infer that for ordinary turbulence $D \approx 2 \times 10^5$, increasing in cumulus and cumulonimbus to $10^6$ and $10^7$. During the night, $D$ decreases to about $5 \times 10^4$ when winds are weak. A conservative average may be $D = 4 \times 10^5$ on which the calculations of Fig. 6 were based. With this value of $D$, fairly uniform distribution throughout the troposphere is approached after two days.

As mentioned above, the collection of sea salt particles by falling raindrops is decisive for the $Cl^-$ content of the rain water when the major part of the sea salt is concentrated in the lowest layers. If by the mixing over land these layers rapidly disappear, the $Cl^-$ content reflects primarily this decrease of the total sea salt content below the cloud base. According to Fig. 4 and Fig. 6, this value decreases by a factor of about 5 to 10, which is in good agreement with the observed decrease in $Cl^-$ concentration inland given in Fig. 3. After uniform mixing is approached, the salinity of rain water farther inland stays fairly constant because the depletion by washout proceeds slowly. There is indication on some of the monthly maps that the lowest $Cl^-$ concentrations are found in the lee of the Rocky Mountains with westerly winds or north of the Appalachians at times with southerly winds. This is probably due to the much more efficient washout due to orographic lifting of the air mass. The higher efficiency of washout in this case is, of course, caused by the fact that all rain in a certain air mass falls approximately simultaneously, so that no intermediate mixing can occur.

We infer from Fig. 6 that with an onshore wind of 10—15 mph in Southern U.S. the decrease in $Cl^-$ content of rain water with distance inland should cease at about 400—600 km, which agrees fairly well with the observations.

During the winter and at more northerly latitudes the vertical mixing process is not as intense, so that it is expected that the $Cl^-$ concentration in rain will not drop as quickly. This effect, however, does not seem important, as indicated by the Swedish data in Fig. 3. The average winter map of the U.S. (Fig. 2) shows slightly higher $Cl^-$ levels which may be due in part to this effect, and in part to a higher sea spray content in marine air during the stormy season.

## Conclusions

It is shown that the distribution of $Cl^-$ concentration in rain water over the U.S. gives a fairly true picture of the distribution of the total tropospheric sea salt content. This distribution is primarily determined by the change in the vertical distribution of the sea spray particles when the maritime air masses move inland. The large scale washout of these particles is rather inefficient and cannot influence very much the features of the $Cl^-$ distribution maps. This was demonstrated in four different ways:

1. Comparison of data on the total particle counts over sea and land;
2. Calculation of the washout efficiency in an air mass, and comparison with known data from radioactive work;
3. Calculation of the depletion rate of sea salt by rain; and
4. Calculation of the vertical mixing process over land.

The four methods agree and give a consistent picture. The important result of this analysis is that even large-sized hygroscopic aerosols may travel considerable distances in the atmosphere without being removed by precipitation. In the west wind belt, times of 6 to 25 days are required to reduce the concentrations of aerosols by a factor of 2, the longer times probably referring to smaller and less hygroscopic particles. Values of the half lifetime of atmospheric aerosols as a function of their size, perhaps their hygroscopicity, and the average amount of rainfall are of increasing importance with respect to worldwide air pollution problems.

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