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November 13, 1956

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Dr. J. C. Potts
U. S. Atomic Energy Commission
Washington 25, D. C.

31961

Dear Dr. Potts:

I am to deliver a talk at the Washington Academy of Sciences, as indicated in the enclosed paper on Thursday, November 15. Since Dr. Libby's name is mentioned liberally, and since this paper deals at length with the Strontium-90 problem, I feel that you and Dr. Libby should have the opportunity of commenting on it. I shall leave it to your best judgement to decide whether it is worth taking Dr. Libby's time to review it.

I have submitted the paper to Mr. Nash of the Division of Classification for security review.

In the event that either you or Dr. Libby have comments which you feel should be included, I would appreciate hearing about them by Thursday morning, to be able to incorporate them in the talk.

Sincerely yours,

Lester Machta

Lester Machta, Chief
Special Projects Section
Office of Meteorological Research

Enclosure

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Meteorological Factors Affecting Spread of Radioactivity from Nuclear Bombs

By

Lester Machta
U. S. Weather Bureau
Washington 25, D.C.
November 15, 1956

A talk delivered at the Washington Academy of Sciences

Introduction

The fissioning of a nuclear weapon is the first step in a chain of events which may ultimately result in the ill-effects of radioactivity on man. The dispersal from test site to human environment or food takes place largely through atmospheric transport. The problem, however, is more than the application of the usual winds to carry the particulate debris. It will be apparent that eddy diffusion and precipitation, for example, also play an important role.

Most persons who have treated the entire radioactive fallout spectrum have found it convenient to divide the history of the particulate fallout into three categories. First, that which deposits out in the first day, or less. This is designated as local fallout since in this short time the horizontal displacement cannot have been very great. The second class is deposited between about 1 day and a few months. For lack of a better word, it will be termed intermediate fallout. Finally, the fallout delivered beyond a few months is called delayed fallout. Because the particles are injected in the stratosphere and found over much of the globe, it is also referred to as "world-wide stratospheric fallout".

It is the purpose of this discussion to point out the meteorological considerations which account for all ~~of the~~ atmospheric transport of the debris. Remarks on local and intermediate ranges will be brief, while those pertaining to delayed fallout, more extensive.

Local Fallout

Local fallout is characterized by the fact that the deposition is largely by the settling of the particles due to their weight. Given the size, shape, and specific gravity of the particles, it is possible to predict their fall velocity.

However, even in the absence of such exact information, it is possible to produce useful results as to where and when particles will reach the ground. This is because the relative time of fall as a function of altitude ^{the importance of distance} is insensitive to the particle's physical properties, and because ~~it is the relative time of fall with height which turns out to be important.~~

There are at least two other processes which produced a downward flux of particles in the atmosphere. First, vertical mixing acting upon a cloud of particles at upper altitudes will transfer some of these to lower altitudes. Although we cannot be sure, the magnitude of usual tropospheric vertical eddy speeds is of the order of a few tenths to a few miles per hour. It is likely, therefore, that particles which have settling speeds of a tenth to a hundredth of the eddy velocity will have their vertical component of motion controlled mainly by atmospheric turbulence while those with fall velocities equal to or greater than a few miles per hour will descend mainly because of gravitational settling. The point of division for a spherical particle of specific gravity of 2.5 is between 50 and 80 microns in diameter. Most of the sizes of particles ^{the heavy} in local fallout exceed this diameter.

A second means of downward transfer of the particles occurs by precipitation. As far as possible, nuclear tests by the United States are conducted in the absence of naturally-occurring precipitation. However, in the Pacific Proving Grounds, it is likely that the huge amount of moisture entrained by the rising fireball condenses upon cooling and that some downward transfer of the particles results from scavenging of falling precipitation.

The horizontal motion of the falling particles results primarily from the action ^{of} winds which are observed by conventional wind measuring equipment. The procedures for utilizing winds for the determination of fallout areas are entirely straight-forward. The falling particle is transported horizontally by the wind in a layer depending upon its stay in the layer before settling to the next lower level. The summation of the horizontal displacements ends with the ground deposition of the particle.

In addition to transport by the usually-observed winds, there is a certain amount of lateral dispersion because of horizontal turbulence in the atmosphere. Experience suggests that in most cases, the spread of falling particles by the winds so greatly exceed the spread due to small-scale turbulence that the latter effect is usually ignored.

In the prediction of radiation gamma intensities/ ^{from the ground} it is necessary to know not only the winds and physical characteristics of the falling particles, but also the amount of radioactivity as a function of particle size ^{and} altitude. This latter information is essentially ~~a~~ non-meteorological problem and will not be dealt with at this time. An example of the final result of the application of meteorological winds to a model of radioactivity as a function of altitude and particle size is shown in the first few figures.

The first figure shows the actual and predicted isolines of gamma activity in milliroentgens per hour referred to twelve hours. The light dashes, the observed field, were derived from ground monitoring along various roads around the Nevada Test Site. The heavy lines show the predicted fallout pattern using a Weather Bureau model of cloud radioactivity. On this picture, the winds measured near the explosion place and time, which were mainly from south to north, were assumed to carry particles at all later times and places. It is evident that the prediction, the type made most

frequently, fails to reflect the bending of the pattern to the east in northern Nevada. The next figure contains a fallout pattern, as the heavy lines, which include the best estimate of the change of the wind field in time and in space. It is evident that incorporation of the changing winds provides a better verification. This after-the-fact fallout pattern is typical of most cases which have been attempted.

Intermediate Fallout

After most of the particles whose size are large enough to have a significant fall velocity have been removed from the atmosphere, the remaining particles are part of either the intermediate or delayed fallout. For weapons which have been fired at the Nevada Test Site, the intermediate fallout originates in the troposphere since the nuclear cloud tops rarely penetrated the tropopause. However, for larger calibre detonations, the intermediate fallout may originate either from tropospheric or stratospheric material. ~~If the stratosphere is the source for some of the intermediate fallout, then the particle sizes should be greater than about 5 microns for a specific gravity of 2.5. This is necessary since the vertical turbulent transfer in the stratosphere is very small.~~ For the intermediate fallout originating from the troposphere it is likely that the sizes will be less than about 20 microns ~~else the larger particles would be part of the local fallout.~~ ^{and} At the moment, it is not possible to say with confidence which source contributes the larger fraction for high-yield explosions.

It is characteristic of the motions in the atmosphere that air masses are carried zonally, that is, around circles of latitudes, much more rapidly than in the north-south direction. This is true both in the troposphere and stratosphere. The result of this meteorological fact is the predominance of intermediate fallout

in the same latitude band as the test locations. In Figure 3, the 35-day cumulative fallout from CASTLE BRAVO is shown as a series of isolines on the lower part of the figure. The upper portion of the figure shows as the shaded area the regions of the globe which, from meteorological considerations, were expected to have fallout in the first week or so. It is evident that the band of highest fallout circles the earth in the tropics of the northern hemisphere with only occasional north-south excursions.

The removal of particles in the intermediate range from the atmosphere during dry weather is assisted by vertical eddies near the earth's surface and by interception by obstacles. However, it is likely that the ^{an} most important fraction is removed by precipitation scavenging. It has been found, for example, following most of our Nevada test operations, that on days with rain, there is 10 times more radioactivity deposited on the ground than on days with no rain. The particles are probably large enough so that scavenging by inertial effects are still important. That is, the small radioactive particle, because of their inertia cannot escape from the path of the falling raindrop.

The budgeting of the fraction of radioactivity deposited in the intermediate range as well as that in the delayed fallout is quite uncertain. Careful analysis of the world-wide fallout network of the U. S. Atomic Energy Commission after the IVI and CASTLE test series suggests that between 1 and 5% of the fission products which were formed fell out between about 1 and 30 days after each test operation. The same numbers for the intermediate fallout from the Nevada tests amounts to perhaps 25%. In the latter case, the amount of the close-in fallout is known quite exactly and since there is no delayed fallout, we can add the local and intermediate fallout together to obtain the total. The result shows that something of the order of 50% of the radioactivity is still unaccounted for.

It is possible that the local fallout has been underestimated but this is unlikely. The more reasonable explanation is given in terms of our inability to accurately measure the true removal of particles from the atmosphere with simple devices. The estimate just quoted is based on the measurement of fallout on gummed films exposed horizontally. Even if this were 100% efficient (and it is believed to be about 70% efficient for gross fission products now under discussion), it fails to detect radioactive particles removed by vertical surfaces. The Naval Research Laboratory has reported an amount of radioactivity on a vertically-exposed piece of cheesecloth equal to that deposited on the ground for the same time interval and area. Other experiments confirm the fact that the particles in the intermediate range are collected by other than horizontally-exposed face-up surfaces. The question which is still unanswered, however, is ^{how much more is removed} ~~the actual magnitude of~~ this removal in comparison ^{than} with that observed by the gummed film ^{network}.

Thus, the cumulative contribution of the intermediate fallout and the delayed fallout over that portion of the world lying in the belt surrounding the nuclear proving grounds is still somewhat questionable. When soil samples are collected, say over the U. S., which have integrated all fallout since the atomic age, questions may be raised concerning the apportionment of the fallout.

Although, on the average, the bulk of the radioactivity in the intermediate range lies in a belt surrounding the latitude of the test site, there can be anomalous situations which will either spread the debris over a very large north-south range or carry the entire nuclear cloud in toto initially to a new and distant latitude for zonal transport there.

Delayed Fallout

The delayed fallout represents that fraction of the radioactivity which may still remain to be deposited out on the earth's surface. In a sense, therefore, it is an unknown quantity although a very reasonable upper limit can be placed on ^{its} magnitude from past explosions. Furthermore, evidence points to the fact that this fallout is affecting the nations of all the world rather than the country within which the tests took place or those immediately downwind of it.

Since the particulate debris which is located in the troposphere can be shown to be removed within a matter of weeks or months, ^{even after the delayed fallout} it follows that ~~in the absence of very frequent tests,~~ delayed fallout must be stored somewhere. Aircraft sampling and balloon flights have clearly established that the stratosphere is the storage region for delayed fallout. It can also be deduced that, since the particles take longer than a few months to fall out of the atmosphere, their size must be exceedingly small and it would be estimated that they are no larger than 1 micron and probably well below 0.1 microns in diameter. However, despite this apparent smallness in size, we have no positive proof at this point that gravitational settling is not important in the transfer of radioactive particles from the stratosphere into the troposphere, and, in fact, there is the slightest suggestion that gravitational settling may even be important.

It is perhaps worthwhile at this point to review the questions which can be asked concerning the meteorological aspects of delayed fallout. First, how long will the particulate debris remain in the stratosphere, and second, where will it come out of the stratosphere to be deposited on the earth's surface.

In order to make a prediction of contamination, it is essential to know the source of the pollutants. The high-altitude measurements

at this stage are inadequate to provide the answer to the question of where the debris is located in the stratosphere. The next figure shows some of the reasoning in trying to determine even the initial distribution. To the left, we see a drawing of a nuclear cloud with the familiar stem and mushroom. In this hypothetical high-yield explosion, the entire mushroom has been placed in the stratosphere above the tropopause. To the right, we see, as the dashed line, the profile of the visible cloud ~~to~~^{of} the mushroom. If one assumes that extreme turbulence exists in this cloud so that it is thoroughly mixed, then at every point the amount of radioactivity per gram of air ~~may be assumed~~^{is} to be constant. Since the mass of air per unit volume or density decreases with height, there would be more radioactivity near the bottom of the cloud. This is illustrated by the solid line. On the other hand, it can be argued that the small particles remain with the fireball and that the fireball becomes the torroidal ring present with most, if not all, nuclear explosions. If this is the case, then it is possible that the radioactivity is distributed according to profile B.

The geographical locale for the debris is less uncertain. In Figure 5, a vertical cross section from pole to pole is shown with the approximate latitudes of high-yield explosions indicated at the upper border. As previously noted, only those small particles initially injected into the stratosphere are of concern. It is seen that the height of the bottom of the stratosphere, the tropopause, varies with latitude. It also varies in altitude day by day and season ^{ally} at the same place.

The stratosphere differs from the troposphere in two important aspects. First and foremost, whereas the temperature decreases on the average of 6.5° Centigrade per kilometer in the troposphere there is either no temperature change with height or even, as in the

tropics, an increase of temperature with height in the stratosphere. Second, the stratosphere is practically cloudless and has no precipitation falling from it. The first difference means that there is very little vertical turbulence in the stratosphere compared with the troposphere, so that vertical turbulent mixing is much slower. The absence of clouds means that the removal of debris cannot be aided by falling precipitation elements.

The absence of mixing in the vertical coordinate does not preclude horizontal diffusion, and in fact, Parr's principle, would suggest that horizontal mixing might be greater on account of the lack of vertical mixing. However, except for the spread of dust from Krakatao, ^{The volcanic eruption of 1883} there is little or no evidence on either side. Unfortunately, the optical detection of the Krakatao dust is not quantitative in the sense that it is possible to assign concentrations to the spread of the dust. There is reason to believe that the equatorial band of the lower stratosphere may prevent exchange of air between the hemispheres just as is the case for the lower troposphere. In the troposphere, it is the convergence of air into the Intertropical Convergence Zone, as shown in Figure 5, which prevents exchange. In the lower stratosphere, it is the steadiness of the east to west wind which would suggest little or no north-south exchange. This point is of some consequence. It may mean that the hemisphere with the atomic tests will obtain a disproportionate share of delayed fallout. On the other hand, the measurement of delayed fallout in the southern hemisphere means that some stratospheric debris must have come into the southern hemisphere. Conceivably, the path of the debris may be through the northern hemisphere tropopause and then to the southern hemisphere via the upper troposphere.

Since the vertical diffusion is very slow in the stratosphere and the north-south mixing possibly slower than the troposphere, the meridional circulations may become important instruments for the transfer of the debris. In the next figure, are four published models of meridional circulations in the lower stratosphere. The models of Wulf, Goldie and the lower part of Palmer display a cell which calls for sinking motion in the equatorial area, rising motion over the poles and appropriate north-south motions to complete the cells. The Palmer model suggests that above 83,600 feet in the tropics, there is a reversal to rising motion. Kellogg and Schilling indicate that the main north-south drift will be from the summer toward the winter hemisphere. It should be remembered that the magnitude of the air motions associated with arrows on this figure are exceedingly small compared to the west-east air motions; centimeters per second or less compared to meters or tens of meters per second.

There is nothing unique about the problem of mixing across the tropopause. It is probable that the tropopause is the separation between fast and slow vertical mixing, but in itself represents no unusual barrier. In addition to air exchange through the tropopause, there are other possibilities, ^{for escape from the stratosphere} As was evident from many of the figures, the tropopause is believed to have a break in the temperate latitudes and the air exchange may occur here without any crossing of a tropopause. Secondly, in the course of everyday weather the tropopause disappears and reforms at different altitudes so that exchange of tropospheric and stratospheric can take place again without crossing the tropopause. Finally, in the polar areas the tropopause is often very indistinct and this combined with the expected sinking motion over cold sources suggests that ~~there is a~~ ^{the} possibility that the polar areas may be a region of transport from stratosphere to troposphere.

In theory, it is possible to predict the removal of the radioactive debris from the stratosphere given these non-weather data: the particle size, shape and specific gravities to determine the extent of gravitational settling and the distribution of radioactivity in space. In practice, not only are the non-weather data unavailable, but the meteorological ingredients described above are not sufficiently well known.

Dr. W. F. Libby has published information which permits one to bypass all of the ^{problems} ~~uncertainties~~. He does not specify anything but the total amount of radioactivity available for delayed fallout and the amount which has been deposited during these first few years of the thermonuclear age. Libby computes the fraction of the stratospheric burden deposited each year, finds it to be roughly 10% per year and assumes that this fraction can be extrapolated into the future. This assumption yields an exponential decrease in the amount removed each year.

Libby's approach is very appealing in that it is now possible to obtain an answer to the first question: how long will the debris remain in the stratosphere. At the moment, there is no alternative to Libby's analysis except to argue for changes in details.

Two ^{elements} ingredients enter into the calculation of 10% removal per year. These are: the quantity in the stratosphere at the beginning of a given time interval and the amount removed from the atmosphere during the interval. It is not possible to quarrel with Libby's estimate of the initial stratospheric burden for reasons of possible infringement on classified data.

The question of the removal of radioactive debris from the atmosphere, and in particular, Strontium-90, is important to resolve not only for the problem of stratospheric storage time but also for the understanding of the present level Strontium-90 on earth.

The most important ^{part} ~~ing~~ of delayed fallout

All groups studying Strontium-90 fallout agree on two points: first, that most of it comes down in precipitation and second, that the Strontium-90 is probably soluble in water. The Canadians and British state that about 85% comes down in rain. Libby has pointed out that Antofagasta in Chile, a location with essentially no precipitation has a lower Strontium-90 soil content by a factor of perhaps 50 than any other place on earth that has been sampled.

To understand how one computes the total Strontium-90 deposition per year, it is necessary to review the sampling devices in the light of the importance of precipitation. First, is the soil itself. Use of the soil as a method of integrating the world-wide delayed fallout has the following shortcomings:

1. The soil contains fallout from all times; it is a cumulative rather than a differential sampling device. However, successive sampling from the same locale may overcome this difficulty.
2. The ~~degree of~~ leeching of Strontium-90 is assumed to be small, since the upper several inches are ~~assumed to contain~~ all that has fallen out.
3. There are water losses during moderate and heavy rains or melting of snow packs due to runoff.
4. Removal by impaction on vertical surfaces is not measured.
5. Sampling over the oceans and other water bodies is impossible. This is serious only if spray, wave action or some other ocean phenomena causes a systematic bias of the results compared with land.
6. Radiochemical analysis is difficult so that only limited numbers of geographical areas have been sampled.

The second most reliable sampling technique is collection of precipitation samples by a pot or funnel. These devices collect dry fallout as well as rain drops. It is evident that the pot or funnel is a differential sampler, suffers no leeching or runoff losses, could be used over oceans, and the radiochemical analysis is simpler than for soil. On the other hand, it probably is very

inefficient for dry fallout and still provides no measure of interception on vertical surfaces. ~~The interpretation of isolated sampling points as extrapolated to the entire world must be approached with caution since the amount of Strontium-90 removed by rainfall may depend on many local characteristics of the rain-- as will be noted later.~~ A major shortcoming of the past has been the lack of observing points. Until recently, routine analysis of Strontium-90 ^{at the sample} has been limited to one or two places.

On the other hand, there has been ^{an} extensive gummied film network. But the gummied film suffers from a series of shortcomings.

1. With the small lip surrounding the film, ^{the} water running off the paper may be considerable. In fact, in a study by the Armour Research Foundation, the paper was found to be only about 10% efficient for a rainfall of 0.1" in one hour, for soluble substances.

2. During dry weather, the gummied film was found to develop an electrostatic charge at relative humidities below 45%. The result was a difference in collection efficiency by a factor of 6 between high and low humidities.

On the other hand, the gummied film does produce numbers which appear to be consistent with the pot method at ^a the few places, using an overall collection efficiency of about 40%.

It has been the purpose of this review of collection methods to enumerate the likely sources of error in determining the total amount of fallout of Strontium-90 per year. It is evident that large random errors are likely and that there are errors whose magnitude and sense cannot even be estimated. But of those uncertainties whose sense can be determined, it is the present view that ~~the estimates of annual fallout made initially by Libby~~ ^{initial estimate of 10 to 15} are too low. For example, Libby assumes no impaction on vertical surfaces, no runoff or leaching losses for his soil samples,

and a 79% rather than 40% efficiency for the gummed film. It is, therefore, concluded that more than 10% comes down per year. Dr. Libby's more recent talks reflect this possibility. It would be highly desirable to ascertain whether the calculated removal of 10% per year very seriously underestimates the true deposition since long stratospheric residence permits harmless decay and increases the likelihood of diffusion dilution.

Aside from the question of the actual deposition during the years following thermonuclear tests, there are also problems concerning the propriety of extrapolating the fractional removal of the stratospheric debris into the future. It is implied that the removal process is by diffusion and that the initial distribution is not such that the first year or two would yield anomalous results. With time, it will become evident whether the 10% is constant. Already, however, there is data suggesting irregularities in the stratospheric removal. During 1955, a large fraction of the Strontium-90 fallout occurred in ^{only} about 3 or 4 months in New York City rather than being spread evenly over the year. Lest one conclude that this is a seasonal affair, it may be noted that it did not take place in the same months of 1954 or in 1956. This was the only regional increase.

It is concluded that Libby's approach to the determination of the rate of removal from the stratosphere is ^{the best available} ~~better than any other one~~ but that it would be no surprise to find the percentage stratospheric removal to be far greater than 10% and to vary from year to year.

Rainfall

Before concluding it may be worthwhile to evaluate more carefully how rainfall removes the Strontium-90 from the atmosphere in view of the fact that it is attached to ^{such} ~~the~~ very small particles of delayed fallout. For particles whose diameters exceed a few microns, it has been demonstrated that inertia and interception by falling rain are reasonably efficient scrubbing agents but that for particles less than one micron, ~~this~~ type of collection efficiency

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is zero. Mr. Stanley Greenfield of the Rand Corporation has analyzed another mechanism of precipitation scavenging. This is an agglomeration process whereby the agitation of the small particle results in a probability of ~~ultimate~~ ^{of sub-micron particles} impact on the water element. The scavenging efficiency is shown in the final figure as the ordinate with the particle size as the abscissa. It is seen that to the right, with large particles, the efficiency is high as ^{is the case} for particles smaller than 0.1 microns, although for different reasons. In between, there is a "window" in which the removal by rain may be expected to be low. It is presumed that the delayed fallout is associated with particles below 0.1 microns, and therefore readily scavenged.

The delayed fallout, according to this hypothesis, should be removed by rain from the atmosphere largely at the level of the water cloud since the falling raindrop may not have enough time to allow agglomeration. However, actual comparison between Strontium-90 content of rain and in the air suggests that a very thick layer of air must have had its particles scavenged. This suggests that by a mechanism, still not understood, even the falling droplet is capable of scavenging the particles of delayed fallout.

To this, one may add yet another bit of evidence which fails to fit into a simple picture. Although Antofagasta with its dry weather apparently contains ^{little} ~~no~~ Strontium-90, the correlation between rainfall and delayed fallout is poor either when one considers the total amount of rain or the number of days with rain. Thus, Paris, a wet place, shows only half as much Strontium-90 in the soil as does Damascus, Syria, a dry region.

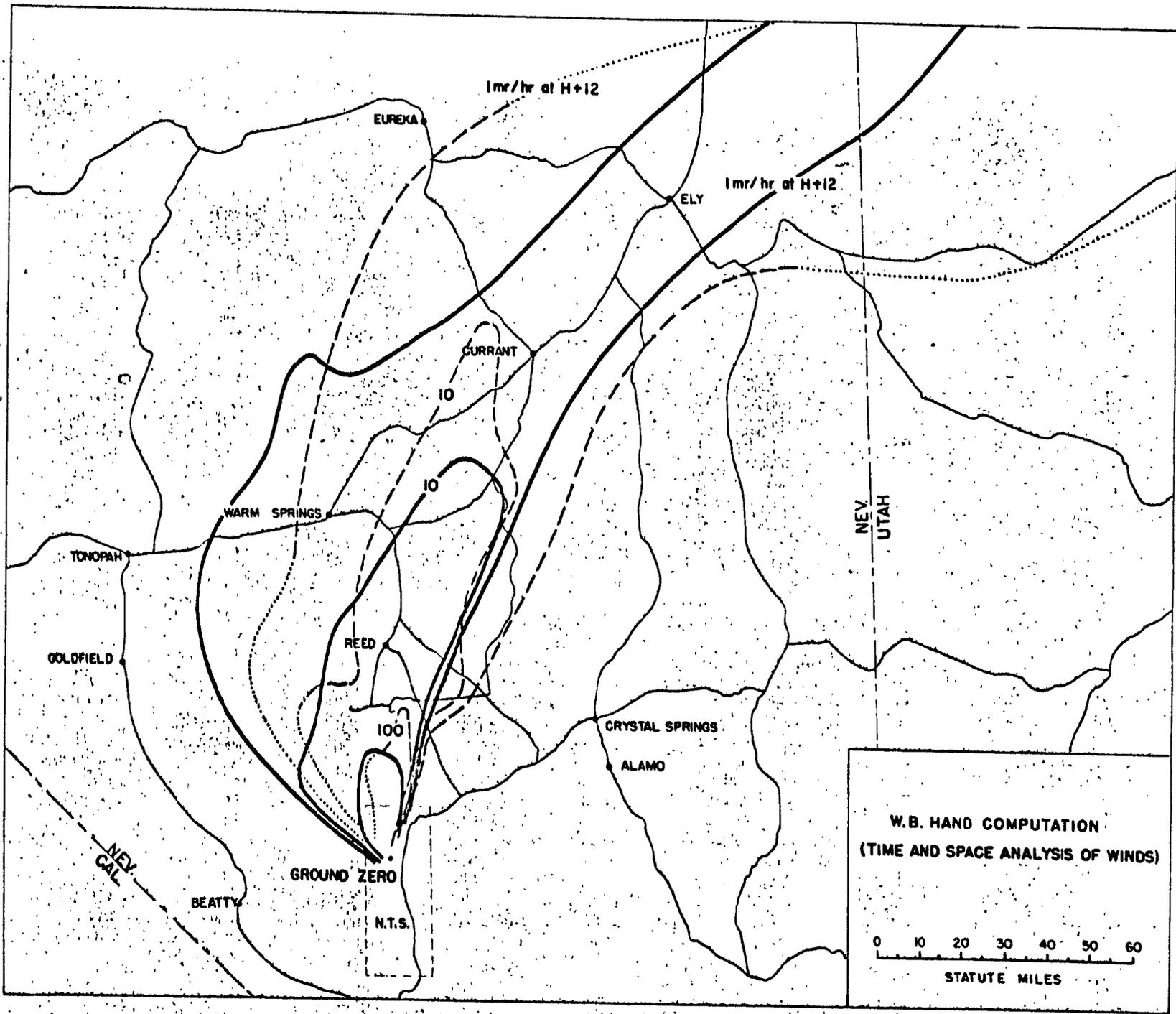
It is hoped that further research on rainfall in relation to Strontium-90 content will explain the numerous discrepancies that appear to be present.

Conclusion

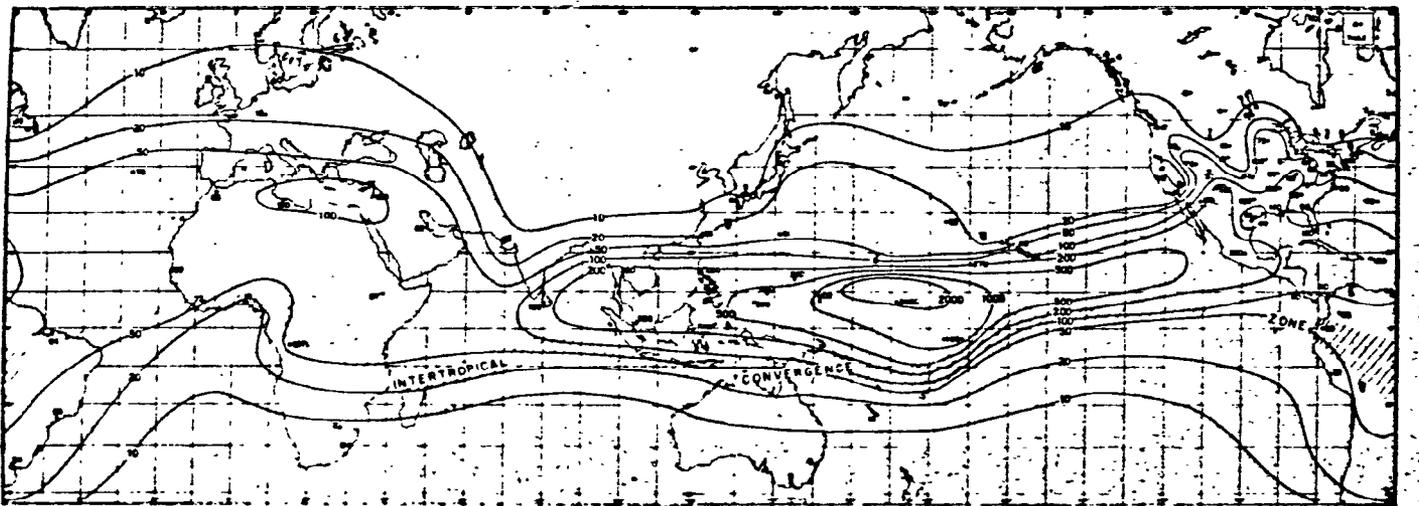
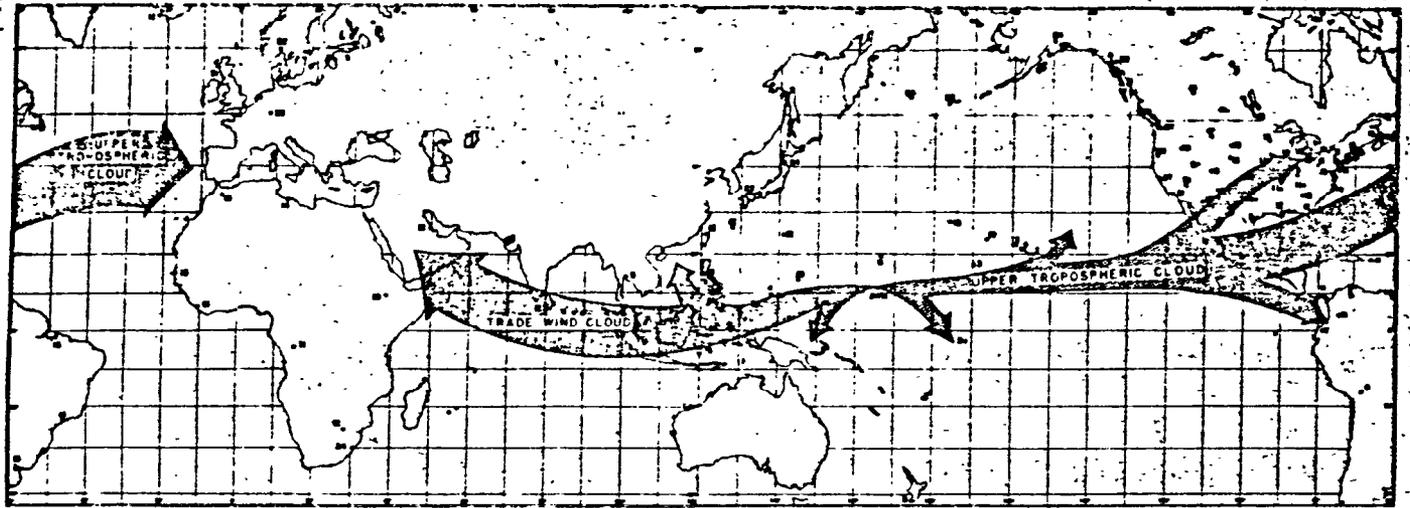
Although, in principle, it is possible to plan a procedure for predicting the removal rate of delayed fallout from the stratosphere, in practice it is necessary to rely on the limited observations available to make the most reasonable estimate. It is recognized, however, that because the actual details are being omitted, the chance that serious errors are present becomes greater.

It is felt that stratospheric sampling holds a key to a more rational interpretation of the storage time. The distribution in time and space ^{in the stratosphere} may allow the meteorologist, with even his meagre information, to predict the desired quantities more reliably than at present.

As a final word, it might be added that the meteorologist stands to share in the knowledge which will be collected in the solution of the Strontium-90 fallout problem.

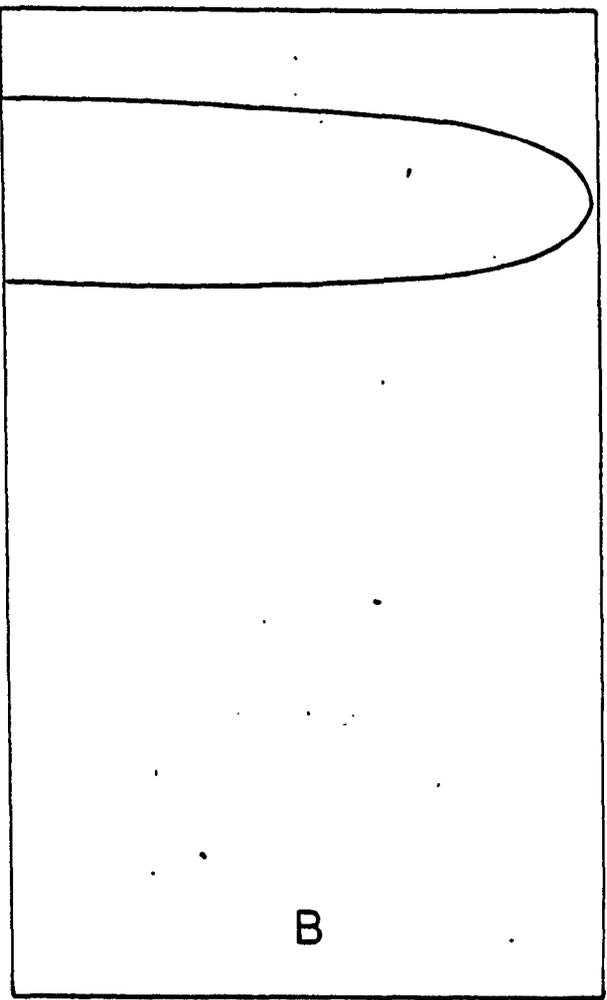
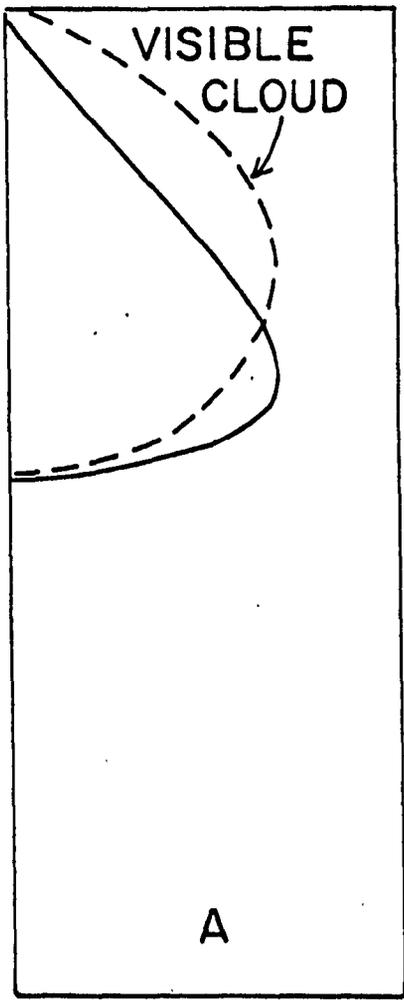
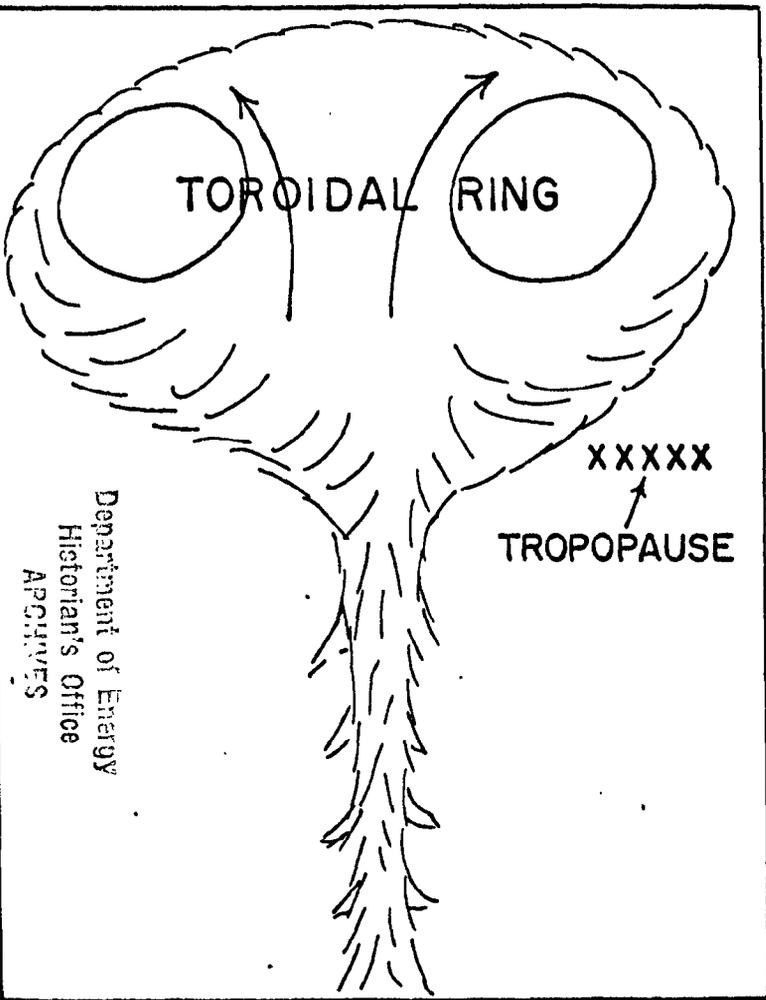


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Fig. 3



SCHEMATIC VERTICAL DISTRIBUTIONS OF SMALL PARTICLES IN NUCLEAR CLOUD

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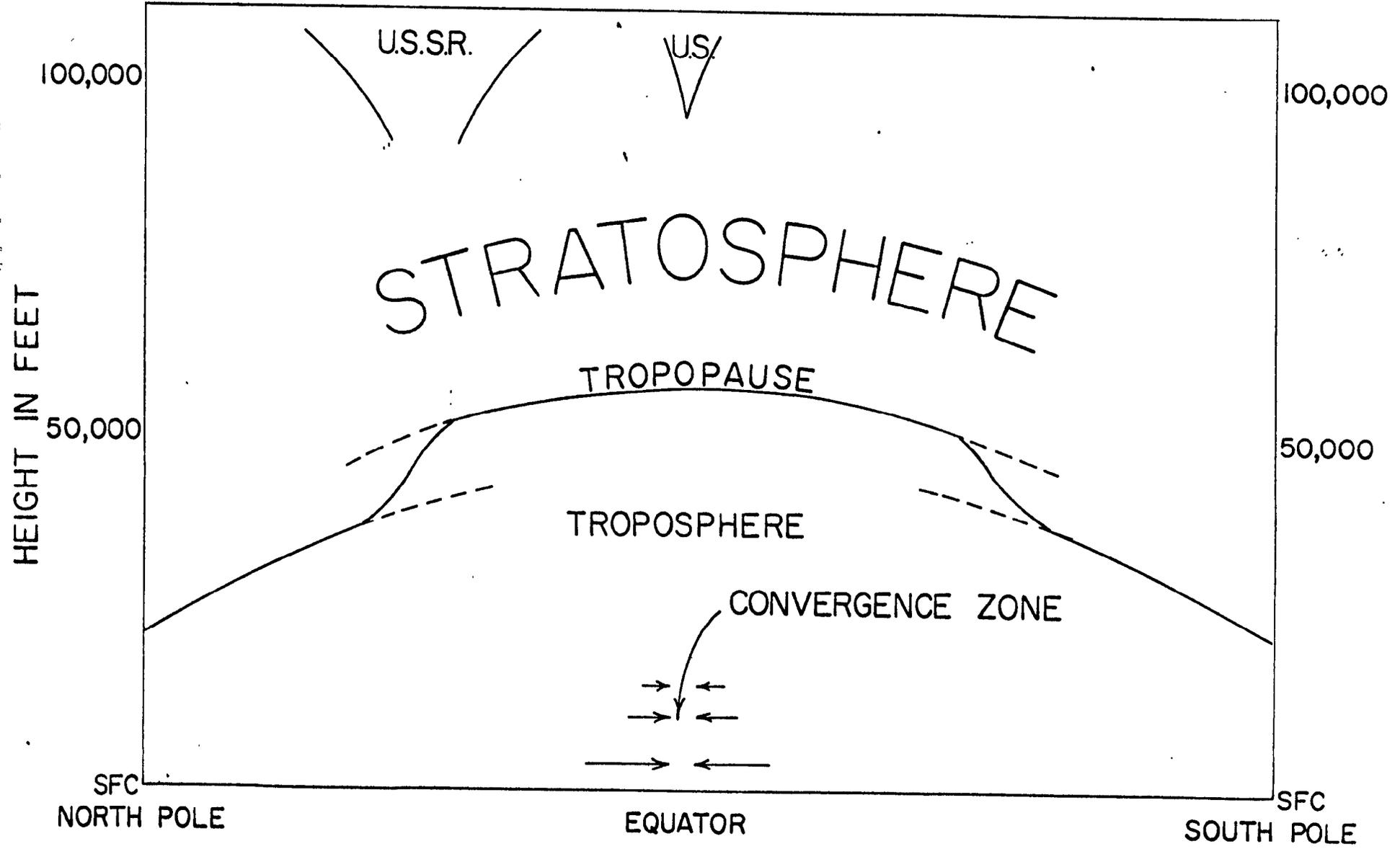
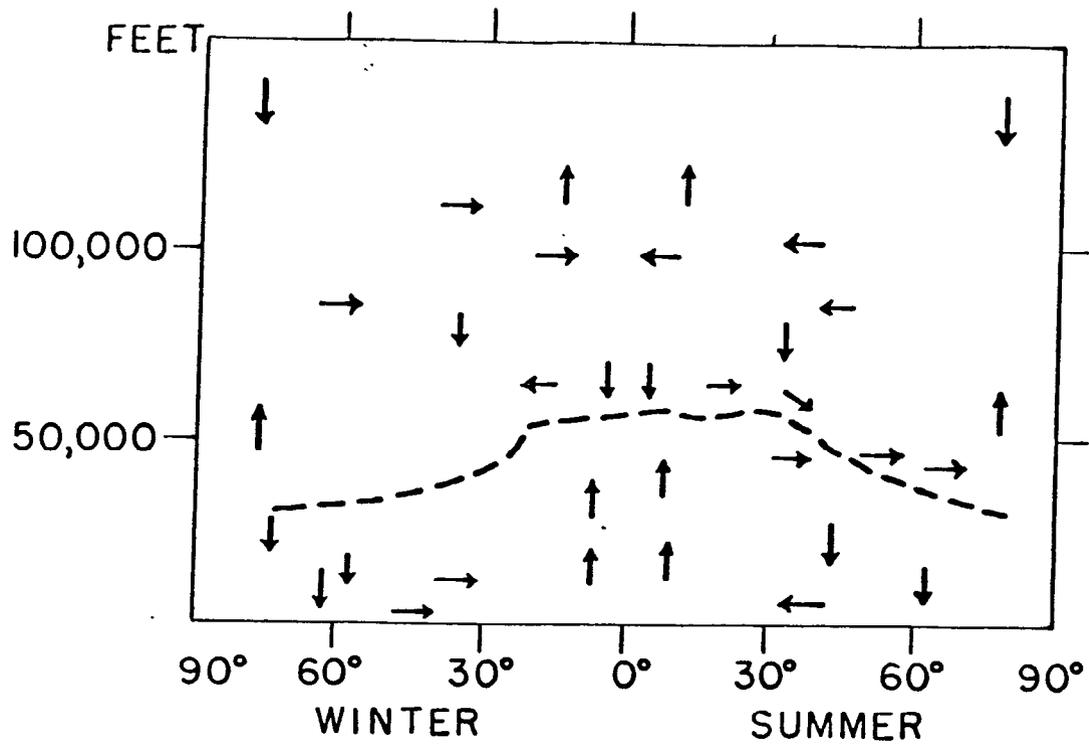
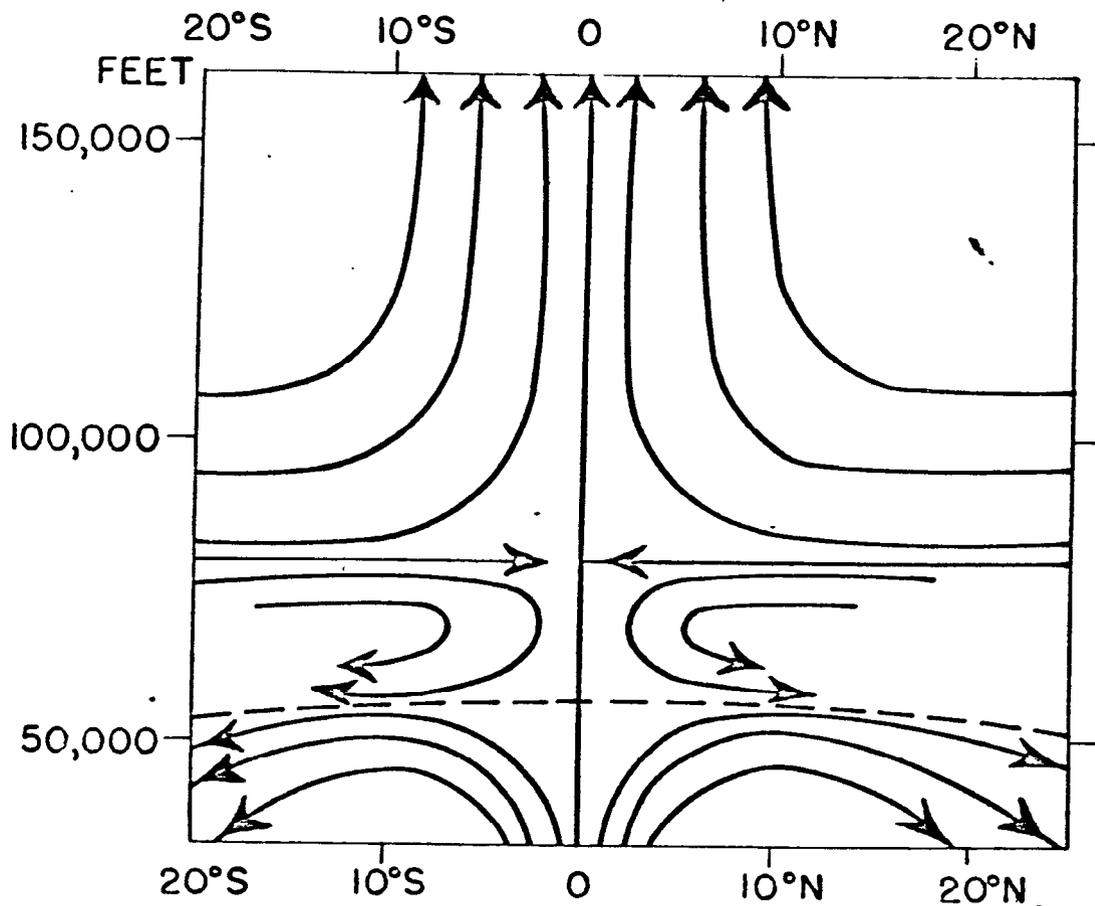


FIG 5

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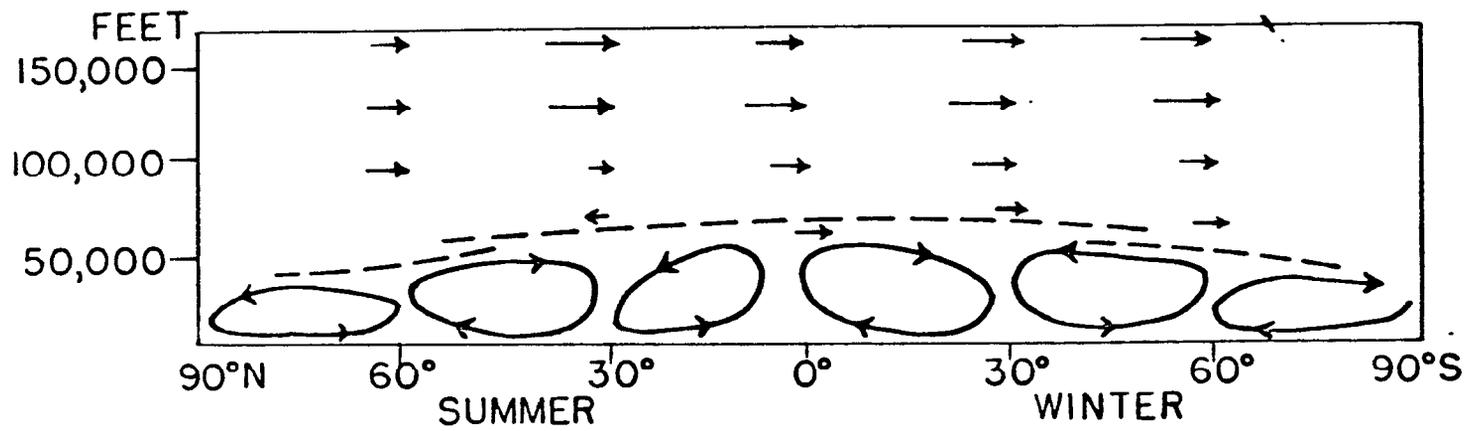
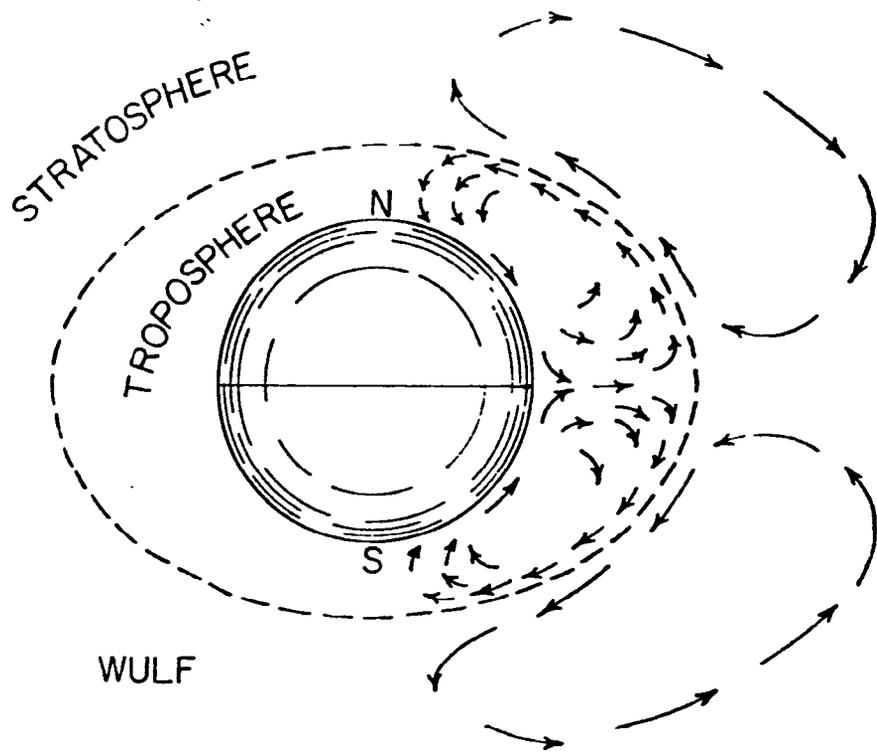


GOLDIE



PALMER

FIG 6a



KELLOGG AND SCHILLING

FIG. 6 b

Department of Commerce
 Bureau of Oceanography
 Washington, D. C.

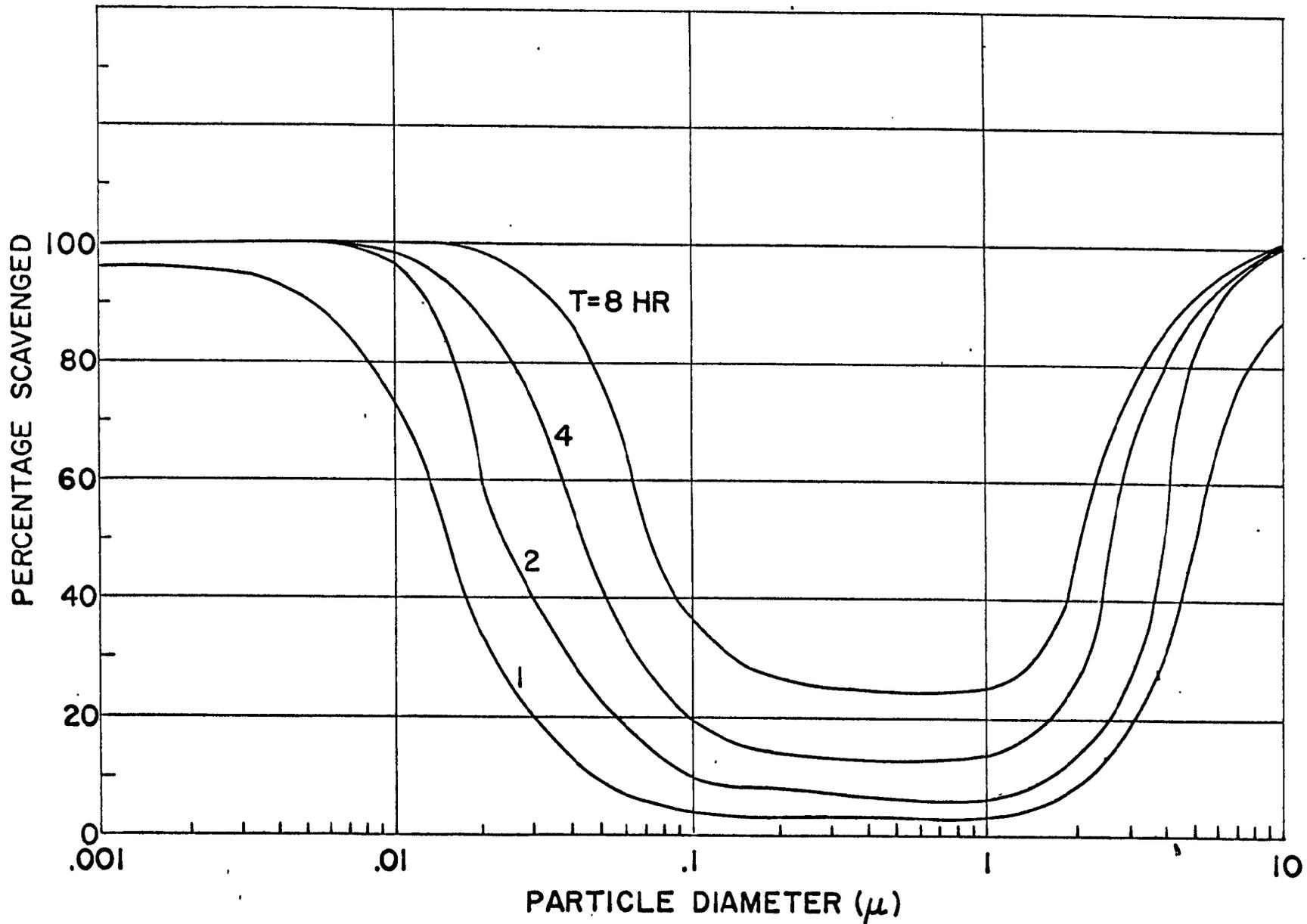


FIG 7