

OROGRAPHICAL AND CLIMATOLOGICAL INFLUENCES ON DEPOSITION OF NUCLEAR-BOMB DEBRIS

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ABSTRACT

The behavior of nuclear bomb debris in the atmosphere is reviewed in order to see how far such debris might be used as a tracer in atmospheric physics. Much debris seems to be captured by orographic precipitation, and information about the development of orographic clouds might be found by measurements of radioactivity in precipitation.

Deposition of bomb debris from the stratosphere is examined by available world-wide measurements of long-lived radioactive isotopes. The simple assumption that Sr^{90} is more rapidly brought down to earth by precipitation than is Cs^{137} is shown to be reasonable. This might explain Norwegian, Danish, and British measurements of the $\text{Cs}^{137}/\text{Sr}^{90}$ ratio in precipitation, provided that various climatic factors are taken into account. This interpretation of the $\text{Cs}^{137}/\text{Sr}^{90}$ ratio together with measured total Sr^{90} deposition is in agreement with accepted latitudinal flow models for low and medium latitudes.

1. Introduction

When radioactive particles from detonated nuclear bombs were found for the first time all over the earth, optimistic prophecies were made: meteorologists had been presented with tracer materials. Up to now, however, use of these tracer materials has been limited.

Measurements of radioactivity are more difficult and more expensive than ordinary meteorological measurements. This could no doubt be accepted if the meteorologists felt convinced that radioactive bomb debris provided satisfactory tracer materials.

This may not be the case.

The total radioactivity from a single explosion or the total activity in the whole atmosphere is not known at all accurately.

Insertion in the atmosphere, time and place, is probably not suitable from a scientific meteorological point of view.

Representative sampling at a given time and place is difficult, especially above the surface of the earth.

Radioactive particles cannot be considered as conservative qualities of the air masses. The content depends at least on precipitation, and it is not yet certain that the particles themselves have no influence upon weather development.

Even if bomb debris is not the best tracer, it would be unwise to neglect its possibilities completely. To see what might be possible, one must know the mechanism of particle formation and have some idea of the distribution of particles in the atmosphere.

2. Formation and spread of particles

A nuclear detonation liberates large amounts of energy. The bomb material, the adjacent air and, in

some cases, earth and stone, evaporate and form a ball of fire. This is cooled by radiation, expansion, and mixing with other air, the vapors condense, and particles are formed.

Condensation temperature is estimated at about 2500K in a "Sunshine"-report [1], and for a nominal bomb (20 kiloton TNT equivalent) this should occur after about 3.5 sec; for a 1-megaton bomb, after 25 sec; and, for a 10-megaton bomb, after 80 sec.

The hot masses ascend rapidly and are further cooled, mostly by mixing with adjacent air. It is generally accepted that for bombs in the megaton class most of the debris is carried above the tropopause. For smaller bombs exploded near the ground, most of the debris remains in the troposphere.

Some of the particles formed are large and fall rapidly as close-in fallout. World-wide radioactive deposition is composed of smaller particles for which the velocity of fall is often negligible. Other mechanisms are thought to be responsible for bringing these particles down to the earth.

The zonal arrangement of the currents in the atmosphere prevents rapid latitudinal spread of particles, and one might for some time expect to find most of the debris not very much displaced from the explosion latitude. However, the spread of particles may not be considered insignificant. Machta, List, and Hubert [2] have sketched trajectories after two explosions in equatorial Pacific in 1952 and 1954. The spread in one case was over 50 deg of lat and in the other case over 70 deg of lat. Measured deposition agreed well.

However, it is possible that particles from explosions at higher latitudes (e.g., Nevada or Siberia) might

keep themselves closer together near the latitude of the explosion.

In connection with bomb debris, it is customary to distinguish between particles originally transported to the stratosphere and particles which came to rest in the troposphere.

The residence time in the atmosphere cannot yet be calculated from direct deposition measurements. By means of 18 weeks of concentration measurements made by aircraft over the Atlantic ocean, Stewart, Crooks, and Fisher [3] have estimated a 22-day half-life for tropospheric particles from Russian and American explosions in the autumn of 1951. The corresponding mean life value of about 1 month can be accepted as residence time for the particles in the troposphere only if no appreciable spread southward has occurred. However, spreading might well be expected and should lead to an increase in the quoted figure.

Another problem is the longitudinal spread of particles. Measurements of the radioactivity in precipitation show great variations during months after termination of an explosion series. This indicates more or less diffused particle clouds in the troposphere.

Because of greater stability in the stratosphere, the vertical turbulent transport is expected to be less than in the troposphere. This should result in a much longer residence time. Direct measurements made by aircraft in the lower stratosphere show very high bomb-debris concentration, indicating that the stratosphere may act as a reservoir [3; 4]. This should be most evident for long-lived fission products long after bomb explosions have taken place.

Horizontal spread in the stratosphere is a difficult question. The great variations in measured concentrations in the lower stratosphere might be interpreted as due to exchange between the cleaner tropospheric air and a reservoir above the aeroplane altitude. However, weather maps from the stratosphere seem to be less impressed by eddies suitable for spreading particle clouds. One of the highest radioactivity levels recorded by the Norwegian Defence Research Establishment indicates concentrated clouds in the stratosphere. The particles were shown to be 3 months old by decay analysis. It is possible that more-or-less diffuse clouds exist for years in the stratosphere. After such a long time, the latitudinal distribution of the clouds is probably somewhat arbitrary.

Radioactive particles in the stratosphere gradually enter the troposphere, most probably through air exchanges. The residence time in the troposphere might be expected to be longer than for originally tropospheric particles because the former enter at the top of the troposphere and the latter might have an initial distribution centered well below the tropopause.

Measurements of ground deposition indicate that

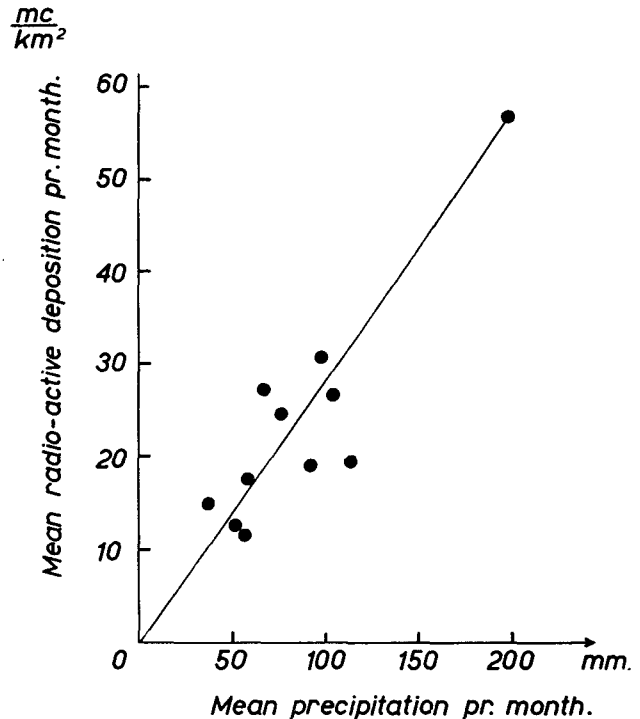


FIG. 1. Scandinavian deposition *versus* precipitation, 1957.

radioactive particles are brought down mostly by precipitation. The determining agents in deposition seem to be a combination of radioactive-particle supply and amounts (and probably types) of precipitation. Supply might predominantly reflect stratospheric or upper tropospheric conditions, while precipitation reflects lower tropospheric conditions. The relative importance of upper-level and lower-level conditions might change with place and problem.

If deposition and rainfall are compared over limited districts, there seems to be a surprising correspondence for longer times. Fig. 1 shows monthly means of precipitation *versus* radioactive bomb-debris deposition for Swedish and Norwegian stations in 1957. A linear relationship seems to cover the measurements, indicating no significant differences in conditions affecting supply from above Scandinavia.

3. Orographical effects

At present, it is difficult to see how atmospheric bomb debris can be successfully used to follow weather development. On the other hand, if deposition of radioactivity shows definite patterns or trends, increased understanding of certain phenomena might be obtained.

In fig. 2, observations from the weather ship "Polar-front," 66N-2E, are compared with observations from stations along the Norwegian coast. Both precipitation and deposition are greater at the land stations,

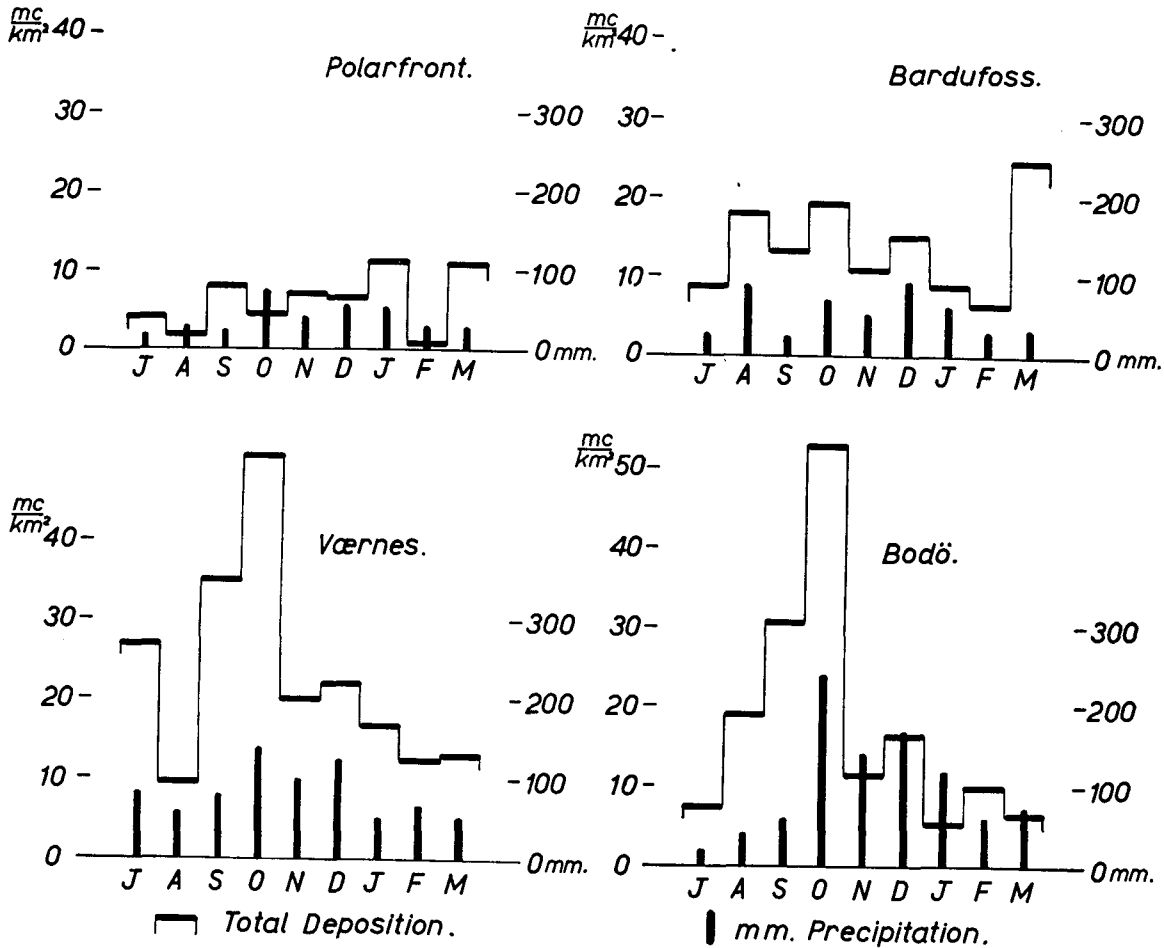


FIG. 2. Deposition measurements.

which may indicate that land areas, or rather coastal areas, have more than the average amounts of deposited bomb debris.

This question might be approached from another angle. Aircraft measurements of air concentrations make it possible to estimate the tropospheric content of activity in the area concerned, and this can be compared with deposition.

Calculations based on measurements for January and May 1957 over southern Norway showed that the content of radioactive bomb debris in the troposphere above the country could be deposited within two weeks. When compared with the earlier estimate of mean residence time of more than one month in the troposphere, this indicates a deposition rate over southern Norway which is higher than the mean in the westerlies.

The large deposition is probably a climatological feature. Norway is situated on the Atlantic seaboard in an area where moist onland winds are frequent and orographical precipitation is favoured. The maritime air masses might even be relatively rich in radioactive bomb debris in the lower layer because of reduced precipitation and washout over the ocean.

Western Norway is very suitably situated for studying orographic effects because the mean westerly air current is forced to pass a north-south mountain ridge. Measurements are available from two stations, Bergen on the coast and Finse near the crest of the mountain ridge. There appears to be less radioactive bomb debris per liter precipitation at Finse than at Bergen during winter, but no significant difference has been noted during summer. The same effect has been found for another pair of stations, Vaernes and Storlien, which are somewhat similarly sited. This is shown on fig. 3 where monthly means of the ratios between deposition per liter precipitation are plotted as a function of time for the two pairs of stations.

As shown on fig. 4, such an effect might be explained by the more stable wintertime air masses. When streaming from the coast towards the mountain ridge, more and more precipitation is exhausted from the air elements without new air being drawn into the precipitating cloud. As the radioactive particles are gradually washed out, the activity per liter precipitation decreases towards the mountain top.

The more unstable summertime air masses might

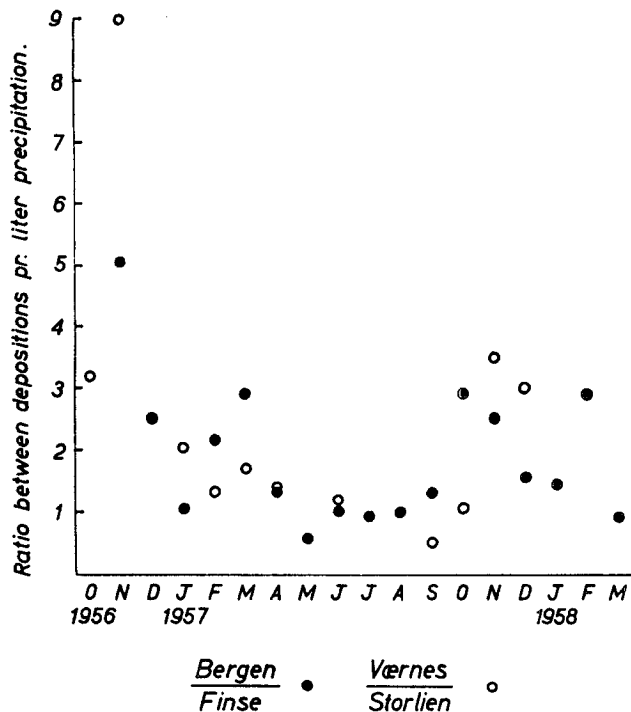


FIG. 3. Ratios between specific deposition at coast and mountain stations.

be expected to show no appreciable clean-up effect because of new air being steadily drawn into the cloud systems.

Between Bergen and Finse, no continuous monitoring of radioactive particle deposition is made at present. During the summer of 1956, a few snow samples were collected in the mountains near Voss, midway between Bergen and Finse. These showed much higher activity than similar samples from Finse, indicating an orographic effect.

4. Selective deposition

The possibility of selective fission-product transport in the atmosphere is much debated. Such a transport may be brought about because of the tendency on the part of certain isotopes to form part of larger or smaller particles, or because of differences in volatilities, moisture absorption, etc. Capture by precipitation might also be selective. S. M. Greenfield [5] has, for instance, shown that particle size is a determining agent in capture by water drops.

The rate at which a certain type of particle is brought down to earth from an air element might be written as

$$D_1 = \frac{-dN_1}{dt} = a \cdot N_a \cdot \exp(-at) \quad (1)$$

where N_a is the initial amount of particles, t is time, and a is a rapidity constant. It is assumed that no new particles enter the element.



FIG. 4. Particle distribution in stable orographic precipitation area (in outline).

Another type of particle may not be so easily brought down. If the new rapidity constant is b , $b < a$, then the ratio between the deposited quantities of the particle types is

$$\frac{D_1}{D_2} = \frac{a \cdot N_a}{b \cdot N_b} \cdot \exp[-(a - b)t] \quad (2)$$

The initial deposition will consequently show an excess of the particles most easily brought down, the position being reversed later on.

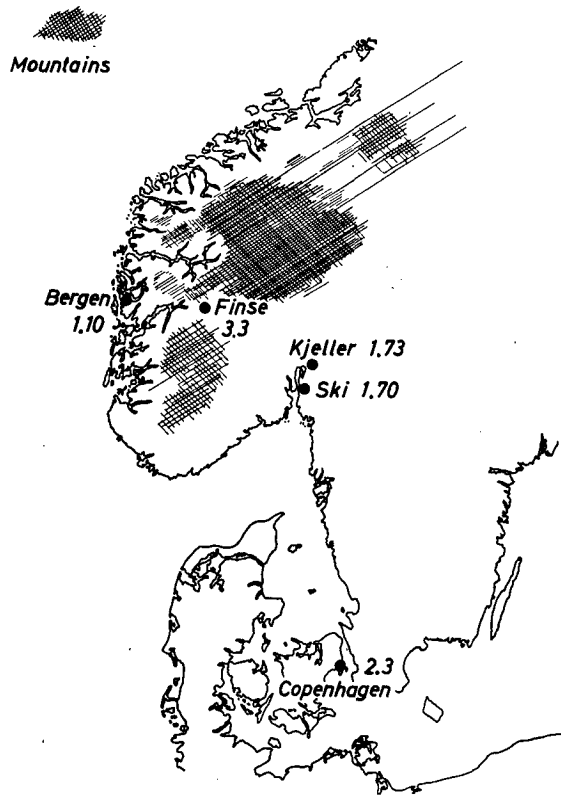
This effect will not appear over an area with a closed air-mass circulation with even or arbitrarily distributed precipitation, because all debris brought down into clouds must be deposited eventually. The air concentration of the more slowly captured particles will show a relative increase until the rate of entry equals the rate of removal.

In a district with rather fixed precipitation conditions, the circumstances may be otherwise. In an area of orographically strengthened precipitation, a previously-equilibrium mixture is set aside, and one might expect an excess of particles most easily brought down at the start of the precipitation area and an excess of the more slowly captured particles towards the end of the precipitation area.

Stable orographic precipitation areas mostly occur in the wintertime, and in western Norway certain effects might appear in the snow cover.

One snow sample from the Finse snow analysed in Harwell, England showed a Cs^{137}/Sr^{90} ratio of 3.3 when the value 1.5 was the expected equilibrium value. This might mean that Sr^{90} is more rapidly brought down by precipitation than Cs^{137} . The snow sample was taken in summertime, and the isotope content might have changed after the snow had been deposited because of melting or percolating rainwater.

However, analyses made by the chemistry division of the Norwegian Defence Research Establishment provide a further indication. Cs^{137}/Sr^{90} ratios in precipitation during the autumn of 1957 were as follows [6].

FIG. 5. Cs^{137}/Sr^{90} ratios.

(preliminary values; calibration will be changed):

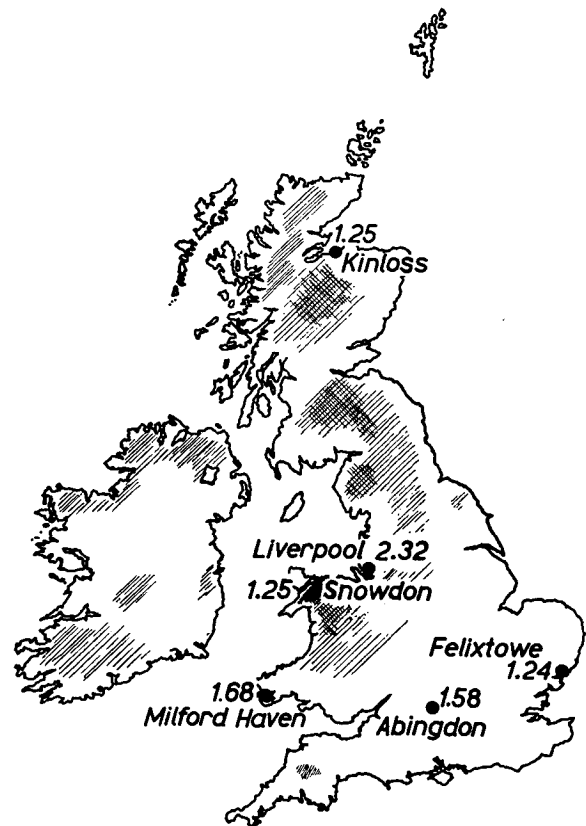
357-mm precipitation in Bergen, mean value 1.10
 160-mm precipitation at Kjeller, mean value 1.73
 78-mm precipitation at Ski, mean value 1.70.

The figures are plotted on the map (fig. 5).

Orographic precipitation very often begins near Bergen, so that a low value of the ratio might be expected. Kjeller and Ski, lying in southeast Norway, have less predominantly orographic precipitation. However, the air masses giving precipitation here have at first often given precipitation in the western and southern parts of southern Norway. This should wash out some of the Sr^{90} content and give a higher value for the Cs^{137}/Sr^{90} ratio.

A corresponding observation [7] was made in Copenhagen, where the Cs^{137}/Sr^{90} ratio was found to be 2.3 in precipitation falling between October 1957 and January 1958. The general moist westerly air currents can be expected to give some orographic-influenced precipitation before reaching Copenhagen from the west coast of Denmark. The high value is therefore reasonable, but it must be emphasized that no value from the western part of Denmark is available with which the figure might be compared.

Stewart, Osmond, Crooks, and Fisher [8] have published measurements from several English stations for

FIG. 6. Cs^{137}/Sr^{90} ratios.

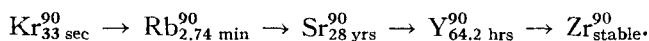
1956. From the map given in fig. 6, two stations especially seem to be orographically influenced in the west-southwesterly air current which is the most common source of precipitation. These are Snowdon, lying on the windward side of the Welsh mountains where the Cs^{137}/Sr^{90} ratio is 1.25, and Liverpool, lying nearly on the leeward side of the Welsh mountains where the ratio is 2.32.

The expected ratio is about 1.5, and the measurements might be explained if it is accepted that Sr^{90} is more easily deposited in the orographical precipitation area formed by the Welsh mountains, so that air masses reaching Liverpool give a relatively larger Cs^{137} deposit.

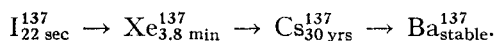
5. Formation and deposition of Sr^{90} and Cs^{137}

The observations quoted are sparse, they cover short periods, the degree of representativity is not examined, and sampling and analysis procedures might give place for errors. One might wonder, however, about possible differences between Sr^{90} and Cs^{137} which might cause selective capture. This can, for instance, be brought about because of differences in chemical and physical properties. The conditions just after explosion might, for instance, be responsible for a possible size difference leading to different behaviour in the atmosphere.

The decay scheme of which Sr^{90} forms a part is



The corresponding scheme for Cs^{137} is



The half-life of each isotope is given beneath the chemical symbol.

Following the fission (splitting) of a heavy atomic nucleus, two daughter nuclei are usually formed, both of which generally have too many neutrons to be stable. The isotopes occurring first in the decay chains tend to be formed directly, and most of the Sr^{90} and Cs^{137} are formed by decay from these.

Kr and Xe are noble gases and will take very little part in particle formation. The condensation phase occurs less than 5 sec after the detonation of a nominal bomb, when most of the isotopes of mass number 90 and 137 exist as noble gases or their predecessors. Following decay of the noble gases, the chemically active elements Rb and Cs are formed in atomic form and are apt to be adsorbed and to react with particles present.

Capture by particles is presumed to occur by diffusion, and this will favour deposition on smaller particles. It is generally accepted that Cs^{137} and Sr^{90} occur on smaller particles than do most other isotopes.

However, it is probably the bombs in the megaton class which are responsible for most of the Sr^{90} and Cs^{137} existing in the atmosphere, and for these large bombs the picture is different. In the decay chain for Sr^{90} , particle formation may start with Rb^{90} , which is formed from Kr^{90} with a 33-sec half-life. If the condensation process starts much later than this, most of the nuclei with mass number 90 will be able to take part in the condensation process, and the Sr^{90} atoms might not be expected to occur on especially small particles.

This is the case for a "typical" megaton bomb of size 10 Mton. Eighty sec elapses before the fire-ball cools to the condensation temperature of 2500K, and most of the later Sr^{90} atoms then exist as Rb^{90} .

In the Cs^{137} chain, the first isotope which forms permanent particles is Cs itself. Its predecessor has a half-life of 228 sec, and it seems that a very much larger bomb is necessary before Cs^{137} can be expected to take part in the condensation process to any significant extent.

It should therefore not be unreasonable to assume that Sr^{90} generally occurs on larger particles than Cs^{137} .

Greenfield [5] has calculated particle capture by water droplets and found a minimum capture efficiency for a particle diameter of 0.5μ . For larger particles, capture by coalescence is predominant; for smaller particles, capture by Brownian motion is predominant.

Capture by coalescence will favour large droplets because of the greater difference in fall velocity between particles and droplets. Capture by Brownian motion will, on the contrary, favour smaller droplets.

The median particle diameter for airborne material is given in a "Sunshine" report [1] as 1.2 and 2.2μ for two air bursts in the kiloton class. Larger bombs have longer condensation times, and it is possible that the particles are greater.

If the median diameter quoted is accepted as typical for Sr^{90} particles, these will predominantly be captured by larger droplets in a cloud. If the Cs^{137} particles are very much smaller, they should be collected predominantly by smaller cloud droplets.

From Langmuir's [9] or Mason's [10] tables for the coalescence collection efficiencies of droplets, it can be easily deduced that the larger droplets have a higher probability for reaching the raindrop stage. The smaller particles should therefore be more delayed in the downward transport by rain than the larger ones.

6. Climatological effects on deposition

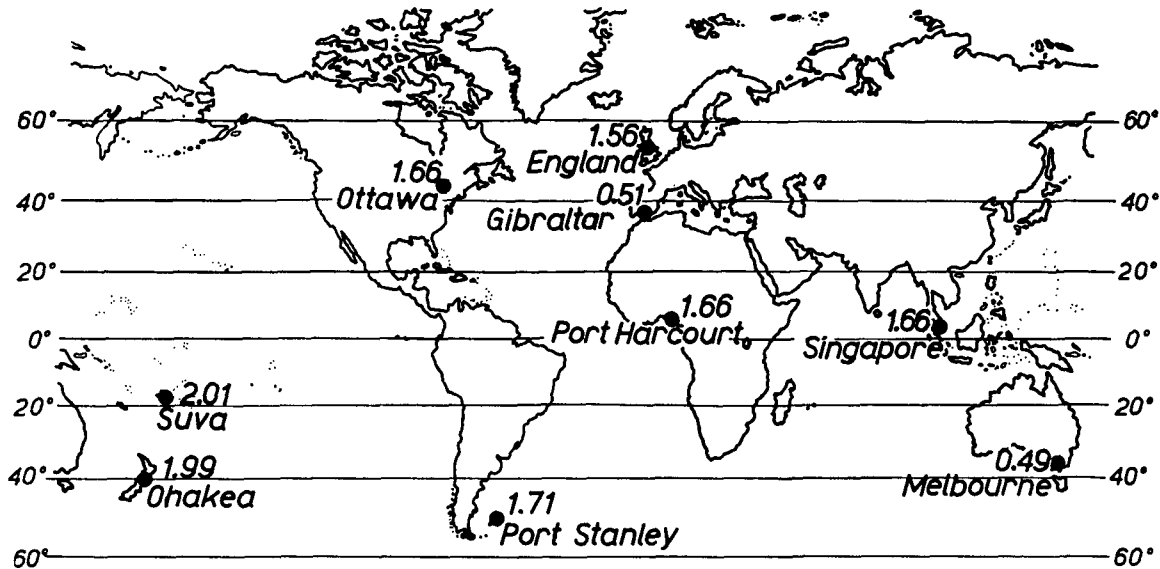
The foregoing examination demonstrates a real possibility that Sr^{90} in the atmosphere might exist in a form making capture by raindrops more rapid than is the case for Cs^{137} .

If this hypothesis is accepted, it is natural to look for a world-wide effect as well as for an orographical effect. Stewart *et al* [8] have published figures for the 1956 $\text{Cs}^{137}/\text{Sr}^{90}$ ratio in bomb debris deposited at a number of stations (fig. 7). Two stations, Gibraltar and Melbourne, have a remarkably low value of about 0.5 compared with the expected value of 1.5. These two stations have the Mediterranean-type climate, and it might be natural to look for climatological causes.

It was mentioned earlier that evenly distributed precipitation in a closed air-mass circulation would cause an equilibrium mixture of particles in the air, so that the amount of particles rapidly captured by rain would decrease. If air renewal is rapid enough, then the selective removal of particles by precipitation will not be sufficient to reach equilibrium mixture of particles in the air, and the rapidly captured particle types will outweigh others in the deposition.

With the distribution of radioactive bomb debris existing today, a rapid renewal of particles implies either a downward air current above a particular region or a steady advection of air from such a region with no significant rainfall produced on the way.

In the Mediterranean-type climate, it is quite possible that such a process is a contributing factor in the deposition occurring. This climate is closely connected with the subtropical highs, and a downward transport of air might well take place here. Rain is usually

FIG. 7. Cs¹³⁷/Sr⁹⁰ ratios.

sparse, and the more rapidly captured particles are apt to dominate the bomb-debris deposition. The very low value of the Cs¹³⁷/Sr⁹⁰ ratio measured agrees well with the hypothesis that Sr⁹⁰ particles are more easily brought down by rain than are the Cs¹³⁷ particles.

The effect must have a counterpart on a world-wide scale. The Sr⁹⁰ amount deposited causes a deficit of this isotope in the air compared with Cs¹³⁷, and the relative deposition of Cs¹³⁷ increases according to equation (2). Air flows out from the highs and can probably most easily be found in a later development stage in the tropics.

The three tropical stations on fig. 7, Port Harcourt, Singapore, and Suva, all have a Cs¹³⁷/Sr⁹⁰ ratio above the mean, but the deviations are not as remarkable as for Melbourne and Gibraltar.

7. World-wide deposition

It is difficult to compare direct measurements of deposited radioactivity made at different places at different times because of the higher activity of newer fission products. One way of avoiding the difficulty is to concentrate on long-lived fission products, whose total activity changes very slowly.

For biological reasons, the most examined long-lived isotope is Sr⁹⁰, with a half-life of 28 yr. The decrease in the amount of Sr⁹⁰ due to decay can for the moment be neglected.

Extensive measurements are made, especially by the Americans [11; 12]. Measurements of Sr⁹⁰ in soil, precipitation, and on sticky papers seem to have some common features.

It appears that very little Sr⁹⁰ is deposited in tropical areas. There seems to be a maximum somewhere be-

tween 30N and 50N and possibly a smaller maximum at the corresponding southerly latitudes. This might be caused by the latitudinal distribution of bomb-test sites, but calculations by Stewart *et al* [8] and by Machta and List [13] indicate that this is not the case. It is more likely that the distribution is affected by the general circulation.

It might be questioned whether Sr⁹⁰ is suitable for investigations on a world-wide scale because of its particle properties. Calculations made [1] show that a 1 μ -diam particle sp w 2 falls from a height of 30 km to 4 km in 5 yr. If the typical Sr⁹⁰ particle is larger, it is doubtful if the distribution of deposition will show effects of possible slow stratospheric movements. However, if the Cs¹³⁷ particles are smaller as assumed, they ought to show such effects better. The difference in the British Cs¹³⁷ and Sr⁹⁰ measurements are indeed interesting in this connection, but they are not conclusive.

Even if a residence time for Sr⁹⁰ in the atmosphere of only a few years is accepted, it is still interesting to see what type of flow is indicated by the measurements. A latitudinal flow model is shown in fig. 8.

The greatest deposition seems to be near the subtropical highs where the Cs¹³⁷/Sr⁹⁰ ratio is low. This indicates fresh air from above, and a regular downward displacement transport is postulated.

The smaller amounts deposited in equatorial areas and the high Cs¹³⁷/Sr⁹⁰ ratio here may be due to the lack of downward transport in this area. It seems that particle advection from the highs is reasonable in the lower layers.

North of the highs, deposition appears to decrease, but the Cs¹³⁷/Sr⁹⁰ ratio is not particularly high. Some

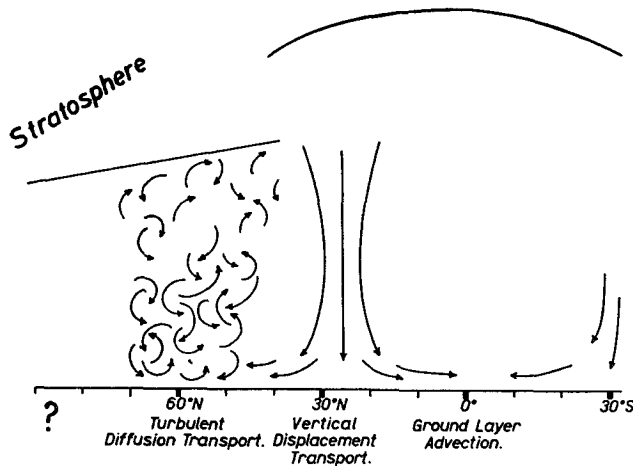


FIG. 8. Flow model.

sort of downward transport is thus necessary, and a large-scale turbulent spreading transport is to be expected in the westerlies. North of this area, lack of published observations makes it impossible to give any flow model based on bomb-debris measurements.

The model presented agrees with models for the general circulation deduced from other types of measurements. It might not be extended to areas not covered by observations such as, for instance, Asia and the Pacific.

If distribution of bomb debris over the earth's surface is determined by the general circulation, variation in strength and position of the main currents should be reflected in deposition of bomb debris. Examination of measurements reveals a seasonal variation.

Fig. 9 shows means of monthly measurements made at Milford Haven from 1954 to 1957 [8], Pittsburgh from 1955 to 1957 [12], and one place in Holland from 1956 to 1957 [14]. The figure may be typical for stations in the westerlies.

One might keep in mind that, if the mechanism of transport from above is different in two regions, the time delays of upper-air variations can be expected to be different. In addition, where transport is affected by turbulent spreading, the amplitude at the surface should be less than where orderly transport occurs.

Time variation might therefore differ somewhat between the westerlies, the subtropics, and the tropics, or perhaps even within the same zone for different climates.

8. Conclusion

When considering the measurements made in the light of the hypothesis put forward in this paper, it is necessary to look not only at the situation of the stations in relation to the general circulation but also for possible orographical influences. Ohakea and Port Stanley lie to the leeward side of large islands, and the

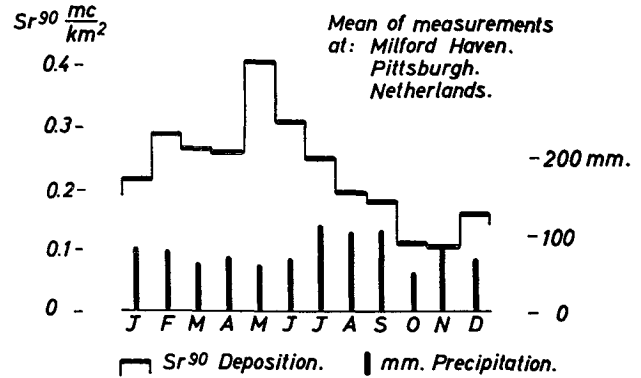


FIG. 9. Time variation of Sr⁹⁰ deposition.

high Cs¹³⁷/Sr⁹⁰ ratio might be caused by orographical precipitation areas.

It seems logical to apply the considerations on a more extensive "orographical" scale. In the general circulation, all west coasts within the westerlies might be expected to show a frictional increase in precipitation, causing in coastal areas an increase in deposition, and also decreasing fractions of easily captured isotopes with distance from the coast, until renewal of the particle content from above outweighs the clean-up effect. The same effect might occur at east coasts in the trade-wind zone and at coasts in the monsoon districts with onland winds.

No mechanism for air exchange between the stratospheric reservoir and the troposphere has been invoked in the model presented. Supply might be via eddies connected with the subtropical jet-stream and by air motions above cyclones in higher latitudes. This would cause maximum amounts of radioactivity in late wintertime in the upper troposphere, at the end of the period with strongest atmospheric activity, agreeing with the observed ground maximum in springtime.

It is hoped that further and more extensive measurements of bomb-debris distribution and deposition will lead to better understanding of the mechanisms involved and that this will again lead to increased knowledge both of the general circulation and local effects.

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