Zeitschrift: Bulletin der Schweizerischen Akademie der Medizinischen Wissenschaften = Bulletin de l'Académie suisse des sciences médicales = Bollettino dell' Accademia svizzera delle scienze mediche

Herausgeber: Schweizerische Akademie der Medizinischen Wissenschaften

Band: 14 (1958)

Heft: 5-6: Symposium sur les effets nocifs de faibles doses de radiation : éléments physiques et aspectes biologiques = Symposium über schädliche Wirkungen schwacher Strahlendosen : physikalische Grundlagen und biologische Aspekte = Symposium on noxious effects of low level radiation : physical elements and biological aspects

Artikel: The world-wide deposition of long-lives fission products from nuclear test explosions

Autor: Stewart, N.G.

DOI: https://doi.org/10.5169/seals-307383

IIa) Distribution des produits de fission
Verteilung der Fissionsprodukte – Distribution of the fission products

Président – Präsident – Chairman:
Dr. W. G. Marley, Harwell

D. C. 539.17:[613.2:611.15]

Atomic Energy Research Establishment, Harwell

The World-wide Deposition of Long-Lived Fission Products from Nuclear Test Explosions

By N. G. Stewart
Health Physics Division

1. Introduction

Measurements of the radioactive dust from nuclear test explosions have been made in the U.K. since 1948. In those early years, it was considered sufficient to measure the gross radioactivity of the atmosphere by drawing air through filters and counting the radioactivity of the collected dust on a simple Geiger counter. By 1951, some of the interest was transferred to deposited material and two stations were set up within the U.K. at which rainwater was collected and filtered and the gross radioactivity again assayed on a Geiger counter system. Then in 1954, the importance of the long-lived isotopes Sr^{90} and Cs^{137} became apparent and at one of the U.K. stations, rainwater was collected over monthly periods and analysed for Sr^{89}, Sr^{90} and Cs^{137}. About the middle of 1955 it was decided to study fallout rates throughout the U.K. and throughout the world, and from then on the number of sampling stations was progressively increased until it reached its present level of 6 in the U.K. and 13 in other parts of the world.

This paper describes the most significant results obtained at the various stages of this expanding project. Some of the important features of the dust clouds and their dependence on meteorological factors will be described. It will be shown that the world pattern of deposition is strikingly non-uniform and hence that global surveys are essential for a true assessment of the fallout problem.

2. Long-Range Air Contamination from Nuclear Test Explosions

When a "nominal" bomb is exploded in the middle latitudes of the northern hemisphere, the dust cloud rises to a height of perhaps 10 000

to 12000 m and travels east with the prevailing westerly winds. As the cloud travels it diffuses both laterally and vertically and ultimately contaminates the lower atmosphere across a broad front at great distances. Although on the average the cloud from, say, a Nevada explosion crosses U.K. on the fifth day after time of burst and thereafter every 4 to 7 weeks as it circulates round the world, the behaviour of any one cloud can depart considerably from this average and to obtain a general idea of the way in which the concentration of dust varies with distance downwind or with time, it is convenient to consider the concentrations over U.K. of the fission products from a series of explosions whose times of burst are close enough together to allow them to be considered as a single event, at least throughout a period starting some 3 to 4 weeks after the mean time of burst.

The specific activity of the air above U.K. in the period following the Russian and American tests in the autumn of 1951 has been plotted in fig. 1A. In this diagram, time is measured from the mean date of the individual explosions, and the specific activity of the air has been averaged over periods of approximately 14 days to smooth out short-period variations. In this presentation, earlier points are omitted since the method cannot reasonably be used within the actual period when the explosions are occurring. A smooth curve has been drawn through the experimental points and it has been found by experience that the shape of this curve is quite characteristic of explosions of near nominal size which take place in the middle latitudes of the northern hemisphere. The curve has been corrected for the radioactive decay of the fission products according to a composite decay curve computed from the sum of the individual decay curves, giving the dotted line in fig. 1 which can then be taken to represent the decrease with time of the specific air content of the dust itself or of the longer-lived isotopes.

The decrease with time of the specific dust content of the air over the U.K. can be due to increased spread of the cloud or to deposition or both. Vertical spread above 12000 m is prevented by the very low diffusion rates in the stratosphere (see below). There is also a barrier of stable air in the lower atmosphere (troposphere) reaching as far as 25° N of the equator which prevents widespread diffusion to the south. Finally, direct observations of the lateral spread of Nevada clouds by sampling planes based in Gibraltar and in Scotland suggest that lateral diffusion is virtually complete by the time the clouds have made one circuit of the world. All the points in fig. 1 refer to clouds which satisfy this criterion and we must conclude that the dotted line in fig. 1A represents the rate of removal of atmospheric dust from the atmosphere by

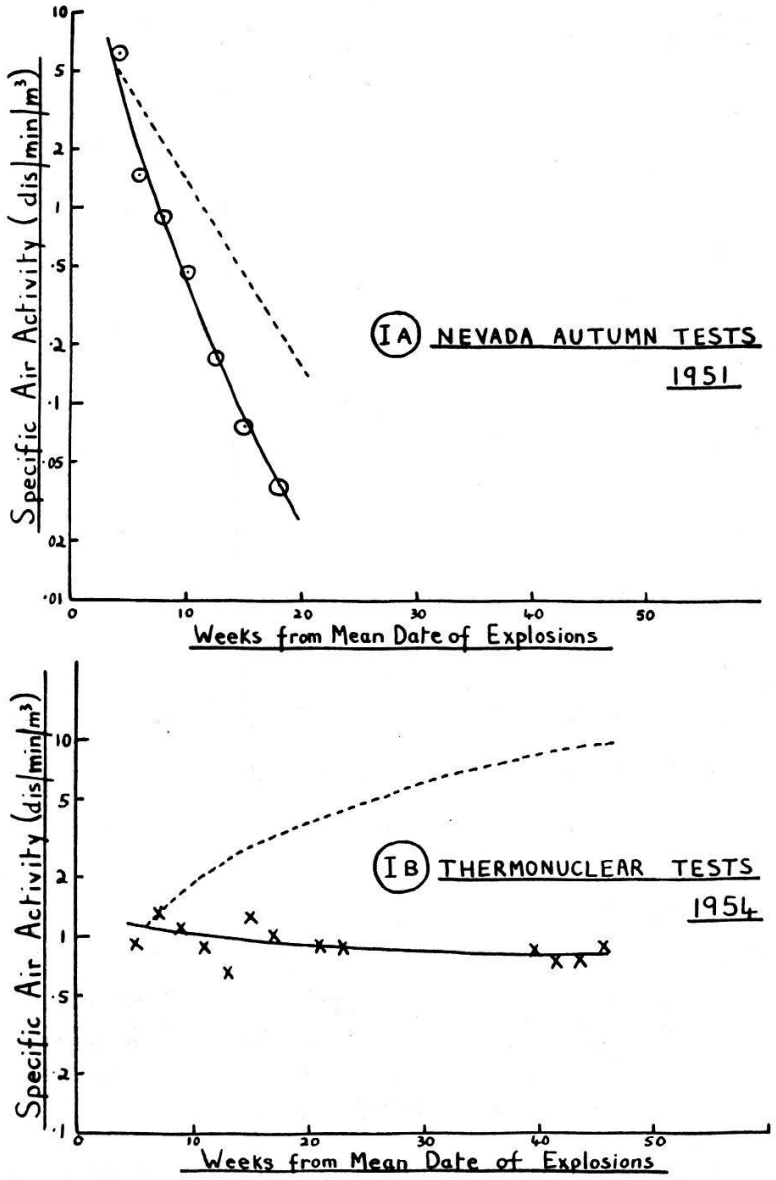

Fig. 1. Specific activity of Atlantic air following nuclear explosions.

deposition alone and that half the material is removed in approximately 22 days. The behaviour of the clouds from thermonuclear explosions is quite different. The fortnightly averages of the specific air activity following the Pacific tests in the spring of 1954 have been plotted in fig. 1 B. There is no sharp decrease of airborne activity such as was obtained after the Nevada explosions, and when the correction is put in for radioactive decay it will be seen that the concentration of dust has in fact been increasing throughout the 10 months following the mean date of the explosions. The explanation is that a thermonuclear dust cloud penetrates into the stratosphere and reaches a height of the order of 30 000 m soon after the explosion. Diffusion is a very slow process in the stratosphere and material returns to the lowest layers of the atmosphere at a much slower rate than in the case of the nominal bomb cloud.

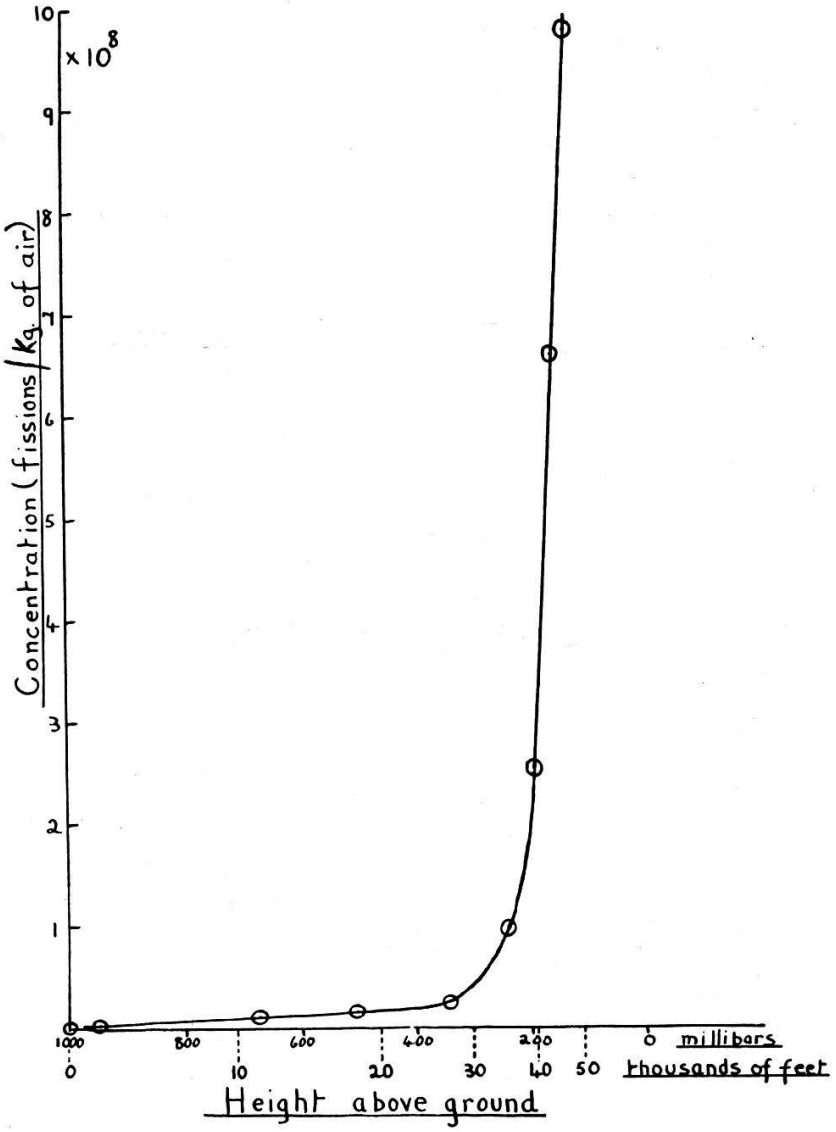

Fig. 2. Vertical distribution of fission products in atmosphere August to September 1954.

The gap in the record in fig. 2 occurs during the period when the lower atmosphere was further contaminated by the explosion of several Russian nominal-sized bombs in the late autumn of 1954. This contamination decreased in the manner shown in fig. 1A for this type of bomb, enabling measurements to be resumed on the residual thermonuclear contamination in January and February, 1955.

Direct confirmation of the hypothesis that the thermonuclear explosion dust is held back in the stratosphere has been provided by a series of experiments conducted in the autumn of 1954 and repeated in the spring and autumn of 1955. In these experiments the concentration of activity was measured at various heights in the atmosphere up to 15 000 m. The mean concentrations, averaged over a period of 2 months in the autumn of 1954, are shown in fig. 2 which demonstrates quite

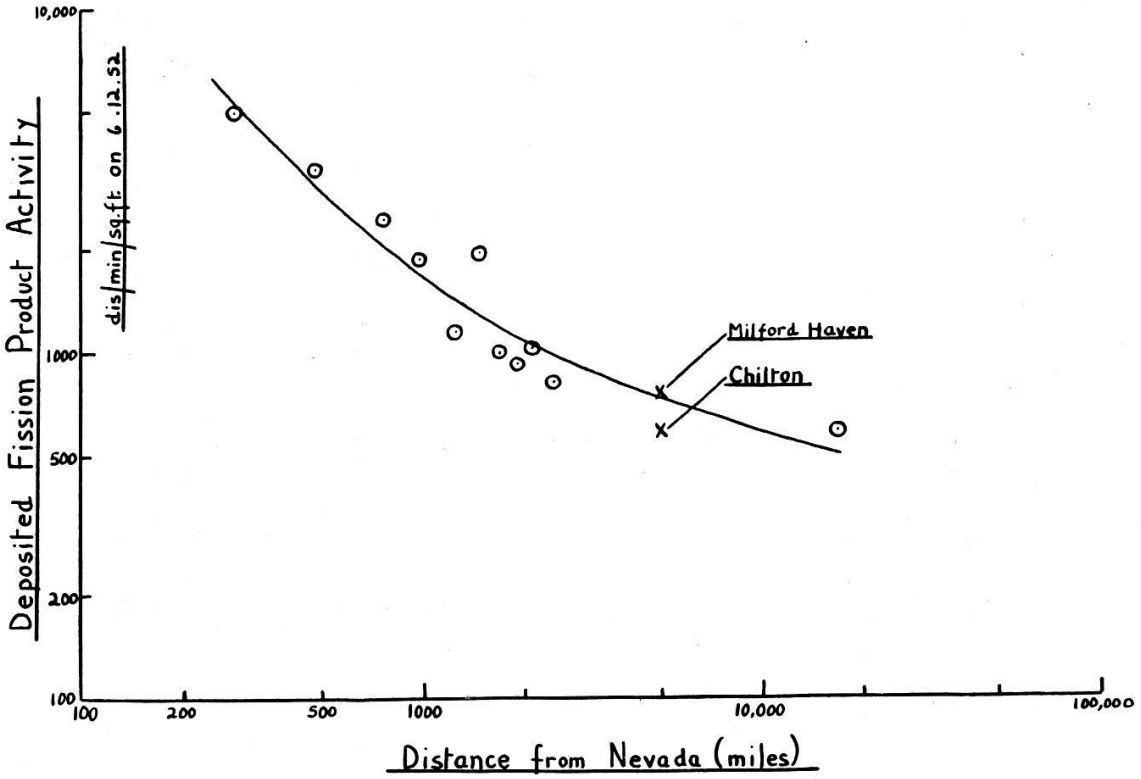

Fig. 3. Deposition from Nevada tests, 1952. Variation with distance.

strikingly the existence of large accumulation a of fine dust in the stratosphere several months after the tests. The origin of the dust was identified by chemical analysis of the fission products.

3. Deposition of Radioactive Dust from "Nominal" Test Explosions

Eisenbud and *Harley* (1) have published figures for the total deposited radioactivity at 121 stations within the United States in the period following the eight test explosions in Nevada in 1952. The average values of the deposited fission product activity were determined at various distances between 300 and 2 400 miles downwind from the test site and these have been plotted in fig. 3. The one additional point plotted at 17 000 miles is the average deposition on the U.S. west coast and represents the deposition of material which has travelled once round the world, together with that due to any temporary "back-flow" from the test site. The deposition figures obtained at the two sampling stations in the U.K. are also shown and the general agreement with the U.S. data is very satisfactory. The graph shows a marked decrease in deposited radioactivity with distance so that the mean deposition at the U.K. stations is about 10 per cent of that occurring about 200 miles from the test site.

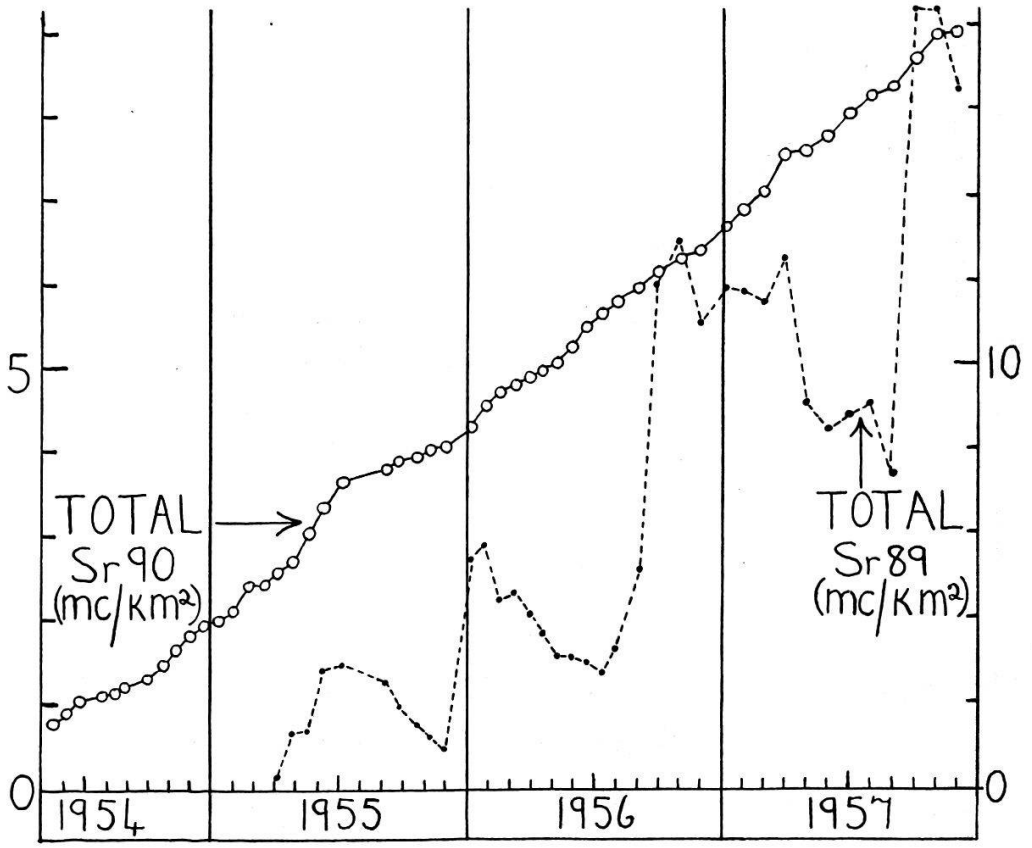

Fig. 4. Cumulative deposition in the U.K.

4. Deposition of Radioactive Dust from Large-Scale Test Explosions

a) Deposition Measurements in the United Kingdom

Rainwater has been collected over monthly periods at Milford Haven, U.K., since May 1954, and analysed for Sr^{89}, Sr^{90} and Cs^{137}. In general, the amount of Cs^{137} deposition maintains a steady ratio of 1.7 relative to Sr^{90} so that the general character of the observations can be described with reference to Sr^{90} only.

The cumulative curves of deposition of Sr^{89} and Sr^{90} are plotted in fig. 4. In the successive yearly periods between May 1954 and May 1957 the amount of Sr^{90} deposited was 2.06, 2.24 and 2.55 mc/km² respectively and the cumulative total in October 1957 was 8.6 mc/km².

By examining the ratio of Sr^{89}:Sr^{90} in the monthly samples it is possible to show that the deposited Sr^{90} has been derived mainly from old fission products which have been stored in the stratosphere and that the contribution of "nominal-class" explosions to the total is less than 10 per cent. Most of the Sr^{90}, at least up to the end of 1956, can be shown to be derived from the tests which occurred in 1954.

The specific Sr^{90} content of Milford Haven rainwater has been plotted

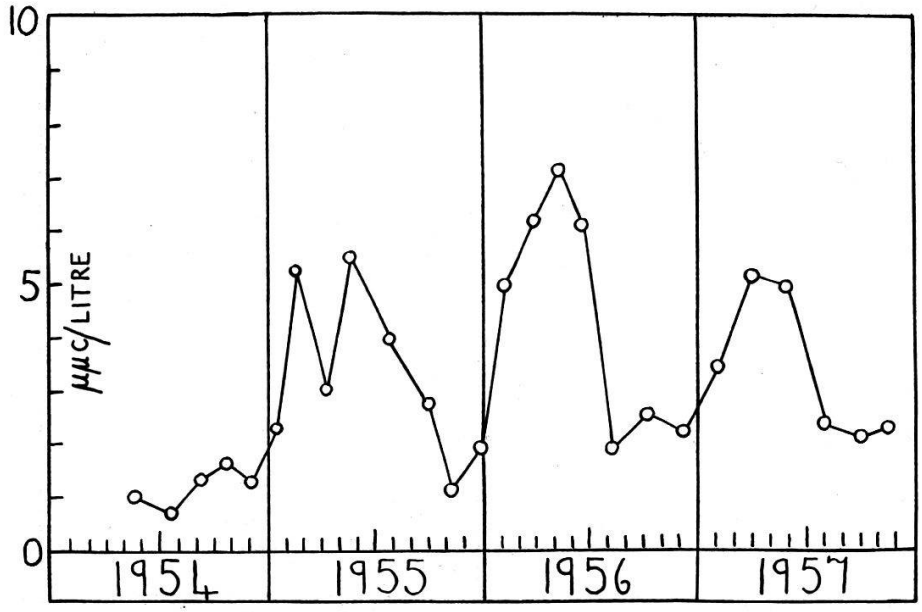

Fig. 5. Sr^{90} in rain.

against time in fig. 5. The graph reveals a marked seasonal variation in the specific activity of the rainwater with peaks in the late spring and troughs in the late autumn of 1955, 1956 and 1957. This result suggests that the dust stored in the stratosphere is returned to earth in a periodic manner and not at a uniform rate as previously envisaged.

The results obtained at the other five U.K. stations suggest that the differences in the amounts of Sr^{90} deposited at each reflect differences in rainfall rather than differences in the specific contents of the rainwater. In the following table, the overall specific activities of the rainwater at the various sites have been expressed as percentages of that at Milford Haven over the common period of sampling. Rainfall rates have been similarly expressed.

Station:	Kinloss	Liverpool	Snowdon	Abingdon	Milford Haven	Felixstowe
Relative specific activity of rain	90	88	80	106	100	102
Relative rainfall rates	89	84	409	68	100	42

The specific activity of rain is apparently insensitive to the amount of rain which falls, and there are therefore good grounds for believing that the cumulative deposition of Sr^{90} at any point in the U.K. will be proportional to rainfall. The results of *Bryant* et al. (2) obtained from the radiochemical analysis of soils confirm this.

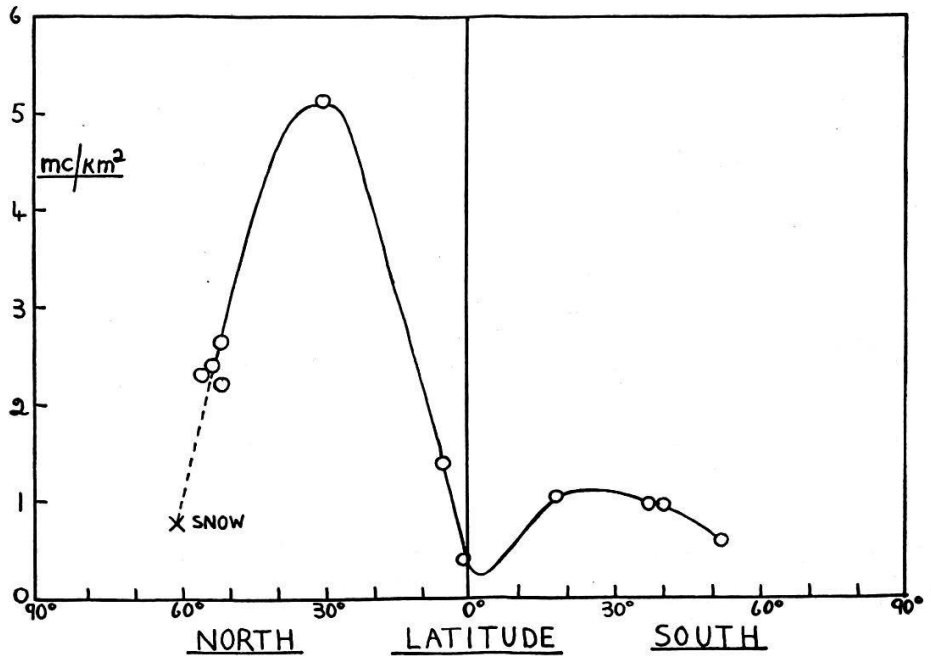

Fig. 6. Total deposition of Sr^{90} in 1956 at various latitudes.

b) Deposition Measurements on the World Network

The most striking result of the world network measurements is that very little Sr^{90} is being deposited in equatorial regions as compared with the middle latitudes of the northern hemisphere. Since the stations were commissioned at different times it is difficult to present the observations in a straightforward manner but fig. 6, in which the total amount of Sr^{90} deposited at the various stations in 1956 is plotted against latitude, illustrates the general nature of the results.

The specific activity of the rainwater has been averaged over the total period of sampling at each station and normalized relative to that at Milford Haven over the common period. The curve is shown in fig. 7 which has the same general characteristic as the previous diagram but with a more pronounced equatorial minimum. Since a smooth curve can be drawn through the points, despite large differences in the longitudes of the sampling stations, it is reasonable to suppose that the mean concentration of Sr^{90} in rainwater is a function of latitude only. Fig. 7 can then be used to compute the accumulated fallout at any place remote from a test site when the latitude and the rainfall at the site are known. If the rainfall at the site is markedly seasonal, due allowance must be made for the seasonal effects shown in fig. 5.

An interesting result is demonstrated in fig. 8 in which the specific activities of rainwater at Milford Haven and at Ohakea in New Zealand have been plotted. The Milford Haven curve is a portion of the curve in fig. 5 showing the pronounced seasonal effect. The New Zealand curve

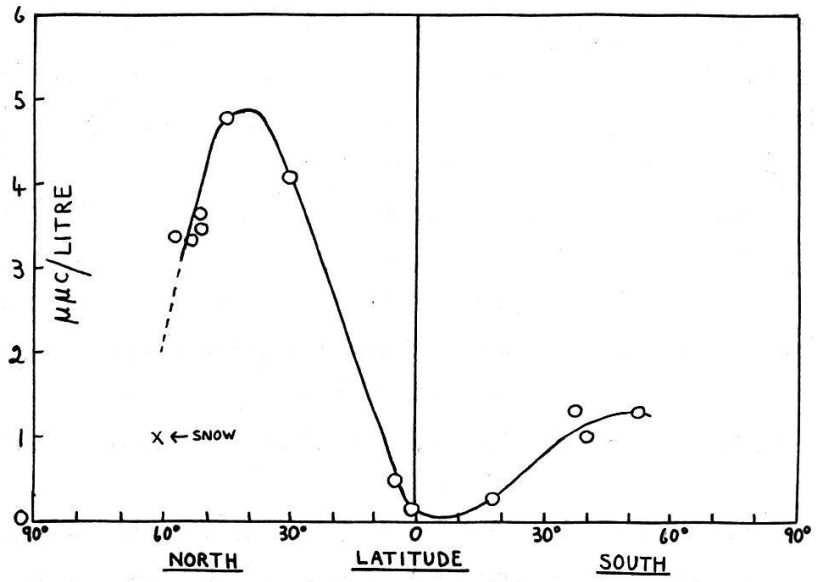

Fig. 7. Mean Sr^{90} content of rainwater at various latitudes 1955–1957.

also shows a seasonal effect but of the opposite phase to that in the northern hemisphere.

5. *Sr^{90} and the General Circulation of the Atmosphere*

The programme of measurements has revealed general facts about the Sr^{90} in the atmosphere, in rainwater and on the surface of the earth:

a) Since 1954, nearly all the Sr^{90} deposited at places remote from test sites has been derived from large-scale nuclear explosions and has been fed gradually from the stratosphere.

b) The concentration of Sr^{90} in rainwater, and hence in tropospheric air, shows a genuine seasonal variation, having opposite phases in the

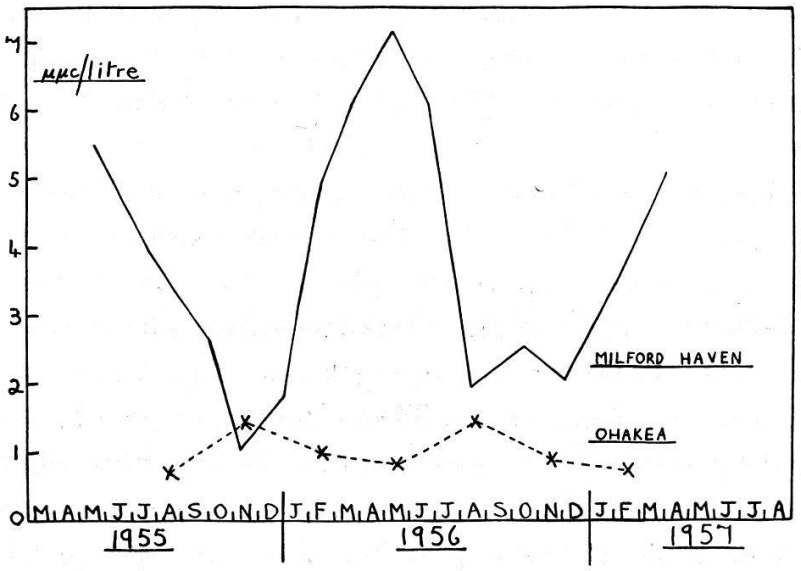

Fig. 8. Seasonal variation of Sr^{90} content of rainwater at Milford Haven and Ohakea.

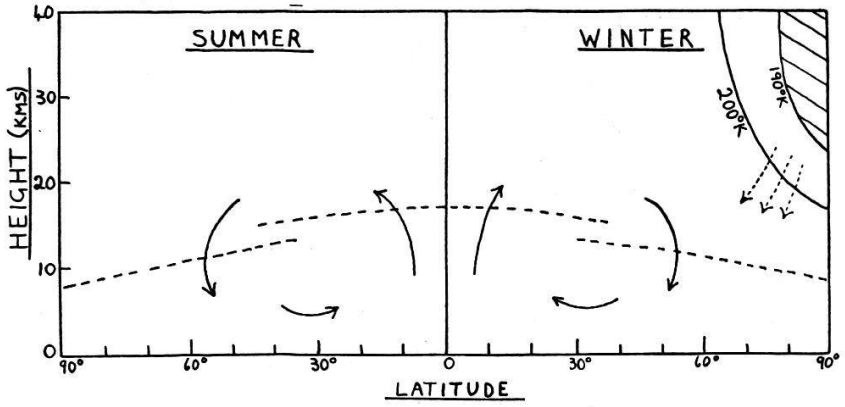

Fig. 9. Atmospheric circulation model (after *Dobson* and *Brewer*).

two hemispheres. There is a strong indication, in the northern hemisphere at least, that this variation is in step with a similar variation in the lower stratosphere.

c) The greatest deposition takes place in the middle latitudes.

d) Deposition in the northern hemisphere is greater than that in the southern hemisphere.

The seasonal variation of the Sr^{90} is remarkably similar to the seasonal variation of the total ozone in the atmosphere which has been observed by many workers. Based on measurements of the distribution of ozone and water vapour in the atmosphere, *Dobson* (3) has proposed a model for the general circulation which offers a satisfactory explanation of the observed Sr^{90} data. In *Dobson*'s model, a very cold pool of air forms above the winter pole during the late winter months when the air lies in shadow (Fig. 9). The ultimate sinking of this pool, carrying ozone-rich air to lower levels in the stratosphere, is believed to be the cause of the rapid increase in total ozone in early spring in high latitudes. Since the Sr^{90} concentration in the stratosphere increases rapidly with height, this subsidence would also bring Sr^{90}-rich air into the lower stratosphere in early spring, leading to a seasonal variation of the concentration in this region of the atmosphere.

The interchange of air between the stratosphere and the troposphere has been discussed by *Brewer* (4) who, in order to explain the form of the water-vapour curve in the stratosphere, has suggested a circulation system in which tropospheric air enters the stratosphere at the equator, travels in the stratosphere to temperate and high latitudes and then sinks again into the troposphere. This circulation provides the means for bringing stratospheric Sr^{90} down into the troposphere where the concentration would be expected to show the seasonal features discussed above. The form of the global deposition curve (fig. 7) supports the view that the stratospheric air enters the troposphere in the middle latitudes,

bringing with it Sr^{90} which is progressively washed out of the troposphere by rainwater as it travels north and south from the region of entry.

Finally, the subsidence of the belt of cold air in the stratosphere above the winter pole would be expected, from continuity considerations, to initiate a meridional circulation in the stratosphere in which there would be a more or less continuous flow from the summer to the winter hemisphere (5). This flow might provide the explanation for the presence and deposition in the southern hemisphere of Sr^{90} from clouds which were generated in the northern hemisphere.

6. Conclusions

The programme of measurements described has revealed some interesting and important features of the exchange processes between the stratosphere and the troposphere which give rise to a non-uniform deposition pattern of long-lived fission products over the surface of the earth. The relative smoothness of this pattern suggests that with a sufficient number of sampling points it might be possible to obtain reasonable values for the integral of Sr^{90} deposition over the surface of the earth. A summary of the main results is given in the abstract at the front of the report.

Summary

The results are given of several investigations which have been made in the United Kingdom into the long-range diffusion and deposition from nuclear test explosions. In recent years these investigations have been extended to cover the deposition of Sr^{89}, Sr^{90} and Cs^{137} in various parts of the world and an account is given of the results obtained so far.

It is shown that most of the Sr^{90} deposited is derived from those nuclear explosions whose clouds enter the stratosphere and return to earth slowly over a number of years. In the successive yearly periods between May 1954 and May 1957 the Sr^{90} deposition at a representative station in the U.K. amounted to 2.06, 2.24 and 2.55 mc/km^2 respectively and the cumulative total in October, 1957 was 8.6 mc/km^2. Cs^{137} levels are about 70 per cent higher than those of Sr^{90}. The experimental data indicate that within a given region, fallout is proportional to rainfall.

It has been found that the mean Sr^{90} concentration in rainwater in the U.K. shows a marked seasonal variation with peaks in the late spring and troughs in the late autumn of 1955, 1956 and 1957 and the concentration in the lower stratosphere appears to vary in a similar manner. The maximum to minimum ratio is about 6:1. A similar but less marked

variation of the opposite phase has been observed in New Zealand rainwater. It has also been observed that the deposition rate of Sr^{90} has a minimum value near the equator, and there appears to be a pronounced maximum in the middle latitudes of the northern hemisphere. These results are shown to be consistent with a model for the general circulation of the atmosphere proposed by *Dobson* and *Brewer* as a result of their observations of ozone and water vapour in the atmosphere.

Zusammenfassung

In 6 Stationen Großbritanniens und in 13 Stationen an anderen Orten der Erde wird in monatlichen oder dreimonatlichen Abständen Regenwasser gesammelt und auf den Gehalt an Sr^{89}, Sr^{90} und Cs^{137} geprüft. Diese Stationen sind seit Mai 1954 eine nach der andern in Betrieb gesetzt worden. Im folgenden ist eine Zusammenstellung der bisher erhaltenen Ergebnisse zu finden.

Es wird gezeigt, daß die größte Menge von Sr^{90} von jenen Kernexplosionen stammt, deren Wolken in die Stratosphäre dringen und nachher während einer Reihe von Jahren wieder langsam zur Erde zurücksinken. In den aufeinanderfolgenden jährlichen Zeiträumen zwischen Mai 1954 und Mai 1957 stieg die Sr^{90}-Ablagerung in einer typischen Station im Vereinigten Königreich um je 2,06, 2,24 und 2,55 mc/km^2 pro Jahr und betrug Ende Oktober 1957 gesamthaft 8,6 mc/km^2. Der Cs^{137}-Gehalt liegt um etwa 70% höher als jener des Strontium-90. Die Untersuchungsergebnisse zeigen auf, daß die radioaktiven Niederschläge in einem bestimmten Gebiet dem Regenfall mengenmäßig proportional sind.

Es zeigte sich, daß die mittlere Sr^{90}-Konzentration im Regenwasser im Vereinigten Königreich eine ausgesprochen jahreszeitlich gebundene Schwankung aufweist mit einem Maximum im späteren Frühjahr und einem Minimum im späteren Herbst 1955, 1956 und 1957; auch die Konzentration in den unteren Teilen der Stratosphäre scheint in ähnlicher Weise zu variieren. Das Verhältnis von Maximum zu Minimum ist ungefähr 6:1. Eine ähnliche, aber weniger ausgesprochene Schwankung auf der entgegengesetzten Hemisphäre ist im neuseeländischen Regenwasser beobachtet worden. Es wurde ebenfalls festgestellt, daß die Ablagerungsrate von Sr^{90} in der Nähe des Äquators einen Minimalwert aufweist, ein ausgesprochener Maximalwert scheint in den mittleren Breiten der nördlichen Hemisphäre aufzutreten. Diese Ergebnisse entsprechen einem Modell für die allgemeine atmosphärische Strömung, wie es von *Dobson* und *Brewer* als Resultat von Beobachtungen über die Bewegung von Ozon und Wasserdämpfen in der Atmosphäre aufgestellt wurde.

Résumé

Un réseau de six stations en Grande Bretagne et de treize stations dans d'autres parties du monde s'occupe de rassembler les eaux de pluie et d'étudier, tous les mois ou tous les trois mois, leur teneur en Sr^{89}, Sr^{90} et Cs^{137}. Ces stations ont été mises en service l'une après l'autre depuis mai 1954; et on fait une récapitulation des résultats obtenus jusqu'ici.

Il est démontré que la plupart du Sr^{90} retombé est dû aux explosions nucléaires, dont les nuages ont pénétré dans la stratosphère et redescendent lentement sur terre dans le courant de plusieurs années. Au cours des années allant de mai 1954 à mai 1957, on a trouvé une sédimentation de Sr^{90} sur une station représentative de la Grande Bretagne correspondant à 2,06, 2,24 et 2,55 mc/km² par année, avec un total en octobre 1957 de 8,6 mc/km². Les valeurs pour le Cs^{137} sont d'environ 70% plus élevées que celles du Sr^{90}. Les observations faites montrent que pour une région donnée les retombées radioactives sont proportionnelles aux chutes de pluie.

On a trouvé que la concentration moyenne de Sr^{90} dans les eaux de pluie tombant en Grande Bretagne montre une variation saisonnière avec maximum à la fin du printemps et minimum à la fin de l'automne, en 1955, 1956 et 1957; les concentrations dans les couches stratosphériques inférieures semblent montrer les mêmes variations. Les variations du maximum au minimum sont dans le rapport de 6:1. Des variations semblables mais moins accentuées ont été observées dans les eaux de pluie de la Nouvelle Zélande. On a également trouvé que la sédimentation de Sr^{90} a une valeur minimum près de l'équateur, et que le maximum semble être dans les latitudes moyennes de l'hémisphère nord. Ces résultats semblent confirmer les vues sur la circulation générale de l'atmosphère de *Dobson* et *Brewer*, qui ont étudié le comportement de l'ozone et des vapeurs atmosphériques.

Acknowledgements

The data presented in this paper have been drawn mainly from two AERE reports (6, 7).

1. *Eisenbud, M.*, and *Harley, J. H.:* Science **117**, 141 (1953).
2. *Bryant, F. J., Chamberlain, A. C., Morgan, A.*, and *Spicer, G. S.:* AERE HP/R 2353.
3. *Dobson, G. M. B.:* Proc. roy. Soc. Med. A **236**, 187 (1956).
4. *Brewer, A. W.:* Quart. J. roy. med. Soc. **75**, 351 (1949).
5. *Kellogg, W. W.:* J. Meteorol. **9**, 446 (1952).
6. *Stewart, N. G., Crooks, R. N.*, and *Fisher, E. M. R.:* AERE HP/R 2017.
7. *Stewart, N. G., Osmond, R. G. D., Crooks, R. N.*, and *Fisher, E. M. R.:* AERE HP/R 2354.