SHORTER CONTRIBUTION

Radioactive fallout in rain at Melbourne, 1958 through 1970

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(Manuscript received October 4, 1971; revised version January 20, 1972)

ABSTRACT

Gross beta activity measurements are analysed from the point of view of the annual cycle in the lower troposphere. The annual cycles found in rainfall concentration of radioactive fallout and in the total fallout deposition peak at different times, the former in December and the latter in August. The difference is explained in terms of the variation in rainfall. A phase difference is apparent between these cycles and *in situ* stratospheric data, suggesting that ground level air lags behind the stratosphere by about one or two months.

1. Introduction

In previous reports (Dyer & Yeo, 1960; Dyer & Hicks, 1965) specific features of the general circulation of the atmosphere as evidenced by measurements of radioactive fallout made at Aspendale, near Melbourne, Australia, were studied. Among the aspects investigated were the stratospheric global circulation time (about 3 weeks at our latitudes), the stratospheric half-residence time for equatorial inputs (about 2 years), and the short-term behaviour following particular atomic weapons explosions. However, recent technological advances have provided meteorologically-oriented researchers with better tools for studying the atmosphere, the GHOST balloon program being an obvious example.

The present purpose is to summarize a longer period of Aspendale data than has been available before. We will look, in particular, at the implications of and inferences to be drawn about the annual cycle as observed at ground level, and about the stratospheric residence time.

2. Method of measurement

Since early 1958, total deposited radioactivity has been collected at monthly or more frequent intervals by exposing plastic-coated funnels attached to collecting bottles. After collection, the exposed funnel surfaces are scrubbed and washed in distilled water which is added to the previously measured rainfall aliquot. Radioactivity is removed by two passes through a selected ion-exchange resin, which is subsequently dried and ashed in preparation for low-level beta counting using a conventional thin-window Geiger-Müller counter.

Samples are counted to better than 90 % certainty.

Calibration is against natural potassium, which will admittedly introduce a small consistent error due to the difference in energies between this and average fallout radionuclides. No attempt has been made to correct for this, and any errors which result apply throughout the entire period of measurement.

Total deposition is calculated from the known collecting area. In the calculation of the radioactivity concentration of the rainfall (often referred to as the "specific activity") dry deposition is neglected. In fact, gravitational settling sedimentation of particles often accounts for as much as 10 % of the total deposition.

Measurements of the volume of rainfall collected agree well with precipitation rates as

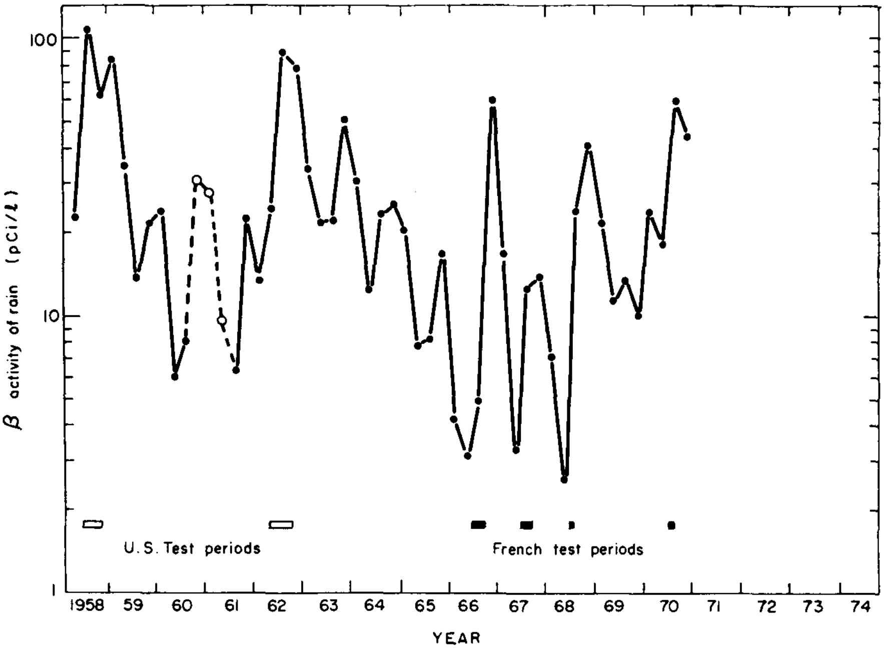

Fig. 1. Fallout in rainfall at Aspendale (38° S). Quarterly averages.

recorded by standard rain gauges at the same location.

3. Quarterly means of fallout

In Fig. 1, quarterly means (calculated after due weighting according to rainfall) are plotted. A large amount of variation is obvious, largely associated with the various injections as indicated. An annual cycle is also clear, particularly between 1962 and 1966.

Some of the variability in Fig. 1 must result from changes in type and amount of rainfall (r). An analysis of data collected over much shorter periods shows a linear dependence of total deposition (D) on about the half power of rainfall, so that the concentration in rain (C) is roughly proportional to the inverse half power. A similar behaviour in longer-term data must be expected, even though seasonal variations do not allow this to be verified.

A feature of the data plotted is the general decrease in levels following the equatorial stratospheric inputs of 1958 and 1962. This results from both radioactive decay and depletion of the stratospheric reservoir. In this regard, it must be remembered that tropospheric residence times are much shorter than the time scales being considered here.

Since 1966, a continuing program of weapons testing in the south-eastern Pacific is reflected in the data. The large peak of late 1966 undoubtedly results from the injection, largely tropospheric, of fallout from the preceding French test series (Dyer & Hicks, 1967). The peaks of 1968 and 1970 can be accounted for in the same way.

4. Annual means

More information can be gained from consideration of annual averages, shown in Fig. 2. Here, the annual cycle is necessarily damped, and the gradual depletion of the stratospheric reservoir is quite apparent, as shown by the straight lines drawn. Sudden increases corresponding to the indicated periods of weapons testing are obvious. Chinese and Russian explosions are not indicated since these are conducted at higher northern latitudes and there is no evidence of fallout from them being detected at Aspendale. Also, no sub-surface tests are included.

The consistent decrease in concentration

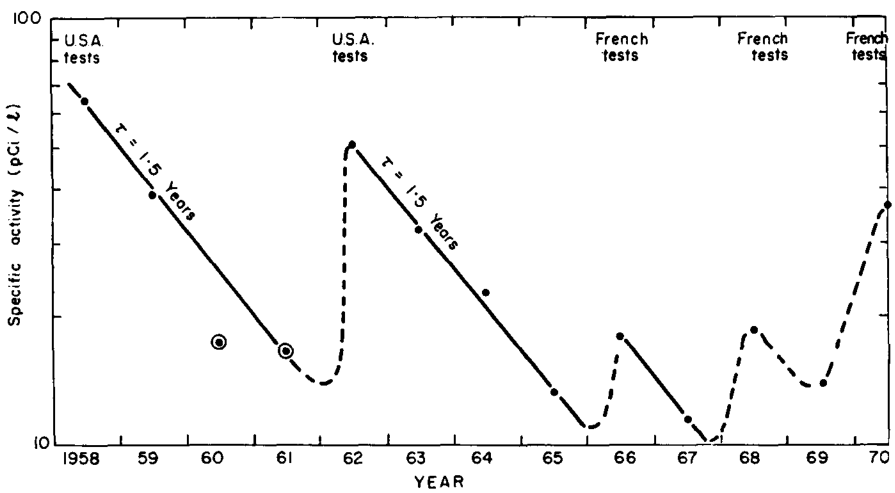

Fig. 2. Fallout in rainfall at Aspendale (38° S). Annual averages.

following the large inputs of 1958 and 1962 is mainly a result of the stratospheric residence time, since the radioactive half-life of the debris likely to be remaining some years after injection is of the order 5 to 10 years (as suggested by Fig. 1 of Hunter & Ballou, 1951). If the radioactive half-life is, in fact, 6 years, then the slope of 1.5 years apparent in Fig. 2 implies a stratospheric half residence time of 2 years. However, if the radioactive half-life is assumed to be less than 6 years, correspondingly higher estimates of the stratospheric half residence time result. A value of about 2.4 years is obtained when the radioactive halflife is 4 years. Values of about 2 years are consisent with other estimates of the residence time obtained for the lower tropical stratosphere.

5. The annual cycle

For study of the annual variability, periods of negligible input are desirable. On the basis of Fig. 2, the years 1959, 1963, 1964, 1965 and 1967 are chosen for this purpose. Two other possibly acceptable years, 1960 and 1961, are not available for selection because some data were lost. The values plotted in the diagrams for these two years are partially inferred from another source (Blake et al., 1962).

Mean annual cycles of specific activity and of total deposition (the gross fallout collected, expressed in mCi km^{-2}) are calculated by normalising quarterly data obtained in each year to the annual average for that year. This removes the year-by-year decrease in levels. Then the normalised values are averaged to give the mean levels appropriate for each quarter.

In this process, we remove the change in average value from year to year, but the diminution through each year remains. Consequently,

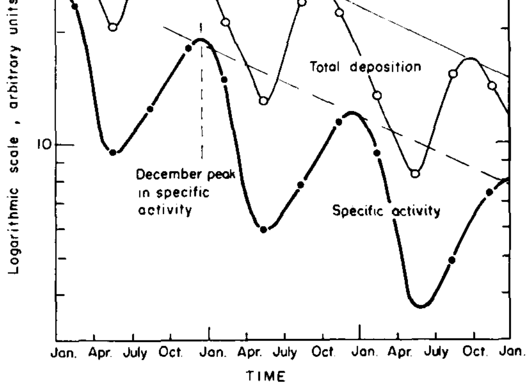

Fig. 3. Mean cycles of specific activity and total deposition at Aspendale.

in Fig. 3 the average cycles obtained are plotted over three years, with the 1.5 year diminution half-life reintroduced: in the first year the basic cycle is plotted, in the second each quarterly value is reduced by a factor of 1.59, etc.

The specific activity cycle shown in Fig. 3 peaks in December, minimizes during May, and has an amplitude of 2.6 (Maximum/Minimum). The total deposition cycle is of about the same amplitude (2.4), but maxima and minima occur in August and May respectively. An obvious question remains to be answered: When does the annual cycle of atmospheric radioactivity near the surface maximise?

The difference between the two cycles plotted in the diagram must be associated with the removal mechanism, mainly rainfall, in the manner mentioned earlier. Since the total depositon tends to be positively correlated with precipitation, and specific activity negatively, we must conclude that near-surface concentrations of fallout radioactivity at 38° S probably maximise in about October. That the air concentrations peak between the maxima in D and in C follows formally from consideration of the correlations with rainfall.

6. Discussion

The stratospheric half-residence time estimated here (about 2 years) is directly comparable to the many other estimates available in the literature. Most experiments using radioactive tracers seem to imply a value of about 2 years over the equator, decreasing to 6 months or less at the poles.

The annual cycle found is similar in many respects to that observed in other tracers in the stratosphere or of stratospheric origin. Total ozone measurements made at 38° S show a cycle peaking in September and minimizing in March–April (as is seen in the graphs presented by Kulkarni, 1966), similar to the present result but with about one to two months phase difference. In this context, it is relevant to recognise that the cycle obtained here represents the net effect of stratospheric transport and stratosphere-troposphere inter-change, in both of which annual cycles probably exist. Thus the one to two months phase lag between stratospheric and lower tropospheric phenomena which is implied here seems reasonable, and is directly comparable with the analogous estimate of Junge (1962) based on surface and stratospheric ozone measurements.

Few suitable sets of data are available with which to compare the amplitude of the present cycle. This is largely because interpretation of northern hemispheric data is complicated by the more frequent atomic weapons tests, and because of the scarcity of observing stations in the southern hemisphere.

7. Conclusions

From the present data we may deduce three conclusions: (1) The relevant stratospheric half-residence time following equatorial (1958 and 1962) and low mid-latitude (1966) injections appears to be about 2 years. (2) The annual cycle resulting near the surface at 38° S is skewed, with minimum in May and maximum in about October. (3) The amplitude of the cycle appears to be about 2.5, expressed as the ratio maximum/minimum.

Acknowledgements

The monitoring program upon which the present report is based was initiated by Dr A. J. Dyer. Mr G. Grauze has performed the analyses.

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РАДИОАКТИВНЫЕ ВЫПАДЕНИЯ В ДОЖДЯХ В МЕЛЬБУРНЕ

С точки зрения годового цикла в нижней тропосфере анализируется большое количество данных измерений бэта-активности. Годовые циклы, найденные в концентрациях радиоактивности в дождях и в полном количестве ее отложений имеют максимумы в различное время, первый — в декабре и последний — в августе. Разница объясняется вариациями в количестве осадков. Заметна разность фаз между этими циклами и данными стратосферных измерений, показывающая, что приземный воздух отстает в этом отношении от стратосферы на один или два месяца.