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Seasonal and latitudinal variations of atmospheric radioactivity along Australia's east coast (150°E longitude)

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ABSTRACT

Preliminary results obtained from a network of air sampling stations located between 9° S and 43° S along about 150° E longitude indicate that the "spring" maximum in tropospheric radioactivity concentrations propagates poleward at a rate of about 35 cm s^{-1}. The peak appears to be associated with an injection of air enriched with Be-7, presumably stratospheric, at about 20°S during September–October. A secondary injection of air containing high concentrations of Be-7 is suggested by the results, occurring southward of 35° S during November–December.

Use of artificial radionuclide concentrations as southern hemispheric tracers is severely hampered by continuing weapons testing in the south Pacific region. A complete comparison between the various models of the atmospheric general circulation and the results from studies such as the present awaits the collection of data unaffected by weapons testing programs.

1. Introduction

In an attempt to obtain more information relevant to the continuing debate regarding stratospheric transport mechanisms and stratosphere–troposphere exchange of air, an air sampling program to measure various radionuclides was commenced at Aspendale, Victoria, in 1966. Prime attention was given to the concentrations of the naturally occurring cosmic ray spallation product Be-7, a gamma-emitter primarily produced in the polar stratospheres, having a half-life (about 53 days) convenient for indicating the presence of high-latitude stratospheric air. The approach was considered to be of greater potential power if Be-7 concentrations were compared with those of some suitable fission product, such as Sr-90 or Cs-137 (half-lives of the order 30 years), which then existed in a predominantly equatorial stratospheric reservoir resulting from nuclear weapons tests in previous years. For this purpose, it was originally anticipated that several years of data would be required to determine differences between the unaffected annual cycles of natural and man-made radioactivity concentrations. In this regard, it is unfortunate that during the subsequent period only one year (1969) has been free of southern hemispheric nuclear weapons testing.

In anticipation of the possible conclusion of the French nuclear weapons test series in the South Pacific region (about 20° S, 140° W) and to obtain information on the latitudinal differences in the annual cycles of the two types of atmospheric radioactivity (one of relatively short half-life predominantly from the polar stratosphere and the other of much greater half-life primarily from equatorial stratosphere), a network of sampling stations was set up along the eastern Australian coast during 1971. This was extended to the north in the following year by the installation of equipment in Papua New Guinea.

Since the original aim of the experiment has not yet been met, this report must be considered to be preliminary. The main purpose here is to draw attention to some aspects of the data, particularly those that bear upon the critical questions of seasonal effects within the stratosphere and between it and the troposphere.

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2. Sampling sites and experimental procedures

During 1966 an air sampler was installed on the grounds of C.S.I.R.O., Division of Atmospheric Physics at Aspendale, a residential area about 30 km south of Melbourne, Victoria (38° S, 145° E). Subsequent installations were made at Sandy Bay, 4 km south of Hobart, Tasmania (43° S, 147°E); at Epping, 16 km northwest of Sydney, N.S.W. (34°S, 151°E); at St. Lucia, 4 km south of Brisbane, Queensland (27°S, 153°E); at Upper Ross, 10 km southwest of Townsville, Queensland (19°S, 147°E) and at Waigani, 11 km northeast of Port Moresby, P.N.G. (9.5°S, 147°E). Sampling sites were selected to be free of interference from nearby obstructions. At the sites near Hobart and Brisbane, the sampling equipment was erected on the roofs of buildings, but in all other cases grassed experimental areas were selected and the sampling intakes were about 2 m above the ground surface. None of the sites is subject to pollution from nearby industrial sources, this being a criterion for site acceptance in order to allow a subsidiary study of atmospheric particulate concentrations at each location (Goodman & Hicks, 1973).

The samplers draw air through high quality polyester filter material[1] which effectively retains all particles larger than about 0.2 μm diameter. Filters are exposed for one week. Checks made with various back-up filters and electrostatic precipitation have shown that more than 99% of the particulate Be-7 is collected. Two types of high volume samplers have been used. The earlier type, used at Aspendale prior to 1971, allowed a sampling rate of about 150 $m^3\ h^{-1}$, while the later version has a reduced flow rate of about 50 $m^3\ h^{-1}$. The more recent sampler has been described by Haye (1973).

Exposed filters are analysed at the Aspendale laboratory using a 100-channel gamma spectrometer coupled to a 10 cm × 10 cm NaI(Tl) crystal. The resulting gamma energy spectra are computer analysed for several radionuclides, including Be-7 (0.48 MeV), Cs-137 (0.66 MeV), Nb-Zr-95 (0.76 MeV) and Ce-144 (0.13 MeV). At routine intervals the system is calibrated against standards of known intensity supplied from commercial sources.

Samples are normally counted for 100 minutes, typically allowing better than 90% counting accuracy. To improve the resolution and certainty of the method, samples are recounted at least four times during the 3-month period after collection, thus allowing determination of the radioactive half-life corresponding to each gamma energy peak of specific interest. This provides a means of detecting the presence of the short-lived fission product Ru-103 (39 days), whose gamma emission interferes with that from Be-7. On some occasions, sufficient Ru-103 is detected to make quantitative determination of Be-7 impossible, and hence some periods of missing data occur (usually shortly after periods of high yield weapons testing).

A further source of experimental inaccuracy is the determination of the volume of air sampled. Usually, measurements of the sampling rate are made each day, but even then the error in this particular evaluation is estimated to be about 5%.

The computer analysis procedure accepts spectral information obtained at various times after collection. Two estimates of each radionuclide concentration are provided, corresponding firstly to the assumption that there is no interference by recently produced atomic debris and secondly to the assumption that the sample is contaminated by the presence of some unidentified material. A statistically significant difference between the results is accepted as an indication of contamination, and the data are then studied further using a graphical technique to enable more accurate determination of the radionuclide concentrations. In practice, the procedure has been employed most frequently in the case of Be-7 evaluation, since Ru-103 contamination is relatively common. We believe that the analysis has rejected satisfactorily those occasions when Be-7 determinations are unreliable.

To a substantial extent, the above procedure is necessitated by the 8% resolution of the gamma scintillation detector employed. Use of a more modern germanium detector would alleviate the problem, but at considerable cost.

3. Long-term Aspendale results

Fig. 1 shows the seasonal variations of Cs-137 and Be-7 in ground-level air at Aspendale between 1966 and 1973. Three-point running quarterly means (calculated geometrically from monthly values obtained from basic weekly measurements)

[1] Microsorban special grade S 99/98, manufactured by Delbag-Luftfilter A.G., Berlin, West Germany.

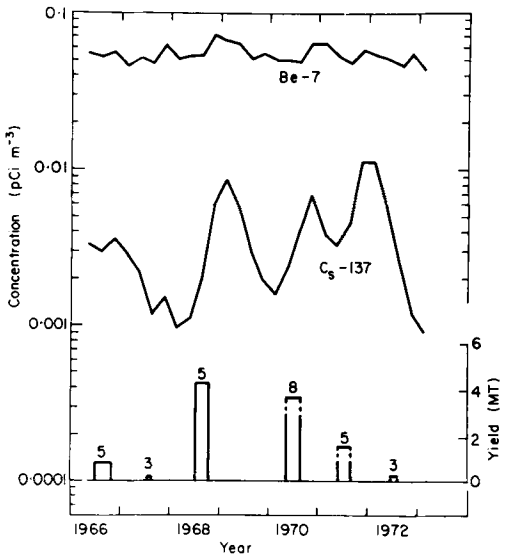

Fig. 1. Seasonal running average concentrations of Be-7 and Cs-137 in ground-level air at Aspendale, 38° S, from 1966 to 1973. Approximate details of total yields and periods of the weapons testing series conducted in the South Pacific Ocean are shown, with numbers indicating the approximate number of explosions in each testing period.

are plotted to give a smooth representation of the seasonal effects. Also shown are the periods of southern hemispheric nuclear weapons testing. Total yields of each test series and the number of explosions in each are obtained from official announcements or from press reports when the former are lacking.

It is immediately obvious in Fig. 1 that large peaks in Cs-137 concentration follow the major test series, lagging by about 5 months on the average. The strong peaks observed following the 1968, 1970 and 1971 inputs are similar to those observed in rainfall concentrations of beta-emitters following equatorial tests of nuclear weapons (see Dyer & Hicks, 1965, for example), in which case the time lag has been interpreted as a measure of the rate of stratospheric transport. However the present data show no indication of a stratospheric input, since the decay rate following each peak of the Cs-137 concentration curve indicates a half-residence time of about 3 months, in comparison with the value 1–2 years normally quoted for the equatorial stratosphere. It seems more likely that in the present case of injection at 23° S the time lag between the input and the observation of a concentration maximum at 38° S is a result of tropospheric latitudinal and vertical mixing. If the time delay is associated with latitudinal mixing alone, then latitudinal mixing rates of the order 5×10^8 $cm^2\ s^{-1}$ are suggested, whereas if the lag is solely a result of vertical mixing from an assumed injection altitude of 15 km, then the observations indicate a vertical diffusivity of about 4×10^4 $cm^2\ s^{-1}$. Both results appear reasonable, and it is not possible to judge which, if either, mechanism predominates. Later consideration of the results from other sites will bear on this question.

Similar peaks in Be-7 concentration follow the periods of nuclear testing (although these are not obvious in Fig. 1 since they are of lower proportional amplitude). Production of Be-7 has been associated with nuclear explosions (Bleichrodt & van Abkoude, 1963), but bearing in mind the decay rate of Be-7 and the fact that detection of the maximum concentration was not until about three half-lives after the supposed injection, it seems unlikely that this is the sole cause of the maxima observed. Nor is it probable that Ru-103 contamination explains the observations, since considerable effort was made to avoid this particular source of error.

Careful inspection of Fig. 1 shows that Be-7 concentration maxima were also observed in those years when no large weapons testing program took place. Fig. 2 shows the average annual cycle for the years 1966, 1967, 1969 and 1972. The summer peak that is apparent is a common feature of studies of this type, and was also evident in the rainfall concentrations of beta-emitters referred to above. The spring peak of August, September and October appears to be real, although of questionable statistical significance since only 4 years of data are involved. However, each individual year exhibits this feature, with high Be-7 concentrations being observed during October in 1966, September in 1967, August in 1969 and September in 1972.

The rapidly changing Cs-137 data do not warrant a similar analysis. However, between mid-1966 and mid-1968 the concentrations of Cs-137 decreased relatively uniformly, at a rate corresponding to a residence time of about 7 months. As is seen in Fig. 3, Cs-137/Be-7 isotope ratios also dropped uniformly, save for a relatively severe decrease in the ratio during the spring of 1967. It is at precisely this time of year that Fig 2 indicates a rather strong increase in the concentration of Be-7.

Thus the data indicate that air of either polar or stratospheric origin contributed to the spring increase in Be-7 concentrations during 1967 (since in both of these regions of the atmosphere Cs-137/Be-7 ratios will have been low). Consideration of isotope ratios, as in Fig. 3, allows some certainty to be associated with conclusions of this general type, since the effects of local tropospheric cleansing processes (such as rainout and dry deposition) are essentially eliminated.

It is considerations such as those outlined above that have been the major interest in the expanded program involving the collection of air samples along the eastern Australian coast.

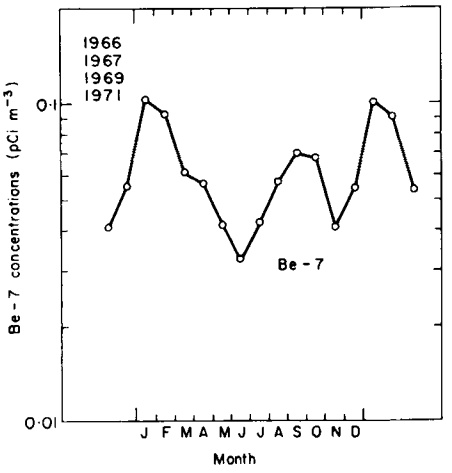

Fig. 2. The average annual cycle of Be-7 concentration at Aspendale.

4. Preliminary network results

Considerably more information can be obtained from a comparison of radioactivity concentrations at different latitudes. Fig 4(a) is a smoothed isopleth representation of the Be-7 results between commencement of the network air sampling program in August 1971, and April 1973. It is evident that the "summer" maximum in Be-7 concentrations observed in 1971 propagated from the equator at a rate of about 35 cm s^{-1}, corresponding to a latitudinal diffusivity of about 7.5×10^8 cm^2 s^{-1}. A similar poleward propagation of the "summer" maximum is seen during 1972, although less clearly defined. It is not intended to suggest that the mechanism is a mean motion of the air, since there is no way that consideration of a single tracer can differentiate between this and alternative explanations such as a stratospheric/tropospheric interchange process that migrates towards the pole.

Fig. 4(b) shows similar features in the Cs-137 data, particularly during 1971, with a somewhat slower rate of poleward progression corresponding to a meridional diffusion coefficient of about 4×10^8 cm^2 s^{-1} (much the same as the estimate derived earlier on the basis of the Aspendale data alone).

In Fig 4(a) it is evident that the 1971 maximum concentration of Be-7 at Townsville (19° S) occurred during November and December. But Fig. 4(b) shows that the peak in Cs-137 at the same location occurred in September. Further comparison of the two diagrams shows that the poleward

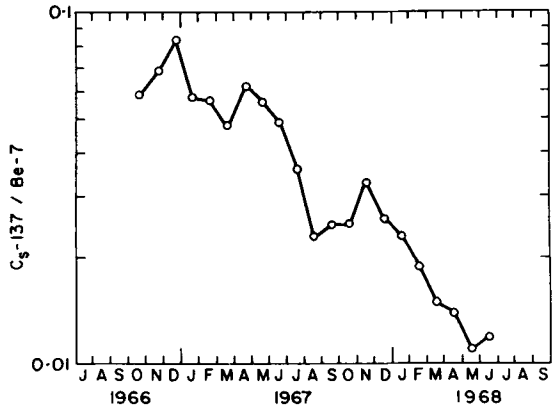

Fig. 3. The change with time of the isotope ratio Cs-137/Be-7 at Aspendale, through 1967, following weapons testing during 1966.

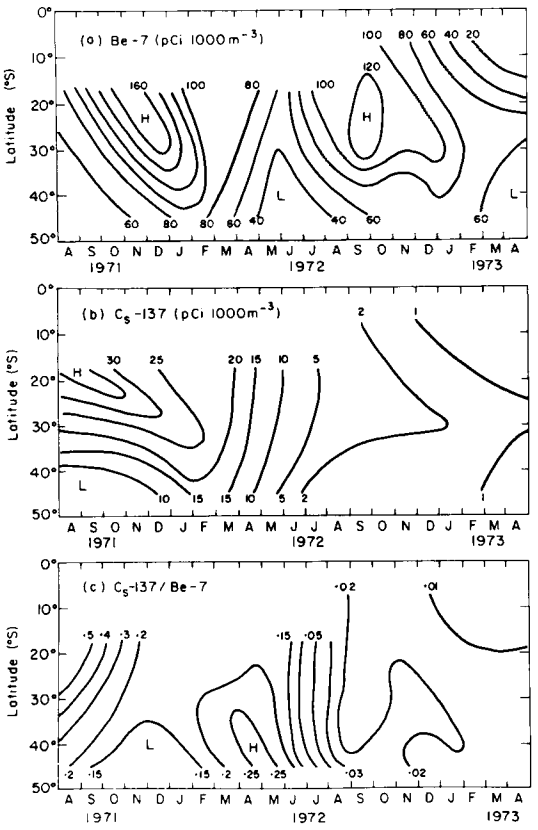

Fig. 4. Latitude-time isopleth diagrams of (a) Be-7 concentration, (b) Cs-137 concentration and (c) the isopleth ratio Cs-137/Be-7, obtained from the network sampling program. Concentrations are in pCi per 1000 m^3.

propagation rates of the maxima in both isotope concentrations were about the same after the Cs-137 peak had arrived at about 30° S. An obvious explanation lies in the difference in sources, with Cs-137 from the 1971 weapons tests slowly dispersing into the southern mid-latitudes and an injection of Be-7 enriched air (presumably from the stratosphere) taking place north of 30° S shortly after September 1971. This overall picture is supported by the 1972 data, with an obvious injection of Be-7 into the lower troposphere occurring during September at about 25° S. There is little evidence for a corresponding increase in Cs-137 concentrations during 1972, as must be expected if in this year the major injection resulted from a stratospheric input (there was little production of Cs-137 during 1972, see Fig. 1).

As mentioned earlier, consideration of individual atmospheric tracers does not allow the drawing of definite conclusions regarding the transport mechanisms involved. In the present, tropospheric, case, it is clear that such factors as rainfall will influence the concentrations of both Be-7 and Cs-137. Thus it is of considerable interest to consider the ratios of concentrations of the two isotopes, by which, hopefully, particular parcels of air can be identified. However, the isotope ratio information presented in Fig. 4(c) is somewhat confused by the injection of Cs-137. The low isotope ratios detected at the most southern stations during November and December 1971 suggest that the southward migration of the radioactivity concentration peak was supported by a further influx of Be-7, again presumably from above. The high ratios found during March, April and May, 1972, at the same southern stations suggest that the rate of input of Be-7 was reduced more rapidly than that of Cs-137, again compatible with the hypothesis of a

stratospheric source of the former late in 1971 while the latter was contained largely within the troposphere. Although by the end of 1972 most of the tropospheric Cs-137 should have been removed, there is some evidence of a similar high latitude minimum in the Cs/Be isotope ratio.

Comparative studies of the concentrations of Be-7 and Cs-137 in surface air have also been reported by Rangarajan & Gopalakrishnan (1970), who investigated the behaviour in tropical and subtropical latitudes. Their finding that in such low latitudes only about 50% of the ground level Be-7 is derived from the stratosphere has relevance to the present work. It is obvious, however, that the implied high levels of the tropospheric "background" concentrations are of little importance in the present analysis. A more critical consideration may well be the role played by the seasonally varying height of the tropopause, particularly since the vertical profiles of concentrations of different radionuclides immediately above the tropopause might be expected to be substantially different. Nevertheless, it is of interest to compare the apparent poleward propagation of the spring–summer peak in Be-7 concentration (Fig. 4(a)) with the meridional transport of volcanic debris in the lower stratosphere as indicated by solar radiation data (Dyer & Hicks, 1968). Both studies suggest a transport mechanism commencing near the equator in midwinter and reaching 40° S in about the following January. While it is tempting to associate both effects with a common stratospheric cause, relating the present ground-level results with high-altitude motions is conjectural at this stage.

5. Conclusions

Although influenced by atomic debris from nuclear weapons tests conducted in the South Pacific Ocean, concentrations of Be-7 and of Cs-137 measured at 38° S between 1966 and 1973 allow some specific aspects of the general circulation of the atmosphere to be identified. In common with other similar tracer results, ground level concentrations of Be-7 increase after winter and reach a maximum in about mid-summer. There is strong evidence of a spring peak preceding the summer maximum, and this peak was associated with a decrease in the Cs-137/Be-7 isotope ratio during 1967 (the only year with data suitable for this type of analysis).

Data obtained at other latitudes indicate that the summer peak in Be-7 concentrations during 1971 migrated towards the pole at a rate of about 35 cm s^{-1}, corresponding to a mixing rate (in a Fickian diffusion model) of about 7.5×10^8 cm^2 s^{-1}. A low-latitude injection of Be-7 (presumably from the stratosphere) during the spring of 1971, and again in the following year, is suggested by the averaged isopleth diagrams presented here, while there is some evidence for a similar incursion at higher latitudes in the isotope ratio data. The raw data show evidence of stratospheric air entering the troposphere as early as July, in low latitudes.

At the present stage of this experiment, statistical significance is lacking, particularly in the case of the latitudinal comparisons. It is to be hoped that future years will be free of southern hemispheric nuclear testing, so allowing a closer study of the various aspects mentioned in this preliminary report. Eventually, it is hoped to provide a sufficiently reliable set of data to allow comparison with the predictions of models of the global general circulation of the atmosphere, and perhaps to supply some input to them.

6. Acknowledgements

Mr G. A. Grauze made most of the radioactivity measurements prior to 1968. The cooperation of many people has been essential to the smooth operation of the sampling network. In particular, mention should be made of the assistance rendered by Dr K. B. Fenton and Mr G. West of the University of Tasmania, Dr E. K. Bigg and Mrs J. Williams of C.S.I.R.O. Division of Cloud Physics, Dr D. F. Robertson and Mr G. Perrin of the University of Queensland, Prof. J. F. Ward and Mr D. Galloway of the James Cook University of North Queensland, and Dr T. G. Emeleus and Mr P. G. Dunks of the University of Papua and New Guinea. Mr B. Turner constructed the network air samplers.

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ШИРОТНЫЕ И СЕЗОННЫЕ ВАРИАЦИИ АТМОСФЕРНОЙ РАДИОАКТИВНОСТИ ВДОЛЬ ВОСТОЧНОГО БЕРЕГА АВСТРАЛИИ (150°В.Д.)

Предварительные результаты, полученные на стеи станций проб воздуха, расположенных между 9° и 43° ю.ш. вдоль, приблизительно, 150°в.д., показывают, что "весенний" максимум в концентрации тропосферной радиоактивности распространяется к полюсу со скоростью около 35 см/с. Представляется, что этот пик связан с инжекцией воздуха, обогащенного Be-7, предположительно, стратосферного происхождения на, примерно, 20°ю.ш. в течение сентября-октября. Вторичная инжекция воздуха, содержащего высокие концентрации Be-7, происходит к югу от 35°ю.м. в течение ноября-декабря. Исподьзование концентраций искусственных радионуклидов как трассеров в южном полушарии сильно затрудняется продолжающимися испытаниями ядерного оружия в южной части Тихого океана. Полное сравнение между различными моделями общей циркуляции атмосферы и результатами исследований, подобных данному, может быть проведено только после сбора данных, не подвергшихся влиянию результатов ядерных испытаний.