A comparison of tritium and strontium-90 fallout in the Southern Hemisphere

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ABSTRACT

The histories of THO and Sr-90 fallout in the Southern Hemisphere are compared for the data available to June 1966.

The seasonal variation pattern (July–June) for THO in mid-latitude oceanic rainwaters is fairly closely reproduced from year to year, and peaks in late winter–spring. The peak coincides with the absence of the temperature inversion at the polar tropopause, suggesting that the stratosphere-troposphere exchange of the bulk of Southern Hemisphere THO fallout takes place over the polar regions.

For Sr-90 in mid-latitude rainwaters, the seasonal variation pattern is not so closely reproduced from year to year, but shows a broad summer maximum and winter minimum. It appears that the bulk of the bomb particulate products mixes southward within the lower stratosphere and enters the troposphere at mid-latitudes, the seasonal fallout variation being similar to that found for the Northern Hemisphere, but 6 months out of phase.

Consideration of the balance of THO in the troposphere leads to a relation which allows a seasonal variation pattern for the stratosphere-troposphere exchange of THO to be deduced from the rainwater concentrations.

Stratospheric measurements of bomb particulate products are taken into account in an assessment of Sr-90 southward mixing paths. An attempt is also made to deduce the origin and possible southward mixing paths for thermonuclear THO appearing in the Southern Hemisphere.

The history of the THO/Sr-90 annual (July–June) mean activity ratio is quite erratic, and it is not constant over the entire Southern Hemisphere at any one time.

Introduction

In a summary of worldwide tritium fallout data from the International Atomic Energy Agency/World Meteorological Organization precipitation network, Thatcher & Payne (1965) have suggested that THO fallout in the Southern Hemisphere may bear a constant relationship to Sr-90 fallout, and that THO fallout may be thereby deduced in unsampled areas, and at unsampled times, if experimental measurements of Sr-90 fallout are available. These authors compared the THO and Sr-90 fallout for several Northern and Southern Hemisphere stations during the period April–December 1963. In the Northern Hemisphere the THO/Sr-90 ratios (average deposition rates expressed as mc per cm of precipitation per km^2) varied considerably (75–364 over 9 stations). For the Southern Hemisphere the ratios were much lower and roughly constant (53–63 over 6 stations between 25° S and 51° S).

Thatcher & Payne suggest that the THO/Sr-90 deposition rate ratio may be constant with time in the Southern Hemisphere. If this is so, the mechanisms of deposition of the two types of fallout would have to be essentially similar.

Closer inspection of the histories of THO and Sr-90 fallout in the Southern Hemisphere shows that the fallout mechanisms are quite dissimilar. The fallout of Sr-90 from injections into the lower tropical stratosphere has followed a fairly typical pattern. Appearance at ground level follows in the first spring season after stratospheric injection, and the mean stratospheric residence time is of order 1 year (e.g quantities of particulate fallout from the 1954 U.S. Castle test series were deposited in Antarctic snows during the 1954–55 southern summer

(Libby, 1956; Picciotto & Wilgain, 1963; Vickers, 1963; Wilgain *et al.*, 1965)). Movement of particulate debris within the lower stratosphere has been well illustrated by the earlier results of the High Altitude Sampling Programme (see e.g. Stebbins, 1961; Friend *et al.*, 1961), and the more recent results of the Project Stardust phase (Feely *et al.*, 1966), which indicate that generally 80 % or more of the stratospheric burden of Sr-90 is in the lower stratosphere.

The behaviour of the Southern Hemisphere THO fallout up to 1964 has been discussed in an earlier paper by the present author (Taylor, 1966). The THO fallout registered at ground level appeared to have no component which was characteristic of the behaviour registered for the particulate fallout originating in the lower tropical stratosphere. For THO the first fallout at ground level after an equatorial test series appeared to occur in the second spring season after injection, and the mean stratospheric residence time exhibited was much greater than 1 year.

This paper proposes to examine more closely the patterns of Sr-90 and THO fallout, in order to show clearly the differences involved in the fallout mechanisms. One section will be devoted to the seasonal variation of both types of fallout to illustrate the differences in the stratosphere-troposphere exchange mechanisms, and in the histories of both forms in the troposphere. Further sections will deal with stratospheric mixing paths, the year-to-year history of each type of fallout, and with the behaviour of the THO/Sr-90 ratio. It will be necessary first to make a digression to consider which data will be used for each isotope. In the case of THO the decision is clear cut, but it will be seen that problems arise over the available Sr-90 data.

Tritium data

The THO fallout data covering the greatest time span for the Southern Hemisphere originates from the Tritium Laboratory of the New Zealand Institute of Nuclear Sciences, Lower Hutt (D.S.I.R.), where tritium measurements have been carried out since 1958. The results obtained at this laboratory till 1964 have already been published (Taylor, 1966). In nearly all cases the activity of samples after enrichment is such that the statistical counting error is less than 2 %. Repeatability tests and the comparison of various sets of interrelated data show that the accuracy of the electrolytical enrichment process is probably somewhat better than the conservatively applied standard deviation of 6 % may suggest. The rainwater collection station at Kaitoke, near Wellington, has supplied a continuous record of THO fallout since 1960. Data for other stations is at present not so comprehensive, but serves to complement the picture obtained from the Kaitoke record.

There is some data available from other laboratories (I.A.E.A. Tritium Lists), but none covering any prolonged period of time. In most cases the data appears to be reasonable, but the author has encountered concentrations measured by Northern Hemisphere laboratories which are unexpectedly high, and, by comparison with the picture suggested by the New Zealand data, appear only to be explainable as being due to contamination during storage and/or processing in the highly tritiated atmosphere of the Northern Hemisphere. Accordingly it is felt that a comparison with the New Zealand data must be used as a guide for the interpretation of other measurements of Southern Hemisphere waters.

Strontium-90 data

Several sets of Sr-90 rainwater concentrations are available for comparison with the THO fallout at Kaitoke. The levels of Sr-90 fallout are low, and experimental and statistical inaccuracies have a considerable influence on the measurements. It was felt advisable to compare the available sets of data to see whether close agreeement between different laboratories was apparent. Four laboratories are involved in measurements over an extremely small area—probably a unique opportunity for intercomparison. The following summary lists the sets of rainwater measurements which were considered.

United Kingdom Atomic Energy Authority, Health Physics and Medical Division, have operated fallout collections at Ohakea Aerodrome (91 km N of Kaitoke) since 1955. Quarterly samples are collected. The data are published in reports issued by the measuring laboratory, and in the quarterly fallout sum-

Table 1. *Precipitation-weighted mean activities of Sr-90 in New Zealand rainwater*

Measuring laboratory	Collection station	Precipitation-weighted mean activities ($\mu c \cdot cm^{-1} cm^{-2}$) in stated "fallout years" 1955–56	1956–57	1957–58	1958–59	1959–60	1960–61	1961–62	1962–63	1963–64	1964–65	1965–66
U.K.A.E.A.	Ohakea	9.9	9.4	7.4	8.8	9.0	11.0	10.6	15.6	20.6	35.1	
D.S.I.R.	Lower Hutt					6.8	7.35	7.55	14.3	17.1	35.0	13.9
H.A.S.L.[a]	Wellington						10.0	7.2	10.9	14.5	25.7	9.6
N.R.L.	Taita					7.1	7.8	8.8	14.5	19.7	34.4	13.8
N.R.L.	New Zealand network					5.4	8.7	10.7	14.6	20.1	32.4	14.8
I.N.S., mean value[a]						7.1	8.7	9.4	14.7	19.4	34.2	14.1
Root square deviation from mean (%)						18	16	14	3	7	3	3

[a] Means exclude H.A.S.L. data.

maries of the U.S.A.E.C. Health and Safety Laboratory (HASL).

National Radiation Laboratory, Christchurch, have operated the collection station at the D.S.I.R. Soil Bureau at Taita (23 km SSW of Kaitoke) since 1959. Monthly composite samples are taken. The same laboratory has operated a network of stations throughout New Zealand. From 1959 to 1962 this network comprised 6 stations. Since 1962 the number of stations has been increased to 9. Data are published in quarterly reports by the measuring laboratory, and also in the HASL quarterly summaries.

The Institute of Nuclear Sciences, D.S.I.R., has a collection station at Gracefield, Lower Hutt (31 km SSW of Kaitoke) Daily samples are taken and combined to form monthly composites. Data are published in the quarterly reports of the National Radiation Laboratory, Christchurch, and also in the HASL quarterly summaries.

HASL operate a collection station for monthly samples in the grounds of the U.S. Embassy in Wellington (43 km SSW of Kaitoke). Data are published in the HASL quarterly summaries. Inspection of the data showed that the precipitation amounts quoted were normally in exact agreement with those recorded by the New Zealand Meteorological Service at Kelburn, only about 1 km away. In a few cases, however, it was clear that the HASL precipitation values were not correct, even allowing for small variations between the two sites, and possible overlap of samples between months. Accordingly, for the following analysis, HASL concentration values are obtained by dividing the measured HASL deposition values by the Kelburn precipitation values. This procedure resulted in slightly better agreement between the HASL concentrations and those recorded at Taita and Gracefield. After this initial correction the monthly activities at these three stations were compared to remove the presence in the HASL data of occasional deposition values which were clearly impossible on the evidence of the concentrations recorded at Taita and Gracefield. In such cases interpolated values were calculated for the deposition at Wellington, based on the concentrations recorded at Taita and Gracefield and the precipitation records. Only a few values required correction in this way, but the influence of the false values on the yearly average concentrations had been considerable.

As has already been applied for THO (Taylor, 1966), the concept of a "fallout year" will be applied to the Sr-90 data, since the fallout pattern for the Southern Hemisphere is out of phase with the calendar year. The "fallout year" will be taken to begin in July; e.g. the term "fallout year 1962–63" means the period July 1, 1962–June 30, 1963.

In Table 1 are listed the mean yearly concentrations of Sr-90 in rainwater (precipitation weighted) recorded by the four laboratories. The results of Table 1 may be summarized as follows. In the years 1962–66 it appears that

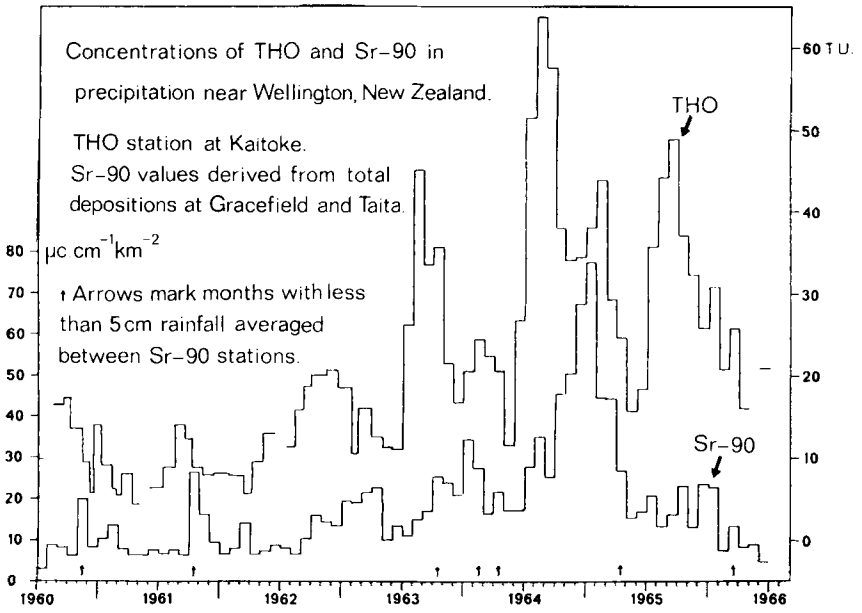

Fig. 1. Tritium and Sr-90 concentrations in rainwater near Wellington, New Zealand, 1960–66.

the higher concentrations, and probably also improvement in experimental accuracy, have made for better agreement than for earlier data. There is good agreement between the N.R.L. Christchurch, U.K.A.E.A. and D.S.I.R. data. The indication is that similar yearly mean activities do occur over the whole area of New Zealand. The HASL data of 1962–66, in addition to the precipitation and deposition errors referred to earlier, are systematically between 20 and 30 % lower than the values from the other laboratories. Since HASL measure deposition directly and then derive the concentration by using the rainfall recorded nearby with a standard rain gauge, it may be that their concentrations are affected by a difference in the collection efficiencies of the fallout collector and the rain gauge.

The monthly mean activities at the individual stations do not correspond so well, partly due to the high statistical error of a single experiment, and also to non-uniformity of monthly rainfall. Studies at the U.K.A.E.A. laboratory (Cambray *et al.*, 1964) have indicated that the standard deviation for a single rainwater sample is about 20 %.

To obtain yearly means of Sr-90 activity to compare with the THO data at Kaitoke, the following procedure has been adopted on the basis of the preceding remarks. All the HASL data is discarded. The means of the other values are then taken and listed at the foot of each column in Table 1, together with the root mean square deviation (in per cent) of the four values from the overall mean. It should be noted that the N.R.L. Taita data is included in the New Zealand network mean, and that the process of applying equal weight to each value may be questionable. However, the network covers the entire latitudinal spread of New Zealand, and therefore the network mean may probably be considered as a "single station mean" at approx. the latitude of Kaitoke. As to weighting, it is found that, during the years 1959–62, the statistical standard deviation of the overall mean activity is so high as to make weighting of no significance. In later years the agreement between the values is much better and the mean would be unaffected by any preferential weighting.

Seasonal patterns of THO and Sr-90 fallout. Tropospheric fallout and stratosphere-troposphere exchange

In Fig. 1 a comparison has been made between the concentrations of THO and Sr-90 recorded in precipitation for the Wellington area.

For Sr-90 the monthly mean activities ($\mu c \cdot cm^{-1}$ km^{-2}) for the Lower Hutt and Taita stations have been used. (The mean values are derived by summing the depositions and dividing by the total precipitation. At Lower Hutt, deposition is measured; for Taita, the measurement gives activity, which is multiplied by precipitation to give the deposition.) From July to December 1960 inclusive the activities are these for the Lower Hutt station alone. The tritium data are for the station at Kaitoke.

When examining Fig. 1, it should be borne in mind that the standard deviation of a tritium measurement is about 6 %, while that for a Sr-90 activity is probably 15–20 %. To keep the diagram as clear as possible error bars are omitted.

It is thought that in months of low precipitation the presence of dry fallout in the Sr-90 samples might tend to give falsely high concentrations. Accordingly, the months where the mean precipitation at the two Sr-90 stations was less than 50 mm have been marked by vertical arrows immediately above the time axis. It is seen that in most of these cases a high concentration was recorded.

Once these anomalous low precipitation Sr-90 activities have been recognized, there are two features of the diagram which stand out very clearly.

(1) In most "fallout years" the main peak of tritium concentration occurs during the late winter and early spring months, July–October. A second feature, not considered in the discussion of the earlier data (Taylor, 1966), but more apparent since new data has become available, is the appearance of a minor peak of concentration in the summer months January–March. This peak is apparent in all years except 1961–62, and possibly 1965–66. Although this peak is not too well marked when one considers the possibility of overlap due to the experimental standard deviation, the appearance over several years is significant. In 1961–62 it is clearly absent. In 1962–63 there is a much broader spring peak than normal, and only a brief drop before a recovery in February–March.

(2) In no year does a peak of Sr-90 activity coincide with the main spring tritium peak. Between 1960–62 there are only faint suggestions of a Sr-90 peak in the summer months. In later years summer Sr-90 peaks are quite definite.

Seasonal variation of THO fallout

Thermonuclear THO passing from stratosphere to troposphere becomes intimately associated with the tropospheric water vapour cycle. The transport of THO in the troposphere has been discussed by various authors (Bolin, 1958; Eriksson, 1965 and 1967; Roether, 1967). Because the sources and sinks for THO differ from those of normal water vapour, there are considerable differences between the atmospheric transports of the two forms. In an oceanic area, such as the area whose meteorological regime encompasses New Zealand, the balance of THO in the troposphere may be expressed by the equation

$$Q \approx \nabla \cdot \mathbf{F}^* + c_P P - \Phi(c_S - h c_V) \qquad (1)$$

ignoring the small terms expressing the changes in tropospheric water vapour content. Here

- Q is the rate at which THO is passed downwards from the stratospheric source.
- $\Delta \cdot \mathbf{F}^*$ expresses the outwards divergence of the horizontal THO flux $\mathbf{F}^*$.
- c_P is the concentration of tritium in precipitation.
- P is the precipitation rate.
- Φ is the total evaporation rate from the ocean surface $\approx E/(1-h)$, where E is the net evaporation rate and h the relative humidity close to the ocean surface.
- c_S is the concentration of tritium in the evaporate from the ocean surface.
- c_V is the tritium concentration of the atmospheric vapour in active exchange with the ocean surface.

As already postulated by Eriksson (1965 and 1967), and now confirmed by actual ocean tritium inventories (Münnich & Roether, 1967), the last term in eq. (1), which describes the molecular exchange of THO between atmosphere and ocean, represents in most areas the major process of removal of THO to the oceans.

On the basis of eq. (1) it is difficult at first sight to predict the interplay of the various balancing terms. For instance, one cannot say whether a period of low precipitation will lead predominantly to an increase of c_V, and hence to an increase in removal by molecular exchange, or whether the balance will be restored by an increase of $\nabla \cdot \mathbf{F}^*$. Likewise it is not clear whether an increase in c_P is due to an increase in Q

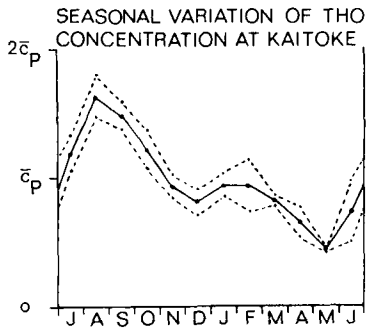

Fig. 2. Seasonal variation of tritium concentration in Kaitoke rainwater. $\bar{c}_P$ is mean concentration over entire "fallout year". Dotted lines indicate mean root square deviations of monthly values from the mean values joined by the central line.

or to a seasonal change in Φ. It is not the concern of this paper to discuss in comprehensive detail such problems of the tropospheric tritium balance, but only to consider whether any simple relationship might exist by which one might deduce the seasonal variation of Q from the tropospheric tritium concentrations. To reach such a relationship, the precipitation term in eq. (1) is eliminated in the following manner. For water the balance equation may be written

$$0 = -\nabla\cdot\mathbf{F} - P + \Phi(1-h), \qquad (2)$$

where $\nabla\cdot\mathbf{F}$ is the divergence of the water vapour flux $\mathbf{F}$. Multiplying eq. (2) by c_P and adding to eq. (1) yields

$$Q = \Phi[(c_P - c_S) - h(c_P - c_V)] + (\nabla\cdot\mathbf{F}^* - c_P\nabla\cdot\mathbf{F}). \qquad (3)$$

The stratosphere-troposphere transfer rate Q is thereby expressed as the sum of an exchange term and a flux divergence term. It will be shown in what follows that the exchange term is dominant.

In the exchange term of eq. (3) it can be taken at all times for the New Zealand area that $c_S \leqslant 0.1\ c_P$. Unfortunately there are relatively few measurements to indicate the relationship between c_P and c_V, but recently Roether (1967) has reported on data for rain and vapour samples taken during cruises of the German research vessel *Meteor*, which suggest that the oceanic rain, on the average, lies very close in concentration to the vapour in the lowest few metres of the troposphere, i.e. $c_P \approx c_V$. It can therefore be said with reasonable certainty that $h(c_P - c_V) < 0.2\ (c_P - c_S)$, and the whole term may be expressed as kc_P, where $k > 0.7$ and has probably very little seasonal variation.

In the flux divergence term of eq. (3) there will be a tendency for the advective components to balance out, although by no means completely since c_P will not in general be equal to the mean concentration of the flux $\mathbf{F}^*$. As regards the eddy components, the meridional distribution of c_P suggests that THO passing downward into the moist layer is reasonably well mixed, at least in the region 30 – 90° S, so that the eddy component in the moist layer may be quite small even although it may have the same sign as $-c_P\nabla\cdot\mathbf{F}$ in the flux divergence term. From eq. (2) it can be deduced that $|\nabla\cdot\mathbf{F}| < 0.2\ \Phi$ unless $P > 2E$. Therefore it is seen that $(\nabla\cdot\mathbf{F}^* - c_P\nabla\cdot\mathbf{F})$ is probably considerably less than $0.2\ c_P\Phi$. Thus the flux divergence term in eq. (3) is very minor in comparison to the exchange term.

The conclusion is reached that Q must be approximately proportional to the function $c_P\Phi$, provided of course that c_P and Φ are averaged over periods long enough to remove the influence of transient conditions. Taking monthly c_P values one can normalize c_P to the yearly (July–June) mean concentration $\bar{c}_P$, i.e. take $c_P = A\bar{c}_P$. Averaging A values arithmetically over several years, one obtains the best approximation to the seasonal variation of c_P.

Table 2. *Monthly variation of tritium concentration and $c_P\Phi$ in New Zealand area, based on Kaitoke tritium data, 1963–66, and evaporation data from Albrecht (1951)*

Month	$A = c_P/\bar{c}_P$	Φ (gm. month^{-1})	$A\Phi$ (gm. month^{-1})
July	1.18 ± 0.14	91.3	108 ± 13
August	1.63 ± 0.16	75.9	124 ± 12
September	1.48 ± 0.11	65.8	98 ± 7
October	1.21 ± 0.15	44.5	54 ± 7
November	0.93 ± 0.09	33.4	31 ± 3
December	0.81 ± 0.10	28.6	23 ± 3
January	0.94 ± 0.09	42.4	40 ± 4
February	0.94 ± 0.20	58.7	55 ± 12
March	0.83 ± 0.04	66.7	55 ± 2.5
April	0.66 ± 0.12	75.9	50 ± 9
May	0.44 ± 0.02	82.5	36 ± 2
June	0.74 ± 0.24	96.5	71 ± 24

Monthly values of c_p for Kaitoke are available for only three years from July 1963. The averaging of these three years data results in the seasonal variation pattern shown in Fig. 2. The mean root square deviations of the averages are enclosed by the dashed lines. It is seen that the spring peak is clear, but that the smaller summer peak is by no means definite within the statistical limits.

For the total evaporation rate Φ, monthly values have been calculated for the New Zealand region of the South Pacific using humidity and net evaporation data of Albrecht (1951). Although the quantitative accuracy of these values is questionable because of the indirect methods of ocean evaporation determination, the relative values from month to month may be more reliable, and the seasonal variation is what matters here. The evaporation rate is higher in winter than in summer because of the influence of wind.

Values of A, Φ and $A\Phi \propto Q$ are shown in Table 2. The seasonal variation of $A\Phi \propto Q$ is shown in Fig. 3, together with the mean root square deviation envelope resulting from the averaging of A. No errors have been estimated for Φ. It is seen that the peak of Q in late winter—early spring is even more pronounced than it appears from the rainwater concentrations (Fig. 2). Also it is clear that the statistically doubtful summer peak of c_p is a response to a peak in Q, and is not a tropospheric buffer effect arising from a lower rate of molecular exchange during the summer. The value for June has a high root mean square deviation, since the June concentrations seem to "anticipate" the coming "fallout year" to a variable extent from year to year, i.e. the new "fallout year" actually begins at some time during June.

It is consequently necessary to postulate a mechanism which might explain the occurrence of the peak of THO concentration in late winter—spring at a season when the Sr-90 concentrations do not peak. The timing of the THO peak coincides closely with the season when no temperature inversion exists at the south polar tropopause (see e.g. Court, 1942; Picciotto *et al.*, 1960; Aldaz, 1965). It therefore seems likely that mixing can occur across the polar tropopause at this season, and that THO is brought down from higher stratospheric altitudes within the circumpolar vortex, and is passed into the troposphere over the polar regions. Such a

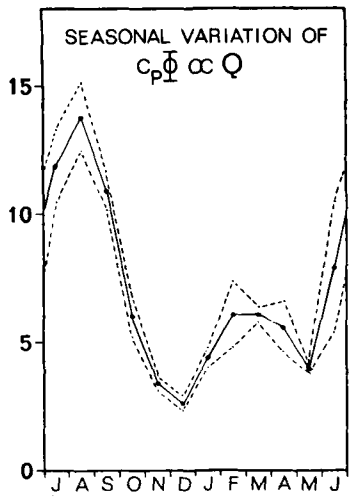

Fig. 3. Seasonal variation of the function $c_p\Phi$, proportional to Q, the stratosphere-troposphere exchange rate of THO in the Southern Hemisphere. c_p values are from the seasonal variation of tritium concentration in Kaitoke rainwater (Fig. 2). Φ from data of Albrecht (1951). No errors incorporated for Φ.

mechanism could also account for the well known maximum of ozone observed during late winter—spring over Antarctica (Aldaz, 1965) and at mid-latitudes (see e.g. Dyer, 1966).

It is natural to enquire whether stratosphere-troposphere exchange of the bulk of THO over the South Pole might produce a difference between the meridional deposition distributions of THO and particulate products if the latter enter the troposphere predominantly at mid-latitudes. However, the possibility of different meridional deposition distributions depends critically on the degree of meridional mixing which occurs in the upper troposphere before the fallout products reach the lower tropospheric regions where they participate in the various processes which deposit them on the earth's surface. Present evidence suggests that the upper tropospheric mixing in the region 30–90° S is intense, so that there is little meridional gradient of mixing ratio by the time fallout products reach the lower troposphere. The amounts of fallout deposited in different localities are then dependent primarily on the final processes of removal, with probably only minor influence due to any latitudinal differences in the stratosphere-troposphere exchange.

As an illustration of this problem it is instructive to examine the relative amounts of THO and Sr-90 fallout in the Antarctic and at southern mid-latitudes.

For Sr-90, the data of Wilgain *et al.* (1965) show that the total deposition at the South Pole to the end of 1962 was only about 8 % of that at 40–50° S, at Scott Base (78° S, 170° E) 16 %, and at Base Roi Baudouin (70° S, 24° E) 20 %. Data for Byrd Station (80° S, 120° W) for the period 1962–64 (Volchok, 1965) indicate deposition of about 10 % of the amount recorded in New Zealand. The atmospheric (Lockhart *et al.*, 1966) and precipitation (Wilgain *et al.*, 1965) Sr-90 concentrations are very close to those at mid-latitudes. For Base Roi Baudouin, the concentrations in snow for the years 1955–62 are very close to those recorded in New Zealand rainwater (Table 1). At the South Pole the concentrations in snow are about twice as high as at Roi Baudouin, although such a concentration difference is not apparent in the atmospheric concentrations of long-lived β-products between the South Pole and coastal stations (Lockhart *et al.*, 1966). This difference in the snow concentrations may possibly be related to the influence of dry fallout. The great difference between the Sr-90 depositions in Antarctica and at mid-latitudes results from the low precipitation in Antarctica. The relative depositions are in approximate proportion to the precipitations.

For THO the deposition mechanisms in Antarctica are complex. In January 1964 a surface snow sample at the South Pole had a tritium concentration of 200 T.U. More samples were taken by the author 11 months later in December 1964. Samples to 3 cm and 60 cm depth (1.2–24 cm water equivalent) at one site and to 3 cm depth at another site, showed uniform concentration of about 650 T.U. If the sample to 60 cm depth represented unconsolidated snow, the deposition of THO for the year Jan. 1964–Jan. 1965 is about $650 \times 7 = 4550$ T.U. cm·cm^{-2}, where 7 cm water equivalent is taken as the annual deposition (Gow, 1965). However, the surface location of this sample was one of smooth snow. Smooth surfaces at the South Pole have been shown (Gow, 1965) to represent consolidated accumulation. Shallow sastrugi formations up to 40 cm high are formed during winter but by mid-January even the largest sastrugi are substantially levelled. Thus the 60 cm sample probably represented about 3 years accumulation of snow, and in this case the THO deposition between January and December, 1964, was at least $(7 \times 650 + 14 \times 450) = 11{,}000$ T.U. cm·cm^{-2}. The accumulation of considerable amounts of THO in sub-surface snow is thought to be due to an exchange effect caused by the penetration of the snow by the persistent winds of the polar plateau.

For the period January–December, 1964, the THO deposition at about 40° S may be estimated as about 2.5 times the deposition by precipitation (thus taking into account the molecular exchange across the ocean surface), or about $2.5 \times 35 \times 150 = 13{,}000$ T.U. cm·cm^{-2}. The deposition of THO per unit area on the Antarctic polar plateau was thus at least 35 % of that at 40° S, but most probably of more equal magnitude. The THO/Sr-90 ratio over the polar plateau is therefore greater by a factor of at least 4, and possibly even as much as 10.

Similarly high concentration (550 T.U., top 30 cm) to the South Pole samples was also found in Dec. 1964 in surface snow on Nimrod Glacier (82° S, 155° E, altitude 2.1 km). In contrast, lower concentrations existed at Shackelton Glacier (84.5° S, 5° W, 1.3 km). 130 T.U., top 60 cm; Byrd Station (1.5 km), 120 T.U., top 3 cm; 185 T.U., top 60 cm; Scott Base (sea level) 180 T.U., blown snow. Thus the deposition of THO in coastal low-lying areas between January and December, 1964, was at least $150 \times 20 = 3000$ T.U. cm·cm^{-2}, this being a low estimate because it does not take into account molecular exchange with the surrounding ocean, the loss of precipitated snow through summer ablation, and the possibility of some exchange of atmospheric vapour with sub-surface snow. In this case an enhanced THO/Sr-90 ratio with respect to mid-latitudes is not definitely confirmed but appears probable.

The THO concentrations just quoted for the Antarctic regions indicate the presence of an altitude effect. The existence of higher atmospheric THO concentrations above about 2 km has been already suggested by snow measurements in the New Zealand Alps (Taylor, 1964), and is clearly indicated by profile measurements of tropospheric water vapour reported by Ehhalt (1967).

To explain the minor summer peak of Q (Fig. 3) at a time which lies at the end of what

will be shown later to be a broad summer maximum of Sr-90 fallout, it is suggested that, after the reformation of the Antarctic tropopause in early summer, some THO remaining in the Antarctic stratosphere will be free to mix towards mid-latitudes to provide the source of this summer peak through a mid-latitude stratosphere-troposphere exchange. Since the Antarctic tropopause will reform at a different time each year, and since the downwards transport over the pole probably varies in strength from year to year, the late summer peak will vary in intensity from year to year, and this appears in the measured concentrations.

The seasonal variation of Kaitoke THO concentration indicated in Fig. 2 is reproduced fairly closely each "fallout year" during the period 1960–66. In 1962–63 a somewhat broader spring summer peak occurred. This may have been due to some THO accompanying the Sr-90 from the 1962 U.S. Pacific test series. However, 1962 was a year of slow polar stratospheric warming between winter and summer, and this may be the reason for the broader peak.

Other island stations in southern mid-latitudes show essentially the same seasonal pattern (see e.g. data for Gough Island (40° S, 10° W) and Marion Island (47° S, 38° E) (Taylor, 1966) measured by the Radioactive Dating Laboratory, Stockholm; such data as are available from the New Zealand Laboratory for stations at Campbell Island, 52° S, 169° E; Invercargill 46° S, 168° E; Kaitaia 35° S, 173° E; Rarotonga 21° S, 160° W). Stations in Australia, South Africa and South America are subject to strong continental influence, and sufficient data is not yet available to discuss their seasonal patterns.

As to prethermonuclear THO, virtually all is produced in the stratosphere, and in the upper tropospheric regions between mid-latitudes and the pole (Lal, 1963). Its fallout distribution over the Southern Hemisphere was therefore probably similar to that of thermonuclear THO, since it should have been similarly distributed as it entered the lower troposphere. The prethermonuclear concentration of Antarctic coastal snows has been fairly well established by various sources (Begemann & Libby, 1957; Begemann, 1958; von Buttlar & Wiik, 1965; Institute of Nuclear Sciences, New Zealand, unpublished measurements) at about 15 T.U. With a precipitation amount of about 20 cm water equivalent the fallout, without including the possibility of molecular exchange over the oceans surrounding coastal Antarctica, is about 300 T.U. $cm \cdot cm^{-2}$. The prethermonuclear tritium concentration in New Zealand rainwaters is thought to have been about 1.5 T.U. (Grant-Taylor & Taylor, 1967). Thus the fallout at mid-latitudes including molecular exchange is about 560 T.U. $cm \cdot cm^{-2}$. The coastal Antarctic prethermonuclear THO deposition was thus about 50 % of that at mid-latitudes, and the concentrations differ by an amount implying similar mixing ratios for THO at each locality.

Seasonal variation of Sr-90 fallout

The Lower Hutt/Taita Sr-90 data for the period 1960–66 (Fig. 1) may be analyzed in the following manner. For each "fallout year" the mean concentration $\bar{C}$ of Sr-90 in rainwater is evaluated by summing the monthly depositions at both stations and dividing by the total precipitation. The monthly concentrations C from Fig. 1 are then expressed as $C = B\bar{C}$, i.e. the C values are normalized to $\bar{C}$. Over the period 1960–66, the B values have been arithmetically averaged to give the seasonal variation pattern depicted in Fig. 4*a*. Here the B averages are joined by the full line. The dotted lines join the points which indicate the mean root square deviations of the individual B values from the mean values. A broad summer maximum is indicated but the detail of the pattern is obscured by the large mean root square deviations, which range from 15 % to 45 %. At a rough estimate the mean root square deviations due to experimental inaccuracies are in the range 6–10 %, so that it is clear that the deviations are due to other causes. In order to see whether these large deviations are reduced by taking data over a larger area, a similar analysis has been performed for the data from the New Zealand network operated by National Radiation Laboratory, Christchurch. The seasonal variation pattern for the network is shown in Fig. 4*b*. The patterns are similar, and for the network the mean root square deviations are on the average slightly less, ranging from 6 % to 34 %, but still considerably greater than the experimental inaccuracies. It appears that the seasonal pattern of Sr-90 concentration in rainwater is not so regular from year to year as was

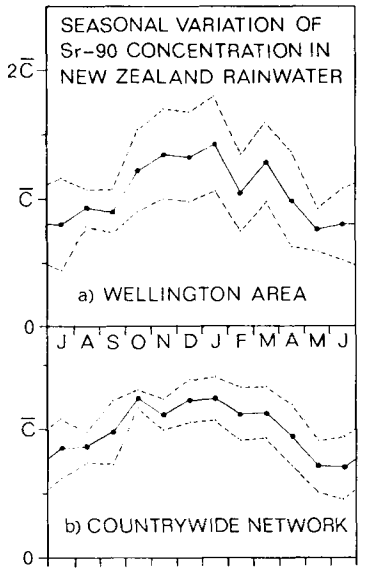

Fig. 4. Seasonal variation of Sr-90 concentration in (*a*) Wellington rainwater (Taita and Gracefield data); (*b*) New Zealand network mean (9 stations). Dotted lines indicate mean root square deviations of monthly values from the mean values joined by the central line. $\bar{C}$ is mean concentration over entire "fallout year" (precipitation-weighted).

found for THO. There are in fact a number of reasons why this should be so. In part the Sr-90 concentrations are affected by the amount of precipitation with a tendency to anomalously high concentrations in dry months because of the effect of additional dry fallout in the collector. Dry fallout has been shown to amount to about 10–20 % of the total fallout at normal rainfall rates (Junge, 1958; Small, 1960). The Sr-90 concentration is affected by the amount of evaporation suffered by raindrops, and the scavenging efficiency for radioactive aerosols, both of which are factors which depend on rain intensity and cloud characteristics. Thus a month characterized by many days of light, highly evaporated rainfall would produce higher concentration than a month where an equivalent amount of rainfall occurred on only a few days with high intensity.

The high deviations of the B values from their mean are also partly attributable to the relative proportions of Sr-90 from two different sources. As will be discussed in the following section, considerable quantities of fission products are mixed southward at low stratospheric altitudes, and appear to be passed to the troposphere at mid-latitudes. It is known that a certain amount of Sr-90 exists above the fallout situated in the lower stratosphere, as particles small enough to be unaffected by sedimentation. The evidence for such fission product fallout is provided by the data obtained for Rh-102 (Kalkstein, 1962) and Cd-109 (List *et al.*, 1966) fallout. Martell (1966) has concluded that the artificial radioactivity which persists above 25 km is associated with particles, presumably micrometeorites, mainly with radius less than 0.02 μ. It is not until these particles are mixed down to the lower stratosphere that they become attached to larger particles and are eventually released to the troposphere. One would expect such fallout to undergo stratospheric mixing similar to that for THO, so that the seasonal pattern for Sr-90 would exhibit a tendency to greater late winter-spring concentrations in years when the high stratospheric component is of magnitude comparable to particulate fallout originating from the lower stratosphere. One judges from the B values that such a situation occurred in "fallout year" 1965–66.

As regards the mid-latitude stratosphere-troposphere transfer, the broad summer peak and winter minimum occurring in the fallout rainwater concentrations suggests two possibilities:—either the peaking is controlled by seasonal variations in the stratosphere-troposphere transfer, or else by seasonal variations in the rate at which the fallout is delivered to the "gap" at which the transfer occurs. The stratospheric concentrations recorded by the HASP programme (Feely *et al.*, 1966) suggest that the peak occurs in the actual stratosphere-troposphere transfer.

Sr-90 fallout in New Zealand in the years 1955–66

It is seen from Table 1 that the Sr-90 activities in New Zealand rainwater remained fairly constant at about 10 mc·cm^{-1} km^{-2} during the period July 1955 to June 1962. A rise to more than 3 times this value occurred during the period July 1962–June 1965. A steep drop then occurred during the "fallout year" 1965–66.

Before examining the situation prior to 1962–63 in greater detail, it will be advisable to consider first the later data in parallel with measurements of fission products in the strato-

sphere between 1962 and 1965. The history of the thermonuclear testing of 1961–62 is by now well known (see e.g. Salter, 1965), and the fission yields are fairly well documented in reports issued by the U.S. Atomic Energy Commission. It appears likely that the yields of the early 1962 U.S. Pacific test series were of magnitude sufficient to account entirely for the increase of mean Sr-90 activity observed in New Zealand in 1962–63. Fresh activity from later tests of this series, and from the July 9 high altitude Starfish test, was certainly present during the "fallout year" 1963–64, but can hardly have accounted for the rise in Sr-90 activity measured in this year. The source of the 1963–64 and the much greater 1964–65 increases is quite clear when one examines the stratospheric patterns of activity measured by the Stardust phase of the American High Altitude Sampling Programme (Salter, 1965; Feely *et al.*, 1966). During 1963 and 1964 a considerable southward spread of particulate debris from the Russian test series of 1961 and 1962 occurred. This is most strikingly shown by the concentrations of Mn-54 recorded in the stratospheric regions below 32 km. Since Mn-54 is a tracer unique to the Russian Arctic test series of 1961–62, this debris cannot be confused with that originating from the 1962 American Pacific test series, as might be the case with other isotopes. A tongue of this Russian debris spread southwards in the stratospheric region between 24 and 32 km. By Jan.–April 1963 the tip of this tongue had just reached the equator. Consequently the first fallout of this debris in the Southern Hemisphere would have occurred during the 1963–64 spring season. In Jan.–April 1964 this tongue of stratospheric activity had extended to southern mid-latitudes and the concentrations were such as to suggest an even greater fallout peak in the subsequent "fallout year" 1964–65. At the 20 km level the Sr-90 and Mn-54 levels increased strongly and simultaneously during mid–1963, but then declined slowly during 1964. The activities of both isotopes show a sharp decline below the 1963 level during early 1965, and this agrees with the maximum fallout of Sr-90 observed in New Zealand precipitation during 1964–65. The levels of activities in the stratosphere at 35°–40° S during 1963–65 correspond roughly to the activities measured in New Zealand precipitation. Thus it seems certain that the Sr-90 precipitation activities between 1962 and 1966 can be explained as follows. During "fallout year" 1962–63, a fresh source of Sr-90 arrived via the lower stratosphere, originating from the 1962 U.S. Pacific test series. If only this series had continued to contribute, levels might have remained roughly the same during 1963–64, according to the decline of the low stratospheric 1962 U.S. contribution and remaining earlier fallout, and with a first fraction appearing from the U.S. 1962 high-altitude debris. (The presence of Sr-90 descending from high altitudes during 1963 was shown by stratospheric balloon measurements over Mildura, Australia, 34° S (Dyer, 1966). The simultaneous downward movement of the Cd-109 tracer from the 1962 high-altitude Starfish test has been reported by List *et al.* (1966).) However, the activities were raised further during "fallout year" 1963–64 by the arrival of the first of the Russian debris spreading southwards at lower stratospheric levels. The stratosphere passed most of this debris to the troposphere during the "fallout year" 1964–65. Although the southern stratospheric inventory was roughly the same between spring 1963 and spring 1964, in 1963 the bulk of the activity was situated close to the equator, and the spreading southwards of this activity during subsequent months provided the situation favourable for the bulk of the activity to be removed to the troposphere. A further point of importance to note about this north-south spread of particulate debris of Russian origin within the lower stratosphere during 1962–64 is that it was not significantly supplemented by particle descent from altitudes above 30 km (Feely *et al.*, 1966), except for the material originating from the American high-altitude tests. Such particle descent would have resulted in an increase in the Mn-54/Sr-90 activity ratio in the lower stratosphere, but this remained steady below 15 during 1963 and 1964, and decreased with time. Also there was no hint of such descent in the stratospheric isotope distributions. Thus it appeared that even the high yield 1961 and 1962 tests did not introduce much particulate debris above the 20 km level. This behaviour had also been suggested for earlier test series at high latitudes (Friend *et al.*, 1961; Feely *et al.*, 1963). This information will be of importance later in this paper, when the

Sr-90 and THO fallout patterns are compared.

Turning now to the activities of Sr-90 in New Zealand rainwater prior to 1962–63, it is at first sight surprising that no strong decrease of activity occurred during the moratorium years 1958–1961. In the light of the conclusions reached above for the fallout after 1962, one must consider the following three sources of activity as being responsible for contributions to the New Zealand Sr-90 fallout after 1958.

(1) The activity injected into the lower stratosphere by Pacific equatorial testing during 1958, and smaller contributions remaining from earlier years. This source of activity should be responsible for the greater part of the Sr-90 in New Zealand precipitation during 1958–59, and perhaps 1959–60, but certainly the contribution from this source will be greatly decreased by 1960–61.

(2) The activity injected into the high equatorial stratosphere by the two U.S. high-altitude tests of 1958, Teak detonated at 77 km, and Orange at 30 km. According to the behaviour of the Rh-102 tracer of the Orange test, the first considerable appearance of this debris at ground level in the Southern Hemisphere occurred during the 1959–60 spring-summer season, and an even greater fraction appeared during 1960–61 (Kalkstein, 1962). This behaviour was also typical of the Cd-109 tracer of the U.S. 1962 Starfish high altitude test, as reported by List *et al.* (1966), where a first appearance occurred in the stratosphere above Mildura, 34° S during 1963, and the first fallout occurred in the 1963–64 spring-summer season.

(3) The activity injected by the Russian Arctic testing of 1957–58. Because of the evidence of the southwards spreading of the 1961–62 Russian debris, only the possibility of fission product debris spreading within the *lower* stratosphere will be considered. The fission yield of the Russian October 1958 series has been estimated at about 12.5–15 MT (Libby, 1959), or about 1.5–2 times the fission yield of the 1958 tests into the lower equatorial stratosphere. The excess over the equatorial low stratosphere yield is thus somewhere in the region 3–8 MT. In 1961 and 1962 the excess fission yields were about 25 and 50 MT respectively (no equatorial testing in 1961). The increase in Sr-90 activity recorded in the Southern Hemisphere during 1964–65 is about 20 $mc \cdot cm^{-1}\ km^{-2}$ above the 1962–63 level. Thus it would be reasonable to expect that the greatest increase arising from the 1958 test series would have occurred in the "fallout year" 1960–61 and would have amounted to an activity of order 1 $mc \cdot cm^{-1}\ km^{-2}$. This contribution should have dropped considerably in 1961–62.

With these three contributions one can explain the maintenance of the Sr-90 activity level up till the end of "fallout year" 1960–61. It remains to explain the level recorded during 1961–62. Balloon soundings fof the stratosphere above Mildura, Australia, 34° S (Dyer, 1966), during 1961, showed isopleths for Sr-90 which suggested vertical transport associated with the descent of the 1958 high altitude debris. It appears therefore that further fallout of 1958 high-altitude debris occurred at ground-level in 1961–62, subsequent to that which had already occurred, on the evidence of the Rh-102 levels, in 1959–60 and 1960–61. This must have served to keep the 1961–62 Sr-90 level roughly equal to that of 1960–61.

The continuing transfer of 1958 high-altitude debris from upper to lower stratosphere as late as 1961 confirms that these fission products are associated with particles small enough to be uninfluenced by sedimentation.

For the earlier data, all that can be said is that the U.K.A.E.A. data indicate that the Sr-90 concentration in New Zealand rainwaters remained approximately at the same level during the years 1955–1959. This behaviour is consistent with the continual renewal of the reservoir of Sr-90 in the equatorial stratosphere by testing during 1956, 1957 and 1958.

The stratospheric mixing paths for THO

The general history of THO fallout in the Southern Hemisphere up to 1964 has been already discussed in a previous paper (Taylor, 1966). In that paper it was deduced that the THO fallout behaved as high-altitude fallout, and that no significant component showed behaviour typical of the lower stratospheric southwards mixing and removal to the troposphere exhibited by particulate products. Undecided were the questions of the height of formation of the THO (i.e. whether it was

created at lower stratospheric altitudes, or fractionated to higher altitudes), and of the removal path from the stratosphere (i.e. if THO is created at low stratospheric altitudes is it immediately removed across the tropopause, or does it mix first to higher altitudes so that all the THO eventually appears as high-altitude debris? — What is the mixing path for the Soviet THO which predominates in the Southern Hemisphere after 1963?). Such questions will be discussed in this section.

In the earlier paper it was deduced that the increase in THO fallout recorded at Kaitoke in 1962–63 and later years would have to be a scribed to THO of Soviet origin, since the published fusion yields of the 1962 U.S. Pacific tests were, on the basis of earlier evidence, not great enough to account for the increases recorded at Kaitoke in these years. (There remains the possibility that the 1962 U.S. series produced very much more THO per MT fusion yield than in previous series. A greatly increased THO yield (by a factor of about 10) for this series could account for the entire subsequent history of THO in the Southern Hemisphere, but it seems unlikely that such information would not be published in view of the open nature of the information about other tracer isotopes. Further the domination of particulate fallout of Soviet origin in the Southern Hemisphere after 1963 leads one to expect that the same would occur for THO even if a different southward mixing path is followed. The possibility of abnormally high THO yield from the U.S. 1962 series is therefore neglected.)

If the 1962–63 rise of 6 T.U. at Kaitoke results from 1961 Soviet THO, then this THO must be mixing southward from the Arctic regions to the southern stratosphere within 1 year. It is clear that this THO cannot mix southward together with the particulate debris whose appearance in the southern stratosphere occurred somewhat later. A path for Arctic THO which accompanies particulate debris equatorwards and then separates from the particulate debris at the equator by being caught up in a Brewer–Dobson type circulation (Brewer, 1949; Dobson, 1956), in which the particulate debris remains at low stratospheric altitudes because of sedimentation, must be rejected also as requiring too much time.

In considering other possibilities of southward mixing paths for the Soviet THO, it is helpful firstly to consider what situation resulted in the Northern Hemisphere after the 1961–62 Arctic testing. In Table 3 the yearly mean concentrations of THO in central European rainwater (Roether, 1967) and for Sr-90 in rainwater at Heidelberg, Germany (Schumann, 1967) are presented for the years 1962–66, together with the THO/Sr-90 ratio. The background activities from earlier testing and estimated small contributions from the American 1962 series have been subtracted, so that the values represent those due to the 1961–62 Soviet series only. The rise of the THO/Sr-90 ratio with time indicates that the mean stratospheric residence time is greater for THO than for Sr-90. (To a small extent the continental THO concentrations will be affected by the re-evaporation into the atmosphere of water of concentration of equal order of magnitude to the atmospheric concentration, which might mean that the concentrations of continental rainwaters might have some 'memory' of recent THO fallout. However it is clear from the THO data for Northern Hemisphere oceanic stations that a similar progress of rainwater concentrations relative to the 1962 level occurred in oceanic acreas as found for Central Europe. The general increase of THO/Sr-90 ratio indicated in Table 3 is fairly representative for the Northern Hemisphere.)

The history of the Northern Hemisphere Sr-90 fallout after 1961 is much as would be expected from the HASP stratospheric measurements, (Feely *et al.*, 1966). A good fit to the Sr-90 variation indicated by Table 3 is obtained by assigning uniform Sr-90 production per MT fission yield for the Soviet series of 1961–62 and applying the 10-month stratospheric half reseidence time found from the HASP data. This does not mean to say that very little of the Sr-90 from these tests was injected into regions which might be typical of longer stratospheric residence times, since the HASP survey suggested that the particulate products were affected by sedimentation after injection, so that their later distribution was at the altitude of settling rather than that of injection. The bulk of the particulate fallout has exhibited a 10-month stratospheric half-residence period and has been located in the lower stratosphere since a few weeks after injection, with no significant addition of material from higher regions.

Table 3. *THO and Sr-90 concentrations in central Europe, 1962–66, originating from the Soviet test series of 1961 and 1962*

Year	Mean THO concentration ($mc \cdot cm^{-1} km^{-2}$)	Mean Sr-90 concentration at Heidelberg ($\mu c \cdot cm^{-1} km^{-2}$)	THO/Sr-90 ratio	Ratio relative to 1962
1962	17.2	160	108	1.00
1963	69.9	395	177	1.64
1964	41.3	213	194	1.80
1965	19.6	65	301	2.69
1966	11.3	32	353	3.27

Thus the particulate product measurements do not suggest anything definite about the initial altitude distribution of the 1961–62 injections.

It might be expected that THO, as a gaseous product, should exhibit, in its fallout over the Northern Hemisphere, a somewhat longer stratospheric residence time than the particulate products, since Feely *et al.* (1966) have found a 16-month half-residence time for C-14. The history of the THO fallout in Table 3 is not easily explainable in terms of a single stratospheric residence time. The U.S.A.E.C. estimated fusion yields for the 1961 and 1962 Soviet test series were 94 and 120 MT respectively. If the THO production per MT fusion yield for these series was reasonably uniform, it would require to assign the greatest fraction of the 1961 THO a residence period of several years, and virtually all the 1962 THO a residence time of order 16 months, in order to obtain something like agreement with the progress of THO fallout of Table 3, in particular the 4-fold increase between 1962 and 1963. The injection of THO to great heights may have been the case for the two large yield devices of 1961 (Oct. 3, 25 MT total yield; Oct. 30, 55 MT total yield; combined fission yield, 8 MT). These two devices were particularly productive of neutron activation products such as Mn-54 and Fe-55 (Feely & Bazan, 1965; Feely *et al.*, 1966). These authors have reported that the Mn-54 vertical distribution during 1962 and 1963 showed no indication of significant debris originating from altitudes above 22 km in the northern stratosphere. However, as mentioned earlier, the Mn-54 distribution may have been affected by sedimentation, and one cannot say that the THO vertical distribution would resemble that of Mn-54.

Another possible explanation of the 4-fold rise of Northern Hemisphere THO fallout between 1962 and 1963 is that the 1961 THO production may have been very much less than the published fusion yields suggested. To suggest that the two large devices of 1961 may not have produced great quantities of THO is pure speculation, but the actual mechanisms of THO production in thermonuclear tests are not clearly understood, and it may be that such unusual devices differed in their THO production as compared to devices of more normal fusion/fission yield ratio. Since C-14 also exhibited approximately the same 1962/1963 tropospheric increase (Nydal, 1966; Olsson *et al.*, 1966; Munnich & Roether, 1967), its production appears to be proportional to that of THO in the 1961–62 series. It seems to be impossible on the evidence of available data to decide what the initial vertical distribution of the 1961 Soviet THO must have been. The progress of the Northern Hemisphere tropospheric THO concentrations after 1963 shows that the 1962 THO, and the bulk of the THO in the northern stratosphere was distributed in much the same way as found for C-14 (Feely *et al.*, 1966), with a 16-month stratospheric half-residence time, and the bulk of the activity in the lower stratosphere.

Turning to the amount of Soviet THO reaching the Southern Hemisphere, it can be estimated roughly that the total which will eventually be removed via the southern stratosphere is about 5 % of the amount removed to the northern troposphere. This would correspond to the production resulting from approximately 8–11 MT fusion yield, depending on the efficiency of THO production per MT fusion yield of the 1961 series. (The equivalent amount of Sr-90 is also about 5 %.)

The appearance of 1961 Soviet THO in the Southern Hemisphere during 1962–63 is unlikely to be due to a mesospheric source, since the Rh-102 data (Kalkstein, 1962) suggests the first downwards transport of mesospheric debris in the second spring season after injection. On the evidence of the Rocket Network wind data (Newell *et al.*, 1966), upper stratospheric mixing appears to be fast enough to allow pole-to-pole transport somewhat faster than the 18-month to 2-year time taken by the particulate products in the lower stratosphere. Southern Hemisphere THO fallout later than 1962–63 may be a combination of THO which has mixed southward at various altitudes from middle stratospheric to mesospheric. Indeed, the very long stratospheric residence time indicated by the 1965–66 and early 1966–67 Southern Hemisphere THO concentrations may even result from a reservoir created by transport into the mesosphere during mixing, at a time much later than that of injection.

In the Northern Hemisphere the seasonal variations of Sr-90 and THO fallout have been very similar. This does not necessarily imply that there is an asymmetry of behaviour between the two Hemispheres. The following situation could be responsible for the seasonal variation of the Northern Hemisphere fallout and yet be consistent with the sharp differences between the two types of fallout in the Southern Hemisphere. Nuclear debris creasted at 75° N during Sept.–Dec. could, according to the Meteorological Rocket Network data (Newell *et al.*, 1966), be subjected to intense mixing at all heights above the tropopause, the mixing extending to mid-latitudes where the equatorward mixing appears to slow down. In this way the debris could become stabilised as an umbrella of fairly uniform concentration centred over the North Pole and extending to mid-latitudes. During the period Jan.–April, the stratosphere over the North Pole may be regarded as being shut off from that at mid-latitudes by the formation of the stratospheric westerly circumpolar vortex. During this time the polar tropopause is generally absent, and the fraction of the fallout products caught north of the circumpolar vortex could be transferred downwards into the troposphere in the same manner as has been suggested above for THO over the south polar regions. Since the area over the polar cap is small compared to the total area of the umbrella extending to mid-latitudes, this transfer will result in a fallout peak smaller than that which occurs when the mid-latitude stratosphere-troposphere exchange is at a maximum level during April–July. Downwards transport from the stratosphere at latitudes greater than 60° N has already been suggested by Peirson (1961) to explain the latitude variation of Sr-90 concentration during 1959, a year following major Soviet testing in the Arctic. Concentrations averaged from January–June of this year peaked at mid-latitudes and increased strongly again above 60° N, with a minimum at about 50° N, suggesting downwards transport over the poles and at mid-latitudes at times during this period. During July–December, a flat maximum of lower concentration existed at mid-latitudes, with no suggestion of an increase towards the pole. Thus it may be that the effects of upper tropospheric mixing are not enough to prevent the detection of significant differences in the latitude distribution of fallout entering by separate paths into the troposphere, provided sufficient data are available to establish a reliable picture of latitudinal deposition.

For THO injected by testing in the equatorial North Pacific the evidence of the THO concentrations recorded in the southern troposphere prior to 1962–63 suggests that any THO created in the lower stratosphere ascends to greater altitudes and does not appear in the southern troposphere until the second spring season after injection. In the case of surface detonations, in which considerable quantities of tropospheric water are injected into the lower stratosphere, large quantities of THO may be removed by sedimentation fallout of ice crystals, as has been suggested by Begemann & Libby (1957) for the early fallout from the U.S. Castle test of 1954.

The behaviour of the THO/Sr-90 ratio in the Southern Hemisphere

The values of the mean annual Sr-90 concentrations in New Zealand rainwater (from Table 1), and the mean annual THO concentrations at Kaitoke are compared in Table 4. The mean Sr-90 concentrations are precipitation-weighted to erase the influence of high concentrations

Table 4. *Mean annual concentrations of Sr-90 in New Zealand, and THO at Kaitoke*

"Fallout year"	Mean Sr-90 concentration ($\mu c \cdot cm^{-1} km^{-2}$)	Mean THO concentration ($\mu c \cdot cm^{-1} km^{-2}$)	The THO/Sr-90 ratio THO/Sr-90 ratio
1955–56	9.9 ± 2.0	160 ± 32	16.2 ± 4.5
1956–57	9.4 ± 1.9	158.5 ± 7	16.9 ± 3.4
1957–58	7.4 ± 1.5	244 ± 10	33.0 ± 6.7
1958–59	8.8 ± 1.8	287 ± 50	32.6 ± 8.6
1959–60	7.1 ± 1.3	287 ± 50	40.4 ± 10.1
1960–61	8.7 ± 1.4	330 ± 10	37.9 ± 6.1
1961–62	9.4 ± 1.3	308 ± 6	32.8 ± 4.6
1962–63	14.7 ± 0.4	500 ± 10	34.0 ± 1.0
1963–64	19.4 ± 1.4	825 ± 17	42.5 ± 3.0
1964–65	34.2 ± 1.0	1220 ± 25	35.7 ± 1.2
1965–66	14.1 ± 0.3	1005 ± 20	71.3 ± 2.0

in months of low precipitation. The mean THO concentrations are unweighted, since the THO concentrations are essentially unaffected by precipitation. Weighting for THO shifts the mean in favour of months of large precipitation and does not give a reliable time mean.

Between 1959 and 1966 the Sr-90 error limits are the mean root square deviations from Table 1. Prior to 1959 an arbitrary limit of 20 % is applied to the U.K.A.E.A. Ohakea data. The limits for the THO data are wide in 1955–56, 1958–59 and 1959–60 because data for these periods are very sparse and the means are indirectly estimated.

The history of the THO/Sr-90 ratio is separated distinctly into three periods. In the first, covering 1955–56 and 1956–57, the ratio is below 20. In the second period covering 1957–58 until 1962–63 the ratio is roughly constant, between 30 and 40, any true variation being masked by the error limits. From 1963–64 the variations lie outside the error limits, and in 1965–66 the ratio rises sharply as the lower stratospheric particulate product reservoir becomes exhausted, while the THO reservoir starts to decline with much longer residence time.

A few general comments will now be made as to the behaviour of the THO/Sr-90 ratio over the remainder of the Southern Hemisphere. Near the equator there is a zone where the influence of fallout spreading from the Northern Hemisphere within the troposphere is considerable, so that a gradual transition from Southern to Northern Hemisphere conditions is expected. This zone will vary with season, but its influence should not extend to latitudes greater than 15° S. In the Antarctic regions the ratios will be very much higher than those of Table 4, since the THO fallout is of the same order of magnitude as at mid-latitudes, whereas the Sr-90 fallout is a factor of 10 lower. Between 40° S and 15° S a region exists where the Sr-90 and THO concentrations both decline, so that for oceanic areas the THO/Sr-90 ratios are probably very close to those of Table 4. For continental areas the THO concentrations will be enhanced, due to the absence of molecular exchange removal and to the reevaporation of freshly precipitated water into the atmosphere, the enhancement depending on distance from the coast, direction of the prevailing wind, and local climate. One therefore expects higher THO/Sr-90 ratios. A further uncertain effect of enhancement is that of altitude.

It is clear that the THO/Sr-90 ratio is not a reliable indicator for THO input in continental areas of the Southern Hemisphere, because of its time variation and uncertain degree of enhancement of THO concentration.

As a basis for the reconstruction of past THO deposition on the land surface throughout the Southern Hemisphere south of 15° S, the following relationship is of value. For two localities P and Q in "fallout years" i and j:

$$\left(\frac{\text{Mean annual THO in } P}{\text{Mean annual THO in } Q}\right)_i = \left(\frac{\text{Mean annual THO in } P}{\text{Mean annual THO in } Q}\right)_j.$$

To reconstruct past THO fallout in an unknown area, a reliable master station is required with a complete record of THO fallout. Sampling must then be undertaken in the unknown area for preferably two fallout years. This establishes the seasonal pattern in the unknown area, and a relationship between the master and unknown areas. Extrapolation to earlier years then becomes possible with a good degree of accuracy. At present there seems to be no alternative to tritium sampling unless more than one master station is available between which interpolation can be made. These

procedures may not be applied between 15° S and the equator because of the spillover of Northern Hemisphere fallout.

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REFERENCES

Albrecht, F. 1951. A. Monatskarten des Niederschlages im Indischen und Stillen Ozean. B. Monatskarten der Verdunstung und des Wasserhaushaltes des Indischen und Stillen Ozeans. *Ber. des Deutschen Wetterdienstes 29*, 39 pp.

Aldaz, L. 1965. Atmospheric ozone in Antarctica. *J. Geophys. Research 70*, 1767–1773.

Begemann, F. & Libby, W. F. 1957, Continental water balance, ground water inventory and storage times, surface ocean mixing rates and world-wide circulation patterns from cosmic-ray and bomb tritum. *Geochim. et Cosmochim. Acta 12*, 277–296.

Begemann, F. 1958, Tritium assays of natural waters. Final report, AF18(600)–569. University of Chicago, pp. 21–24.

Bolin, B. 1958, On the use of tritium as a tracer for water in nature. *Proc. 2nd U.N. Conf. Peaceful Uses of Atomic Energy 18*, 336–343.

Brewer, A. W. 1949. Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere. *Quart. J. Roy. Meteorol. Soc. 75*, 351–363.

Cambray, R. S., Fisher, E. M. R., Spicer, G. S., Wallace, C. G. & Webber, T. J., 1964, Radioactive fallout in air and rain: results to the middle of 1964. U.K.A.E.A., Harwell, Report AERE-R4687, 31 pp.

Court, A. 1942, Tropopause disappearance during the Antarctic winter. *Bull. Am. Meteorolog. Soc. 23*, 220–238.

Dobson, G. M. B. 1956, Origin and distribution of the polyatomic molecules in the atmosphere. *Proc. Roy. Soc. A 236*, 187–193.

Dyer, A. J. 1966, Artificial radio-activity, ozone and volcanic dust as atmospheric tracers in the Southern Hemisphere. *Tellus 18*, 416–419.

Ehhalt, D. 1967, Tritium and deuterium in vertical profiles of tropospheric water vapour. Internat. Union Geodesy and Geophysics, XIVth General Assembly, Committee on Problems of Geochemistry, Berne, Oct. 2 (unpublished).

Eriksson, E. 1965. An account of the major pulses of tritium and their effects in the atmosphere. *Tellus 17*, 118–130.

Eriksson, E. 1967. The atmospheric transport of tritium, "Isotope Techniques in the Hydrologic Cycle". American Geophysical Union, Geophysical Monograph Series No. 11, pp. 56–57.

Feely, H. W., Davidson, B., Friend, J. P., Lagomarsino, R. J. & Leo, M. W. M., 1963, The ninth quarterly report on Project Stardust. U.S. Defense Atomic Support Agency, Report DASA 1309.

Feely, H. W. & Bazan, F., 1965, Stratospheric distribution of nuclear debris in 1962, 1963 and 1964. Radioactive Fallout from Nuclear Weapons Tests, Proceedings of the 2nd Conference, Germantown, Maryland, Nov. 3–6, 1964.

Feely, H. W., Seitz, H., Lagomarsino, R. J. & Biscaye, P. E. 1966. Transport and fallout of stratospheric radioactive debris. *Tellus 18*, 316–328.

Friend, J. P., Feely, H. W., Krey, P. W., Spar, J. & Walton, A., 1961, The High Altitude Sampling Programme. U.S. Defense Atomic Support Agency, Report DASA 1300.

Gow, A. J. 1965. On the accumulation and seasonal stratification of snow at the South Pole. *J. Glaciology 5*, 467–477.

Grant-Taylor, T. & Taylor, C. B. 1967, Tritium hydrology in New Zealand, Isotopes in Hydrology. International Atomic Energy Agency, Vienna, pp. 381–400.

I.A.E.A. Tritium Lists, International Atomic Energy Agency, Vienna, Tritium concentration in rain, rivers and other water. Lists 1–6.

Junge, C. E. 1958. Atmospheric chemistry. *Advances in Geophysics 4*, 1–108.

Kalkstein, M. 1962. Rhodium-102 high altitude tracer experiment. *Science 137*, 645–652.

Lal, D. 1963. On the investigations of geophysical processes using cosmic ray produced radioactivity. Earth Science and Meteoritics, North-Holland Publishing Company, Amsterdam, pp. 115–142.

Libby, W. F. 1956. Radioactive strontium fallout. *Proc. National Acad. Sciences, U. S. 42*, 365–390.

Libby, W. F. 1959. Radioactive fallout, particularly from the Russian October series. *Proc. National Acad. Sciences, U.S. 45*, 959–976.

List, R. J., Salter, L. P. & Telegadas, K. 1966. Radioactive debris as a tracer for investigating stratospheric motions. *Tellus 18*, 345–353.

Lockhart, L. B., Patterson, R. L. & Saunders, A. W. 1966. Airborne radioactivity in Antarctica. *J. Geophys. Research 71*, 1985–1991.

Martell, E. A. 1966. The size distribution and interaction of radioactive and natural aerosols in the stratosphere. *Tellus 18*, pp. 486–498.

Münnich, K. O. & Roether, W. 1967. Transfer of bomb C-14 and tritium from the atmosphere to the ocean. Internal mixing of the ocean on

the basis of tritium and C-14 profiles. International Atomic Energy Agency (Vienna), Symposium on Radioactive Dating and Low-level Counting, Monaco, 2–10 March 1967, pp. 93–104.

Newell, R. E., Wallace, J. M. & Mahoney, J. R. 1966. The general circulation of the atmosphere and its effects on the movement of trace substances. Part 2. *Tellus 18*, 363–380.

Nydal, R. 1966. Variation in C-14 concentration in the atmosphere during the last several years. *Tellus 18*, 271–279.

Olsson, I.U., Karlen, I. & Stenberg, A. 1966. Radiocarbon variations in the atmosphere. *Tellus*, pp. 293–297.

Picciotto, E., de Maere, X. & Friedman, I. 1960. Isotopic composition and temperature of formation of Antarctic snows. *Nature 187*, 857–859.

Picciotto, E. & Wilgain, S., 1963, Fission products in Antarctic snow. A reference level for measuring accumulation. *J. Geophys. Research 68*, 5965–5972.

Pierson, D. H. 1961. Transfer of stratospheric fission products into the troposphere. *Nature 192*, 497–500.

Roether, W. 1967. Tritium in Wasserkreislauf, Doctoral Thesis. Univ. of Heidelberg, 67 pp.

Salter, L. P. 1965. Stratospheric radioactivity in the Southern Hemisphere from 1961 and 1962 weapon tests. Radioactive Fallout from Nuclear Weapons Tests. Proceedings of the 2nd Conference, Germantown, Maryland, Nov. 3–6, 1964, pp. 409–421.

Schumann, G. 1967. Personal communication.

Small, S. H. 1960. Wet and dry deposition of fallout materials at Kjeller. *Tellus 12*, 308–314.

Stebbins, A. K. 1961. Reports on the high altitude sampling program. U.S. Defense Atomic Support Agency, Report DASA 539.

Taylor, C. B. 1964. Tritium content of Antarctic snow. *Nature 201*, 146.

Taylor, C. B. 1966. Tritium in Southern Hemisphere precipitation, 1953–1964. *Tellus 18*, 105–131.

Thatcher, L. L. & Payne, B. R. 1965. The distribution of tritium in precipitation over continents and its significance to ground-water dating. Proc. 6th Internat. Conf. Radiocarbon and Tritium Dating, Pullman, Washington, June 7–11, 1965, U.S.A.E.C. CONF-650652, pp. 604–629.

Vickers, W. W. 1963. Geochemical dating techniques applied to Antarctic snow samples. General Assembly of Berkeley, Aug. 1963, Intern. Assoc. Sci. Hydrol., Publ. 61, pp. 199–215.

Volchok, H. L. 1965, Sr-90 Fallout in Antarctica, U.S.A.E.C. Health and Safety Laboratory, Report HASL-161, pp. 286–287.

von Buttlar, H. & Wiik, B. 1965. Enrichment of tritium by thermal diffusion and measurement of dated Antarctic snow samples. *Science 149*, 1371–1373.

Wilgain, S., Picciotto, E., de Brueck, W. 1965, Strontium-90 fallout in Antarctica. *J. Geophys. Research, 70*, 6023–6032.

СРАВНЕНИЕ ВЫПАДЕНИЙ ТРИТИЯ И СТРОНЦИЯ – 90 В ЮЖНОМ ПОЛУШАРИИ

Сравниваются данные, имеющиеся к июню 1966 г. о выпадении ТНО и Sr-90 в южном полушарии. Характер сезонных изменений ТНО (от июля до июня) в дождевой воде над океаном в средних широтах довольно хорошо воспроизводится от года к году, также как и пики в конце зимы — весной. Максимум совпадает с отсутствием температурной инверсии в полярной тропопаузе, причем предполагается, что выпадение основной массы ТНО в южном полушарии из стратосферы в тропосферу происходит над полярными областями. Для Sr-90 в дождевой воде в средних широтах модель сезонных изменений не так точно воспроизводится из года в год, но проявляет широкий летний максимум и зимний минимум. Представляется, что основная часть продуктов ядерных взрывов смешивается в пределах нижней стратосферы, распространяется к югу, и входит в тропосферу в умеренных широтах, причем сезонные изменения выпадения аналогичны сезонному изменению выпадения для северного полушария, но со сдвигом фазы на 6 месяцев. Анализ баланса ТНО в тропосфере приводит к соотношению, которое позволяет получить модель сезонного изменения для обмена ТНО между стратосферой и тропосферой, зная концентрации ТНО в дождевой воде. Стратосферные измерения продуктов распада при ядерных взрывах учитываются при оценке степени диффузии Sr-90 в южном направлении. Сделана также попытка проследить место возникновения, возможные пути диффузии, направленные к югу, для термоядерного ТНО, появляющегося в южном полушарии.

Поведение среднегодового отношения активностей ТНО/Sr-90 совершенно случайно и не является постоянным над всем южным полушарием для любого заданного момента времени.