

# Tritium in Southern Hemisphere precipitation 1953—1964

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## ABSTRACT

THO deposition in the Southern Hemisphere is discussed in the light of currently available tritium measurements of Southern Hemisphere precipitation. Many new results from the New Zealand laboratory are presented. Topics discussed include the seasonal and latitudinal variations of THO fallout, the fallout history of tropospheric and stratospheric thermonuclear THO, and the effects of the continental environment upon many of the reported measurements.

## Introduction

The fallout patterns of fission and fusion products from atomic and thermonuclear devices have been studied extensively in the years since the first thermonuclear device was detonated in 1952. Ground-based and airborne measurements of many radioactive isotopes generated by nuclear tests have served to increase the knowledge of the meteorological exchanges which take place between the troposphere and the stratosphere, and of the mixing and other meteorological and geophysical processes taking place in each of these regions. Because of the unenviable virtual monopoly of nuclear testing enjoyed by the Northern Hemisphere, by far the greater attention has been paid to its fallout patterns. The Southern Hemisphere patterns are less clear because of many fewer observations, although airborne measurements over the last six years have greatly increased the available data.

In this paper are gathered together for discussion all tritium measurements already reported for Southern Hemisphere precipitation, together with many previously unpublished results now available from this laboratory. The results are discussed with regard to their meteorological and hydrological implications.

## Factors of importance to the Southern Hemisphere THO fallout pattern

As an introduction to the interpretation of the data a number of relevant findings and observations are summarised below.

1. THO fallout is to be expected to occur as the result of the atmospheric testing of thermonuclear devices, in addition to the small levels present due to natural production (CRAIG & LAL, 1961; LAL & PETERS, 1962).
2. Thermonuclear detonations can be separated into distinct categories for the analysis of THO fallout. The important factors are the place of detonation, the type (i.e. surface, water or air), the season of year and the yield. The height to which the thermonuclear cloud penetrates is dependent on all these factors, and so also is the later history of the resulting THO fallout.
3. Tropospheric washout of THO in low and mid-latitudes occurs within days of its injection into the lower levels of the troposphere. In southern polar regions the precipitation is very seldom, so that tropospheric wash out should take longer to occur and tritium concentrations in Antarctic snow may be expected to be higher because of the small quantities of diluting tropospheric moisture. At low and mid-latitudes THO may spend, on the average, the greater part of its tropospheric residence time in the higher levels of the troposphere, before being drawn down to lower levels. The maximum mean tropospheric residence time must be set by the observations of the early decay of tritium from the Castle and Redwing Pacific tests measured in precipitation of the Northern Hemisphere (BROWN & GRUMMITT, 1956; BEGEMANN & LIBBY 1957; BEGEMANN, 1958 A;

- GILETTI *et al.*, 1958). This residence time is approximately 1–2 months, and must be considered as a maximum because it reflects also the fallout of a considerable amount of THO from the equatorial stratosphere, (see (5) below). According to JUNGE (1963) the mean residence time for water vapour in the lower troposphere varies with climatological conditions, and he quotes values ranging from 6 to 15 days according to latitude. Because of the considerable THO deposition occurring in oceanic areas by vapour exchange across the ocean surface these residence times, based on precipitation only, will be significantly in excess of the true lower tropospheric residence times for THO.
4. The possibility of exchange of THO across the equator within the troposphere must be envisaged in view of the latitude profiles obtained from measurements of other fallout products, (e.g. MACHTA, 1959; STEBBINS, 1961).
  5. After the formation of a localised quantity of thermonuclear THO its further history will become a meteorological process, but may be influenced in early stages by the presence of other debris formed by the same test. For instance, much of the THO formed in the stratosphere by the large Bravo test of the 1954 U.S. Pacific Castle series is believed to have been removed rapidly from the stratosphere as a constituent of ice crystals formed by large quantities of sea-water carried aloft in the bomb-cloud (BEGEMANN & LIBBY, 1957). Early fallout of THO may also occur by freezing of THO vapour upon individual and coagulated descending debris particles.
  6. Any thermonuclear THO retained for long periods in the stratosphere, and thereby subjected to the slow stratospheric mixing processes, may be expected to enter the troposphere preferentially in mid-latitudes, and with a maximum in the spring period, as has been observed for other fallout products. By this time longitudinal mixing within the stratosphere will be fairly complete. The eventual latitudinal distribution of such stratospheric thermonuclear THO fallout upon the earth's surface may be expected to be much the same as has been deduced by LAL & PETERS (1962) for stratospheric cosmic-ray THO.
  7. Particulate fallout products which remain for long periods in the stratosphere are those whose fall-times are long compared to mixing times in the stratosphere. Such long residence stratospheric debris may be expected to move in the same manner as gaseous fallout products, such as THO and  $C^{14}O_2$ , which are initially stabilised in the same region. These considerations make the stratospheric measurements and fallout estimates of particulate debris of vital interest to the tritium fallout pattern of the Southern Hemisphere. In fact the reports of the High Altitude Sampling Programme (STEBBINS, 1961), the quarterly reports on the fallout programme of the U.S.A.E.C. Health and Safety Laboratory, and the relevant Hearings before the Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the United States (1957, 1959, 1962 and 1963), have formed the greater part of the background against which the Southern Hemisphere tritium data have been appraised.

### Presentation of the data

The results are presented in tabulated form under a three column head (Table 1).

In the first column the collection stations are arranged in order of increasing latitude southwards from the equator. The types of sample are indicated and reference to the source of the data is made for all results not obtained at this laboratory. Appropriate brief comment on the results has been included for certain stations.

In the second column the dates of deposition of each sample have been noted, together with the precipitation over the collection period where known. Preliminary examination of the concentrations indicated that the mid-latitude THO fallout pattern is clearly centred around an annual period which will be termed the "fallout year". The commencement of each "fallout year" heralds a peak influx of stratospheric THO to the troposphere which may or may not contain a first influx from recent tests. The exact date of appearance of the spring maximum is probably variable from year to year, but the result for July 1963 at Kaitoke, Wellington (see Fig. 2), clearly apart from all preceding

TABLE 1

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                | Date and Precipitation<br>in mm |           | Tritium<br>Units |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|-----------|------------------|
| <i>Entebbe Airport, Uganda; 0° S, 32° E</i><br>Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | <i>1959-60</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | March                           | 1960, 306 | 8.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | April                           | 1960, 425 | 6.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | May                             | 1960, 86  | 5.9              |
| This station is not ideal for judging the movement of Northern Hemisphere tropospheric THO across the equator. Its situation is on the shores of Lake Victoria and the tritium concentrations may be affected by re-evaporated continental surface moisture from the Lake and surrounding country. Small variations cannot be considered to represent THO crossing the equator. However the sample for December, 1961, may contain THO from the U.S.S.R. Arctic tests of that year. The high concentrations during January-June, 1963, may be due to the southwards movement of THO from the exceedingly high Northern Hemisphere spring influx of that year, but some of these values appear to be very high in comparison with the concentrations measured at Dar es Salaam during the same period. | <i>1960-61</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | July                            | 1960, 49  | 8.9              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | August                          | 1960, 47  | 11.6             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | September                       | 1960, 44  | 14.5             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | October                         | 1960, 78  | 6.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | November                        | 1960, 62  | 9.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | January                         | 1961, 45  | 8.8              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | February                        | 1961, 86  | 20.8             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | March                           | 1961, 189 | 11.2             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | April                           | 1961, 253 | 13.2             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | May                             | 1961, 169 | 8.9              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | June                            | 1961, 73  | 11.9             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | <i>1961-62</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | July                            | 1961, 77  | 7.1              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | August                          | 1961, 103 | 12.1             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | September                       | 1961, 140 | 11.7             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | October                         | 1961, 255 | 7.8              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | November                        | 1961, 385 | 5.2              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | December                        | 1961, 210 | 34.4             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | <i>1962-63</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | September                       | 1962      | 7.4              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | January                         | 1963, 173 | 73.9             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | February                        | 1963, 177 | 108              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | March                           | 1963      | 148              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | April                           | 1963      | 33.6             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | May                             | 1963      | 94.4             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | June                            | 1963      | 292              |
| <i>Hollandia, Netherlands New Guinea; 2° S, 141° E</i><br>Monthly composite rainwater samples, reported by Institut für Technische Kernphysik der Technischen Hochschule, Darmstadt, in lists of tritium data published by the International Atomic Energy Agency, Vienna.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            | <i>1957-58</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | July                            | 1957, 135 | 8.9 ± 8.9        |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | August                          | 1957, 139 | 10.5 ± 3         |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | September                       | 1957, 214 | 7.4 ± 3          |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | April                           | 1958, 124 | 18.2 ± 18.2      |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | <i>1958-59</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | October                         | 1958, 152 | 24.4 ± 5.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | December                        | 1958, 360 | 21.5 ± 7.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | January                         | 1959, 455 | 22.3 ± 4.5       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | March                           | 1959, 588 | 22.1 ± 4.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | May                             | 1959, 222 | 12.4 ± 3.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | <i>1959-60</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | July                            | 1959, 96  | 35.9 ± 10.0      |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | November                        | 1959, 178 | 16.0 ± 4.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | December                        | 1959, 251 | 11.4 ± 3.0       |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | April                           | 1960, 163 | 10.4 ± 10.4      |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | May                             | 1960, 140 | 9.7 ± 4.0        |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | <i>1960-61</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | October                         | 1960, 64  | 5.0 ± 5.0        |

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                          | Date and Precipitation<br>in mm |            | Tritium<br>Units |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|------------|------------------|
| <i>Dar es Salaam; 7° S, 39° E</i>                                                                                                                                                                                                                                                                                               | <i>1959-60</i>                  |            |                  |
| Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna.                                                                                                                          | March                           | 1960, 304  | 21.6             |
|                                                                                                                                                                                                                                                                                                                                 | April                           | 1960       | 4.9              |
|                                                                                                                                                                                                                                                                                                                                 | May                             | 1960       | 8.7              |
|                                                                                                                                                                                                                                                                                                                                 | June                            | 1960       | 2.1              |
| The concentrations reported for July 1960, January 1961 and September 1961 are anomalously high. Penetration southwards of THO from the Northern Hemisphere during early 1963 appears to be insufficient to carry much further south than this latitude.                                                                        | <i>1960-61</i>                  |            |                  |
|                                                                                                                                                                                                                                                                                                                                 | July                            | 1960       | 36.3             |
|                                                                                                                                                                                                                                                                                                                                 | August                          | 1960       | 15.3             |
|                                                                                                                                                                                                                                                                                                                                 | October                         | 1960       | 2.5              |
|                                                                                                                                                                                                                                                                                                                                 | December                        | 1960       | 2.9              |
|                                                                                                                                                                                                                                                                                                                                 | January                         | 1961       | 38.4             |
|                                                                                                                                                                                                                                                                                                                                 | February                        | 1961       | 3.3              |
|                                                                                                                                                                                                                                                                                                                                 | May                             | 1961, 69   | 2.8              |
|                                                                                                                                                                                                                                                                                                                                 | June                            | 1961, 74.7 | 6.2              |
|                                                                                                                                                                                                                                                                                                                                 | <i>1961-62</i>                  |            |                  |
|                                                                                                                                                                                                                                                                                                                                 | July                            | 1961       | 4.5              |
|                                                                                                                                                                                                                                                                                                                                 | August                          | 1961       | 2.9              |
|                                                                                                                                                                                                                                                                                                                                 | September                       | 1961       | 43.2             |
| October                                                                                                                                                                                                                                                                                                                         | 1961, 282                       | 3.0        |                  |
| November                                                                                                                                                                                                                                                                                                                        | 1961, 415                       | 5.6        |                  |
| December                                                                                                                                                                                                                                                                                                                        | 1961, 308                       | 9.7        |                  |
| January                                                                                                                                                                                                                                                                                                                         | 1962, 83                        | 22.9       |                  |
| February                                                                                                                                                                                                                                                                                                                        | 1962, 47                        | 14.4       |                  |
| March                                                                                                                                                                                                                                                                                                                           | 1962, 154                       | 13.2       |                  |
| April                                                                                                                                                                                                                                                                                                                           | 1962, 313                       | 6.0        |                  |
| May                                                                                                                                                                                                                                                                                                                             | 1962, 54                        | 6.1        |                  |
| June                                                                                                                                                                                                                                                                                                                            | 1962, 8                         | 21.4       |                  |
| <i>1962-63</i>                                                                                                                                                                                                                                                                                                                  |                                 |            |                  |
| August                                                                                                                                                                                                                                                                                                                          | 1962, 95                        | 10.2       |                  |
| September                                                                                                                                                                                                                                                                                                                       | 1962                            | 23.2       |                  |
| October                                                                                                                                                                                                                                                                                                                         | 1962                            | 5.5        |                  |
| January                                                                                                                                                                                                                                                                                                                         | 1963                            | 20.9       |                  |
| February                                                                                                                                                                                                                                                                                                                        | 1963                            | 33.2       |                  |
| March                                                                                                                                                                                                                                                                                                                           | 1963                            | 23.1       |                  |
| April                                                                                                                                                                                                                                                                                                                           | 1963                            | 13.6       |                  |
| May                                                                                                                                                                                                                                                                                                                             | 1963                            | 10.9       |                  |
| June                                                                                                                                                                                                                                                                                                                            | 1963                            | 18.3       |                  |
| <i>1963-64</i>                                                                                                                                                                                                                                                                                                                  |                                 |            |                  |
| July                                                                                                                                                                                                                                                                                                                            | 1963                            | 16.5       |                  |
| <i>1958-59</i>                                                                                                                                                                                                                                                                                                                  |                                 |            |                  |
| <i>Recife, Brazil; 8° S, 35° W</i>                                                                                                                                                                                                                                                                                              | May 2                           | 1959       | 22 ± 1           |
| Individual rain.                                                                                                                                                                                                                                                                                                                |                                 |            |                  |
| LIRBY (1961).                                                                                                                                                                                                                                                                                                                   |                                 |            |                  |
| This result could indicate tropospheric exchange of THO across the equator from the Northern Hemisphere, but may be affected by the continental environment.                                                                                                                                                                    |                                 |            |                  |
| <i>Funafuti Atoll; 9° S, 179° E</i>                                                                                                                                                                                                                                                                                             | <i>1961-62</i>                  |            |                  |
| Composite rainwater samples.                                                                                                                                                                                                                                                                                                    | Approx. 1 week to               |            |                  |
| The increased concentrations measured during May, 1962, coincide with a marked southward drift of debris from the U.S. Christmas Island tests of that period (STALEY, 1963). Later variations during June-August probably indicate less marked transfer of THO across the equator from later tests of this series (see Fig. 6). | May 3, 1962,                    | 10         | 5.0 ± 0.3        |
|                                                                                                                                                                                                                                                                                                                                 | May 3-10, 1962,                 | 8          | 14.0 ± 1.0       |
|                                                                                                                                                                                                                                                                                                                                 | May 10-17, 1962,                | 22         | 13.5 ± 0.9       |
|                                                                                                                                                                                                                                                                                                                                 | May 17-24, 1962,                | 1          | 19.0 ± 1.3       |
|                                                                                                                                                                                                                                                                                                                                 | May 24-31, 1962,                | 12         | 6.7 ± 0.5        |
|                                                                                                                                                                                                                                                                                                                                 | May 31-June 7, 1962,            | 5          | 6.4 ± 0.5        |
|                                                                                                                                                                                                                                                                                                                                 | June 7-14, 1962,                | 100        | 8.1 ± 0.6        |
|                                                                                                                                                                                                                                                                                                                                 | June 14-22, 1962,               | 27         | 6.2 ± 0.4        |

| Source, Sample, Reference and Comments                                                                                                                                                                                  | Date and Precipitation<br>in mm     | Tritium<br>Units |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------|------------------|
|                                                                                                                                                                                                                         | June 22-29, 1962, 25                | $9.4 \pm 0.6$    |
|                                                                                                                                                                                                                         | June 29-July 19,<br>1962, 138       | $8.9 \pm 0.5$    |
|                                                                                                                                                                                                                         | 1962-63                             |                  |
|                                                                                                                                                                                                                         | July 19-Aug. 13,<br>1962, 224       | $9.7 \pm 0.6$    |
|                                                                                                                                                                                                                         | Aug. 13-27, 1962, 53                | $5.5 \pm 0.4$    |
|                                                                                                                                                                                                                         | Aug. 27-Sept. 17,<br>1962, 103      | $6.1 \pm 0.4$    |
|                                                                                                                                                                                                                         | Sept. 17-Oct. 16,<br>1962, 119      | $4.8 \pm 0.4$    |
|                                                                                                                                                                                                                         | Nov. 15-Dec. 27,<br>1962, 733       | $7.5 \pm 0.5$    |
|                                                                                                                                                                                                                         | Dec. 27, 1962-Feb. 12,<br>1963, 369 | $7.2 \pm 0.4$    |
| <i>Apia, Western Samoa; 14° S, 172° W</i>                                                                                                                                                                               | 1961-62                             |                  |
| Composite rainwater samples.                                                                                                                                                                                            | June 4-9, 1962                      | $5.7 \pm 0.5$    |
|                                                                                                                                                                                                                         | June 16-25, 1962                    | $4.3 \pm 0.5$    |
| Both these samples contain somewhat less tritium than those for the same period at Funafuti Atoll, indicating the lowering of concentrations southwards from the equator at low latitudes.                              |                                     |                  |
| <i>Lauhala Bay, Fiji; 18° S, 178° E</i>                                                                                                                                                                                 | 1957-58                             |                  |
| Composite rainwater samples taken at Meteorological Office.                                                                                                                                                             | Feb. 1-4, 1958, 4                   | $2.3 \pm 0.3$    |
|                                                                                                                                                                                                                         | Apr. 1-2, 1958, 13                  | $2.2 \pm 0.3$    |
| First five results quoted are measurements by this laboratory. Other results from LIBBY (1961).                                                                                                                         | June 1-3, 1958, 2                   | $2.7 \pm 0.3$    |
|                                                                                                                                                                                                                         | 1958-59                             |                  |
| The concentration for Aug. 1-3, 1958, may arise from southwards penetration of early THO fallout from the Hardtack Pacific test series. Higher concentrations were also experienced at Raoul Island during this period. | Aug. 1-3, 1958, 2                   | $11.1 \pm 1.2$   |
|                                                                                                                                                                                                                         | Oct. 1-3, 1958, 29                  | $4.1 \pm 0.4$    |
|                                                                                                                                                                                                                         | 1959-60                             |                  |
|                                                                                                                                                                                                                         | August 6, 1959                      | $1.5 \pm 0.6$    |
|                                                                                                                                                                                                                         | November 1959                       | $2.7 \pm 0.7$    |
|                                                                                                                                                                                                                         | January 1, 1960                     | $17.0 \pm 1.9$   |
|                                                                                                                                                                                                                         | April and May, 1960                 | $7.9 \pm 0.7$    |
| The concentrations reported for January 1, 1960, and April and May, 1960, are anomalously high.                                                                                                                         |                                     |                  |
|                                                                                                                                                                                                                         | 1960-61                             |                  |
|                                                                                                                                                                                                                         | August 11, 1960                     | $1.5 \pm 0.6$    |
| <i>Rarotonga, Cook Islands; 21° S, 160° W</i>                                                                                                                                                                           | 1957-58                             |                  |
| Composite rainwater samples taken at Meteorological Office.                                                                                                                                                             | Short period during                 |                  |
|                                                                                                                                                                                                                         | October 1957                        | $4.4 \pm 0.5$    |
|                                                                                                                                                                                                                         | January 1958                        | $2.1 \pm 1.0$    |
|                                                                                                                                                                                                                         | April 1958                          | $3.0 \pm 0.3$    |
|                                                                                                                                                                                                                         | July 1958                           | $3.0 \pm 0.4$    |
|                                                                                                                                                                                                                         | 1962-63                             |                  |
|                                                                                                                                                                                                                         | Oct.-Dec. 1962, 425                 | $9.9 \pm 0.6$    |
|                                                                                                                                                                                                                         | January 1963, 200                   | $8.1 \pm 0.6$    |
| <i>Lourenco Marques, Mozambique; 26° S, 33° E</i>                                                                                                                                                                       | 1959-60                             |                  |
| Rainwater.                                                                                                                                                                                                              | September 14, 1959                  | $1.5 \pm 0.6$    |
| LIBBY (1961).                                                                                                                                                                                                           |                                     |                  |
| This result is similar to that for August 6, 1959, at Fiji. This rain is probably entirely oceanic. The concentrations for Pretoria for the same period are clearly affected by continental environment.                |                                     |                  |
| <i>Pretoria, South Africa; 26° S, 28° E</i>                                                                                                                                                                             | 1957-58                             |                  |
| Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists                                                                                               | January 1958, 125                   | $9.4 \pm 0.5$    |
|                                                                                                                                                                                                                         | February 1958, 94                   | $10.0 \pm 0.5$   |
| Tellus XVIII (1966), 1                                                                                                                                                                                                  |                                     |                  |

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | Date and Precipitation<br>in mm |           | Tritium<br>Units |
|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|-----------|------------------|
| of tritium data published by the International Atomic Energy Agency, Vienna.                                                                                                                                                                                                                                                                                                                                                                                                                                                           | March                           | 1958, 84  | $13.1 \pm 0.5$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | April                           | 1958, 84  | $12.0 \pm 0.5$   |
| Pretoria represents a continental environment of considerable complication. It is likely that the effects of altitude, evaporation of falling raindrops, and reevaporation of surface moisture all contribute appreciably to the measured concentrations.                                                                                                                                                                                                                                                                              | May                             | 1958, 23  | $25.7 \pm 0.8$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1958-59</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | September                       | 1958, 36  | $68.4 \pm 2.0$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | November                        | 1958, 109 | $27.4 \pm 1.0$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | December                        | 1958, 125 | $20.4 \pm 0.8$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January                         | 1958, 127 | $21.4 \pm 0.8$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | February                        | 1959, 99  | $31.8 \pm 1.2$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | March                           | 1959, 46  | $13.2 \pm 0.5$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | April                           | 1959, 31  | $18.9 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | May                             | 1959, 20  | $19.8 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1959-60</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | July                            | 1959, 23  | $12.5 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | September                       | 1959, 9   | $37.4 \pm 1.1$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | October                         | 1959, 16  | $20.5 \pm 0.8$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | November                        | 1959, 109 | $17.4 \pm 0.8$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1961-62</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January                         | 1962, 91  | 16.4             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | February                        | 1962, 147 | 14.6             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | March                           | 1962, 72  | 16.1             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | April                           | 1962, 54  | 15.1             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1962-63</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | October                         | 1962, 36  | 42.7             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | November                        | 1962, 159 | 44.8             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | December                        | 1962, 57  | 62.3             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January                         | 1963, 145 | 37.1             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | February                        | 1963, 12  | 47.7             |
| <i>Raoul Island; 29° S, 178° W</i>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     | <i>1957-58</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January 8, 1958                 |           | $2.1 \pm 0.2$    |
| Rainwater collected at Meteorological Office.<br>The samples for July 2 and August 17, 1958, probably contain some early fallout from the Hardtack Pacific test series of 1958.                                                                                                                                                                                                                                                                                                                                                        | February 28, 1958               |           | $4.4 \pm 0.6$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | May 12, 1958                    |           | $3.5 \pm 0.4$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | July 2, 1958                    |           | $6.7 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1958-59</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | August 17, 1958                 |           | $6.8 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | October 4, 1958                 |           | $2.9 \pm 0.4$    |
| <i>Durban, South Africa; 30° S, 31° E</i>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              | <i>1958-59</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | July 3-12, 1959                 |           | $4.1 \pm 0.3$    |
| LIBBY (1961).<br>This coastal rain is significantly lower in tritium than those at Pretoria.                                                                                                                                                                                                                                                                                                                                                                                                                                           | <i>1957-58</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January 1958                    |           | $5.0 \pm 0.6$    |
| <i>Porto Alegre, Brazil; 30° S, 51° W</i>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              | February 1958                   |           | $3.2 \pm 0.4$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | Feb. 25-Mar. 27, 1958           |           | $4.0 \pm 0.3$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | Mar. 29-Apr. 26, 1958           |           | $6.3 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | June 1958                       |           | $17.0 \pm 0.9$   |
| Composite rainwater samples.<br>LIBBY (1961 and 1963). (On Prof. Libby's advice a result for November, 1958, has been ignored.)<br>The 1958 concentrations agree closely with those for other stations close to this latitude. The June, 1958, value may represent early THO fallout from the Hardtack Pacific test series. The high concentrations measured during 1959 are difficult to understand. Some of the concentrations reported by LIBBY (1961) are ignored when the data were reconsidered in a second paper (LIBBY, 1963.) | <i>1958-59</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | Aug. 1-Sept. 5, 1958            |           | $7.6 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | Sept. 6-30, 1958                |           | $6.9 \pm 0.5$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | Oct. 25-Dec. 2, 1958            |           | $5.8 \pm 0.4$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | December 1958                   |           | $8.5 \pm 0.3$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | January 1959                    |           | $13.0 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        | February 25, 1959               |           | $34.0 \pm 1.8$   |

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                            | Date and Precipitation<br>in mm | Tritium<br>Units |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|------------------|
|                                                                                                                                                                                                                                                                                                                                   | March 1959                      | $3.5 \pm 0.2$    |
|                                                                                                                                                                                                                                                                                                                                   | April 1959                      | 13.3             |
|                                                                                                                                                                                                                                                                                                                                   | May 1959                        | $15.0 \pm 1.7$   |
|                                                                                                                                                                                                                                                                                                                                   | 1959-60                         |                  |
|                                                                                                                                                                                                                                                                                                                                   | July 1959                       | $12.0 \pm 0.6$   |
|                                                                                                                                                                                                                                                                                                                                   | August 1959                     | $20.0 \pm 0.4$   |
|                                                                                                                                                                                                                                                                                                                                   | October 1959                    | $11.0 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                   | December 1959                   | $22.0 \pm 0.6$   |
|                                                                                                                                                                                                                                                                                                                                   | February 1960                   | $7.8 \pm 0.9$    |
| <i>Valparaiso, Chile; 33° S, 72° W</i>                                                                                                                                                                                                                                                                                            | 1953-54                         |                  |
| Rainwater.                                                                                                                                                                                                                                                                                                                        | April 4 or 5, 1954              | $4.3 \pm 0.2$    |
| VON BUTTLAR & LIBBY (1955).                                                                                                                                                                                                                                                                                                       |                                 |                  |
| <i>Malan Airport, South Africa; 34° S, 18.5° E</i>                                                                                                                                                                                                                                                                                | 1960-61                         |                  |
| Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna Vienna.                                                                                                                     | April 1961, 60                  | 16.2             |
|                                                                                                                                                                                                                                                                                                                                   | May 1961, 29                    | 8.8              |
|                                                                                                                                                                                                                                                                                                                                   | June 1961, 94                   | 10.4             |
|                                                                                                                                                                                                                                                                                                                                   | 1961-62                         |                  |
| Peak concentrations at this station appear to occur during the mid-summer months, as an extension of a spring increase which occurs simultaneously with that at Kaitoke. It seems probable that the evaporation of falling raindrops or admixture with continental moisture may contribute to these higher summer concentrations. | July 1961, 44                   | 8.2              |
|                                                                                                                                                                                                                                                                                                                                   | August 1961, 73                 | 5.4              |
|                                                                                                                                                                                                                                                                                                                                   | September 1961, 84              | 12.5             |
|                                                                                                                                                                                                                                                                                                                                   | October 1961, 21                | 14.3             |
|                                                                                                                                                                                                                                                                                                                                   | December 1961, 11               | 15.5             |
|                                                                                                                                                                                                                                                                                                                                   | January 1962, 5                 | 13.7             |
|                                                                                                                                                                                                                                                                                                                                   | February 1962, 40               | 25.9             |
|                                                                                                                                                                                                                                                                                                                                   | March 1962, 40                  | 4.3              |
|                                                                                                                                                                                                                                                                                                                                   | April 1962, 76                  | 8.8              |
|                                                                                                                                                                                                                                                                                                                                   | May 1962, 18                    | 9.9              |
|                                                                                                                                                                                                                                                                                                                                   | June 1962, 225                  | 5.1              |
|                                                                                                                                                                                                                                                                                                                                   | 1962-63                         |                  |
|                                                                                                                                                                                                                                                                                                                                   | July 1962, 65                   | 9.1              |
|                                                                                                                                                                                                                                                                                                                                   | August 1962, 89                 | 15.3             |
|                                                                                                                                                                                                                                                                                                                                   | September 1962, 28              | 8.2              |
|                                                                                                                                                                                                                                                                                                                                   | October 1962, 107               | 14.0             |
|                                                                                                                                                                                                                                                                                                                                   | November 1962, 20               | 11.0             |
|                                                                                                                                                                                                                                                                                                                                   | December 1962, 5                | 10.9             |
|                                                                                                                                                                                                                                                                                                                                   | January 1963, 7                 | 26.7             |
|                                                                                                                                                                                                                                                                                                                                   | February 1963, 2                | 19.4             |
|                                                                                                                                                                                                                                                                                                                                   | March 1963, 4                   | 9.2              |
| <i>Buenos Aires, Argentina; 35° S, 58° W</i>                                                                                                                                                                                                                                                                                      | 1953-59                         |                  |
| Rainwater.                                                                                                                                                                                                                                                                                                                        | April 1959                      | $17 \pm 1.3$     |
| LIBBY (1961).                                                                                                                                                                                                                                                                                                                     | 1959-60                         |                  |
| These concentrations are somewhat higher than those in New Zealand.                                                                                                                                                                                                                                                               | July 1959                       | 18               |
|                                                                                                                                                                                                                                                                                                                                   | November 1959                   | $22 \pm 2.4$     |
| <i>Kaitia, New Zealand; 35° S, 173° E</i>                                                                                                                                                                                                                                                                                         | 1962-63                         |                  |
| Monthly composite rainwater samples.                                                                                                                                                                                                                                                                                              | October 1962, 212               | $15.1 \pm 1.1$   |
|                                                                                                                                                                                                                                                                                                                                   | Nov. and Dec. 1962, 119         | $14.5 \pm 1.2$   |
|                                                                                                                                                                                                                                                                                                                                   | Jan. and Feb. 1963, 140         | $9.0 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                   | Mar. and Apr. 1963, 124         | $8.5 \pm 0.7$    |
| <i>Melbourne, Australia; 38° S, 145° E</i>                                                                                                                                                                                                                                                                                        | 1959-60                         |                  |
| Composite rainwater samples.                                                                                                                                                                                                                                                                                                      | January 1960, 51                | 13.9             |
|                                                                                                                                                                                                                                                                                                                                   | February 1960, 34               | 13.5             |
| Results for 1959-60 and 1960-61 reported by the Radioactive Tellus XVIII (1966), 1                                                                                                                                                                                                                                                | March 1960, 9                   | 21.5             |

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            | Date and Precipitation<br>in mm |           | Tritium<br>Units |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|-----------|------------------|
| Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna. Later results from this laboratory.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | April                           | 1960, 112 | 8.2              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | May                             | 1960, 92  | 9.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | June                            | 1960, 31  | 5.8              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1960-61</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | July                            | 1960, 92  | 11.2             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | August                          | 1960, 67  | 28.2             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | September                       | 1960, 92  | 32.3             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | October                         | 1960      | 20.9             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1961-62</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Jan.-Mar.                       | 1962, 100 | 17.8 $\pm$ 1.2   |
| Wairakei, New Zealand: 39° S, 176° E<br>Rainwater samples collected in waterbutt as part of tritium investigations of the geothermal area.<br><br>Mean concentration over period Aug. 15, 1960-Feb. 13, 1961 = 12.9 T.U. (5 samples).<br><br>Mean concentration at Kaitoke over the same period = 12.8 T.U. (7 samples).<br><br>This comparison shows that Wairakei concentrations are similar to those at Kaitoke, indicating that future Kaitoke rain concentrations could be used to indicate the level of tritium in rainwater at Wairakei.<br><br>Similar comparisons during 1961-62 and 1962-63 are not so fruitful, since sampling over such long periods as those imposed at Wairakei influences the concentrations towards the wet, and away from the dry, intervals of the total collection period. Regular sampling over short periods of about 1 month allows comparisons to be taken. Thus the mean concentration at Wairakei over the period Aug. 19, 1961 - July 16, 1962, was 10.8 T.U while that at Kaitoke was 9.5 T.U. Rainfall records at Wairakei suggest that more frequent sampling there over this period would have produced a lower mean concentration. | Apr. and May                    | 1962, 143 | 8.5 $\pm$ 0.7    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1962-63</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | June and July                   | 1962, 85  | 25.5 $\pm$ 1.8   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Aug. and Sept.                  | 1962, 84  | 26.6 $\pm$ 1.9   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Oct.-Dec.                       | 1962, 173 | 29.8 $\pm$ 2.1   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1959-60</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Sample taken Aug. 21,           | 1959      | 8.2 $\pm$ 0.9    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Aug. 21-Oct. 15,                | 1959      | 13.6 $\pm$ 1.5   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1960-61</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Aug. 15-Sept. 12,               | 1960      | 16.7 $\pm$ 1.8   |
| Gough Island; 40° S, 10° W<br>Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna.<br><br>These results compare well with those at Kaitoke and at Marion Island over the same period. The concentrations at these three stations are plotted in Fig. 7.<br><br>The value for January 1963 is anomalously high.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | Sept. 12-Oct. 10,               | 1960      | 22.9 $\pm$ 2.5   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Oct. 10-Nov. 21,                | 1960      | 9.9 $\pm$ 1.1    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Nov. 21, 1960-Jan. 16,          | 1961      | 11.2 $\pm$ 1.2   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Jan. 16-Feb. 13,                | 1961      | 6.7 $\pm$ 0.7    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Feb. 13-July 19,                | 1961      | 6.0 $\pm$ 0.4    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1961-62</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | July 19-Nov. 17,                | 1961      | 13.8 $\pm$ 1.0   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Nov. 17, 1961-July 16,          | 1962      | 9.7 $\pm$ 0.7    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1962-63</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | July 16-Oct. 5,                 | 1962      | 24.2 $\pm$ 1.7   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1963-64</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | Oct. 5, 1962-Sept. 25,          | 1963      | 21.8 $\pm$ 1.3   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1960-61</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | April                           | 1961      | 3.4              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | May                             | 1961      | 4.5              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | June                            | 1961      | 6.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <i>1961-62</i>                  |           |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | July                            | 1961      | 4.9              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | August                          | 1961      | 7.2              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | September                       | 1961      | 8.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | October                         | 1961      | 6.9              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | November                        | 1961      | 8.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | December                        | 1961      | 6.8              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | January                         | 1962      | 7.5              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | February                        | 1962      | 6.6              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | March                           | 1962      | 4.3              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | April                           | 1962      | 4.2              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | May                             | 1962      | 7.3              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | June                            | 1962      | 7.7              |



| Source, Sample, Reference and Comments                                                                                                                   | Date and Precipitation<br>in mm     | Tritium<br>Units |
|----------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------|------------------|
|                                                                                                                                                          | <i>1962-63</i>                      |                  |
|                                                                                                                                                          | July 1962                           | 10.6             |
|                                                                                                                                                          | August 1962                         | 15.9             |
|                                                                                                                                                          | September 1962                      | 15.0             |
|                                                                                                                                                          | November 1962                       | 11.9             |
|                                                                                                                                                          | December 1962                       | 14.4             |
|                                                                                                                                                          | January 1963                        | 31.2             |
|                                                                                                                                                          | February 1963                       | 11.6             |
|                                                                                                                                                          | March 1963                          | 8.6              |
| <i>Ohakea, New Zealand; 40° S, 174° E.</i>                                                                                                               | <i>1959-60</i>                      |                  |
| Composite rainwater samples.                                                                                                                             | Jan. 28-Mar. 31, 1960               | 9.1 ± 1.0        |
| LIBBY (1961).                                                                                                                                            | Apr. 1-July 1, 1960                 | 4.8 ± 0.4        |
| These results fit well into the pattern of New Zealand results obtained at this laboratory.                                                              |                                     |                  |
| <i>Near Wellington, New Zealand; 41° S, 175° E</i>                                                                                                       |                                     |                  |
| Rainwater.                                                                                                                                               | <i>1953-54</i>                      |                  |
| VON BUTTLAR & LIBBY (1955).                                                                                                                              | October 1, 1953                     | 1.7 ± 0.2        |
| Remaining samples collected at Kaitoke and measured at this laboratory. At Kaitoke the rainfall gauge is situated about 500 m from the sample collector. | <i>1959-60</i>                      |                  |
|                                                                                                                                                          | Approx. 1 month to<br>July 18, 1960 | 9.8 ± 1.1        |
| The mean concentrations (weighted by time intervals of collections), are as follows:                                                                     | <i>1960-61</i>                      |                  |
| 1960-61 10.15 T.U.                                                                                                                                       | Aug. 15-Sept. 21, 1960, 213         | 16.4 ± 1.8       |
| 1961-62 9.45 T.U.                                                                                                                                        | Sept. 21-Oct. 10, 1960, 36          | 17.2 ± 2.0       |
| 1962-63 15.4 T.U.                                                                                                                                        | Oct. 10-Nov. 14, 1960, 161          | 13.5 ± 1.5       |
| 1963-64 25 T.U. (expected value).                                                                                                                        | Nov. 14-Dec. 5, 1960, 139           | 9.4 ± 1.1        |
|                                                                                                                                                          | Dec. 5-Dec. 13, 1960, 114           | 5.7 ± 1.1        |
| 1957-58 7.7 T.U.                                                                                                                                         | Dec. 13, 1960-Jan. 10, 1961, 27     | 14.0 ± 1.6       |
| 1956-57 5.0 T.U.                                                                                                                                         | Jan. 10-Feb. 21, 1961, 129          | 9.0 ± 1.0        |
| (These last two means are deduced from Hutt River measurements.)                                                                                         | Feb. 21-Feb. 26, 1961, 0.5          | 6.2 ± 0.7        |
| 1959-60 Less than 1960-61 but greater than 1957-58.                                                                                                      | Feb. 26-Mar. 7, 1961, 71            | 5.6 ± 0.6        |
| (Deduced by comparison of Kaitoke and Wairakei concentrations.)                                                                                          | Mar. 7-Apr. 11, 1961, 167           | 8.0 ± 0.9        |
|                                                                                                                                                          | Apr. 11-May 10, 1961, 225           | 4.2 ± 0.5        |
|                                                                                                                                                          | June 2-July 19, 1961, 439           | 6.3 ± 0.4        |
|                                                                                                                                                          | <i>1961-62</i>                      |                  |
|                                                                                                                                                          | July 19-Aug. 23, 1961, 342          | 8.7 ± 0.6        |
|                                                                                                                                                          | Aug. 23-Sept. 19, 1961, 361         | 13.9 ± 1.0       |
|                                                                                                                                                          | Sept. 19-Oct. 10, 1961, 58          | 12.2 ± 0.8       |
|                                                                                                                                                          | Oct. 10-Nov. 15, 1961, 112          | 8.8 ± 0.6        |
|                                                                                                                                                          | Nov. 15-Dec. 18, 1961, 129          | 7.9 ± 0.6        |
|                                                                                                                                                          | Dec. 18, 1961-Feb. 1, 1962, 407     | 8.0 ± 0.6        |
|                                                                                                                                                          | Feb. 1-Mar. 15, 1962, 234           | 7.8 ± 0.6        |
|                                                                                                                                                          | Mar. 15-Apr. 6, 1962, 62            | 5.6 ± 0.4        |
|                                                                                                                                                          | Apr. 6-May 11, 1962, 324            | 9.5 ± 1.1        |
|                                                                                                                                                          | May 11-June 19, 1962, 229           | 13.1 ± 0.9       |
|                                                                                                                                                          | <i>1962-63</i>                      |                  |
|                                                                                                                                                          | July 12-Aug. 14, 1962, 252          | 11.2 ± 0.8       |
|                                                                                                                                                          | Aug. 14-Sept. 13, 1962, 219         | 15.8 ± 1.1       |
|                                                                                                                                                          | Sept. 13-Oct. 2, 1962, 60           | 18.4 ± 1.1       |
|                                                                                                                                                          | Oct. 2-Nov. 20, 1962, 374           | 19.9 ± 1.4       |
|                                                                                                                                                          | Nov. 20-Dec. 20, 1962, 107          | 20.6 ± 1.2       |
|                                                                                                                                                          | Dec. 20, 1962-Jan. 30, 1963, 124    | 18.1 ± 1.1       |
|                                                                                                                                                          | Jan. 30-Feb. 21, 1963, 100          | 10.6 ± 0.6       |

| Source, Sample, Reference and Comments                            | Date and Precipitation<br>in mm | Tritium<br>Units |
|-------------------------------------------------------------------|---------------------------------|------------------|
|                                                                   | Feb. 21–Apr. 3, 1963, 126       | $16.0 \pm 1.0$   |
|                                                                   | Apr. 3–May 2, 1963, 115         | $12.6 \pm 0.8$   |
|                                                                   | May 2–June 12, 1963, 341        | $11.2 \pm 0.7$   |
|                                                                   | June 12–July 1, 1963, 112       | $11.1 \pm 0.7$   |
|                                                                   | <i>1963–64</i>                  |                  |
|                                                                   | July 1963, 219                  | $26.0 \pm 1.6$   |
|                                                                   | Aug. 1–Sept. 2, 1963, 311       | $44.9 \pm 3.1$   |
|                                                                   | Sept. 2–Oct. 1, 1963, 187       | $33.6 \pm 2.0$   |
|                                                                   | October 1963, 30                | $35.6 \pm 2.5$   |
|                                                                   | November 1963, 254              | $21.3 \pm 1.3$   |
|                                                                   | December 1963, 128              | $16.9 \pm 1.0$   |
|                                                                   | <i>1964–65</i>                  |                  |
|                                                                   | July 1964, 367                  | $51.5 \pm 3.6$   |
|                                                                   | August 1964, 144                | $63.7 \pm 4.5$   |
| Riverwater from the upper reaches of the Hutt River near Kaitoke. | <i>1955–56</i>                  |                  |
|                                                                   | April 29, 1956                  | $3.7 \pm 0.4$    |
| Mean annual concentrations:                                       | <i>1956–57</i>                  |                  |
| 1956–57 = 4.8 T.U. $\pm$ 0.2                                      | September 15, 1956              | $4.0 \pm 0.4$    |
| 1957–58 = 7.3 T.U. $\pm$ 0.3                                      | October 14, 1956                | $6.6 \pm 0.7$    |
| 1960–61 = 8.8 T.U. $\pm$ 0.3                                      | November 5, 1956                | $5.9 \pm 0.5$    |
|                                                                   | November 30, 1956               | $3.8 \pm 0.5$    |
|                                                                   | January 4, 1957                 | $4.9 \pm 0.6$    |
|                                                                   | February 5, 1957                | $4.1 \pm 0.3$    |
|                                                                   | March 2, 1957                   | $5.3 \pm 0.6$    |
|                                                                   | April 2, 1957                   | $4.7 \pm 0.6$    |
|                                                                   | May 5, 1957                     | $7.9 \pm 0.9$    |
|                                                                   | June 10, 1957                   | $3.1 \pm 0.4$    |
|                                                                   | June 30, 1957                   | $3.9 \pm 0.4$    |
|                                                                   | <i>1957–58</i>                  |                  |
|                                                                   | August 4, 1957                  | $3.4 \pm 0.4$    |
|                                                                   | October 6, 1957                 | $7.5 \pm 0.8$    |
|                                                                   | November 4, 1957                | $9.4 \pm 1.1$    |
|                                                                   | December 10, 1957               | $7.6 \pm 0.8$    |
|                                                                   | January 6, 1958                 | $11.9 \pm 1.3$   |
|                                                                   | February 13, 1958               | $6.1 \pm 0.7$    |
|                                                                   | April 8, 1958                   | $7.5 \pm 0.8$    |
|                                                                   | June 14, 1958                   | $6.6 \pm 0.7$    |
|                                                                   | July 13, 1958                   | $6.1 \pm 0.7$    |
|                                                                   | <i>1958–59</i>                  |                  |
|                                                                   | September 7, 1958               | $7.5 \pm 0.8$    |
|                                                                   | <i>1959–60</i>                  |                  |
|                                                                   | April 21, 1960                  | $6.6 \pm 0.7$    |
|                                                                   | May 17, 1960                    | $7.1 \pm 0.7$    |
|                                                                   | June 28, 1960                   | $6.2 \pm 0.7$    |
|                                                                   | July 18, 1960                   | $7.2 \pm 0.5$    |
|                                                                   | <i>1960–61</i>                  |                  |
|                                                                   | August 15, 1960                 | $8.7 \pm 1.0$    |
|                                                                   | September 21, 1960              | $15.0 \pm 1.6$   |
|                                                                   | November 14, 1960               | $7.8 \pm 0.8$    |
|                                                                   | December 5, 1960                | $6.8 \pm 1.3$    |
|                                                                   | January 10, 1961                | $7.1 \pm 0.8$    |
|                                                                   | February 21, 1961               | $11.4 \pm 1.2$   |
|                                                                   | March 7, 1961                   | $8.2 \pm 0.9$    |
|                                                                   | April 11, 1961                  | $10.6 \pm 1.1$   |
|                                                                   | May 10, 1961                    | $6.6 \pm 0.7$    |
|                                                                   | June 2, 1961                    | $6.1 \pm 0.7$    |

| Source, Sample, Reference and Comments                                                                                                                                                                                                                                                                                                                                                                                         | Date and Precipitation<br>in mm | Tritium<br>Units |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|------------------|
|                                                                                                                                                                                                                                                                                                                                                                                                                                | <i>1961-62</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | August 23, 1961                 | $7.4 \pm 0.6$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | September 19, 1961              | $10.0 \pm 0.7$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | October 10, 1961                | $8.2 \pm 0.6$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | November 15, 1961               | $7.5 \pm 0.5$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | December 18, 1961               | $9.4 \pm 0.7$    |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | <i>1962-63</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | September 13, 1962              | $18.8 \pm 2.5$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | July 1, 1963                    | $11.3 \pm 0.5$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | <i>1963-64</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | January 1, 1964                 | $20.1 \pm 1.2$   |
| <i>Tasman Glacier, Mt. Cook, New Zealand; 44° S, 170° E</i>                                                                                                                                                                                                                                                                                                                                                                    |                                 |                  |
| Snow; taken at 900 m altitude and estimated to be at least 50 years old. If perfect storage conditions existed for 50 years, then this sample contained $16.8 \pm 3.2$ T.U. at the time of deposition. Since this concentration seems high by comparison with more modern post-thermonuclear snows and with the pre-thermonuclear estimate, contamination of the sample, as with the South Pole snow below, is thought likely. | Collected 1959                  | $1.1 \pm 0.2$    |
| Mt. Cook surface snows.                                                                                                                                                                                                                                                                                                                                                                                                        | <i>1958-59</i>                  |                  |
| TAYLOR (1964).                                                                                                                                                                                                                                                                                                                                                                                                                 | Summer layer at 1500 m.         | $10.1 \pm 1.1$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | June, 1959, at 2100 m.          | $17.3 \pm 1.9$   |
| An altitude effect is apparent in these samples by comparison with sea level rainfall at the same times.                                                                                                                                                                                                                                                                                                                       | <i>1960-61</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | Apr. 21, 1961, at 2100 m.       | $35.8 \pm 3.9$   |
| <i>Invercargill, New Zealand; 46° S, 168° E</i>                                                                                                                                                                                                                                                                                                                                                                                | <i>1962-63</i>                  |                  |
| Composite rainwater samples.                                                                                                                                                                                                                                                                                                                                                                                                   | October-December                |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | 1962, 245                       | $19.6 \pm 1.2$   |
| Mean concentration Oct. 1962-June 1963 = 18.0 T.U.                                                                                                                                                                                                                                                                                                                                                                             | January-March 1963, 274         | $15.2 \pm 0.9$   |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | April-June 1963, 296            | $19.2 \pm 1.2$   |
| Mean concentration at Kaitoke over same period = 15.7 T.U.                                                                                                                                                                                                                                                                                                                                                                     |                                 |                  |
| Whether this concentration difference between Kaitoke and Invercargill is significant in terms of latitude variation of tritium fallout will only become clear after further measurements.                                                                                                                                                                                                                                     |                                 |                  |
| <i>Marion Island; 47° S, 38° E</i>                                                                                                                                                                                                                                                                                                                                                                                             | <i>1960-61</i>                  |                  |
| Monthly composite rainwater samples, reported by the Radioactive Dating Laboratory, Geological Survey of Sweden, in lists of tritium data published by the International Atomic Energy Agency, Vienna.                                                                                                                                                                                                                         | April 1961, 189                 | 4.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | May 1961, 100                   | 5.6              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | June 1961, 460                  | 4.6              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | <i>1961-62</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | July 1961, 178                  | 4.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | August 1961, 232                | 16.6             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | September 1961, 174             | 14.1             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | October 1961, 181               | 9.0              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | November 1961, 88               | 8.4              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | December 1961, 260              | 7.4              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | January 1962, 272               | 10.7             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | February 1962, 262              | 7.5              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | March 1962, 195                 | 7.6              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | June 1962, 14                   | 8.7              |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | <i>1962-63</i>                  |                  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | July 1962, 308                  | 17.5             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | August 1962, 178                | 12.5             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | September 1962, 149             | 21.2             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                | October 1962, 146               | 17.5             |

| Source, Sample, Reference and Comments                | Date and Precipitation<br>in mm  | Tritium<br>Units |
|-------------------------------------------------------|----------------------------------|------------------|
|                                                       | November 1962, 720               | 20.1             |
|                                                       | December 1962, 310               | 16.8             |
|                                                       | January 1963, 263                | 19.0             |
|                                                       | February 1963, 251               | 20.0             |
| <i>Campbell Island; 52° S, 169° E</i>                 | 1956-57                          |                  |
| Rainwater samples.                                    | July 4, 1957                     | $2.0 \pm 0.2$    |
|                                                       | 1957-58                          |                  |
|                                                       | November 4, 1957                 | $4.7 \pm 0.5$    |
|                                                       | July 1, 1958                     | $12.5 \pm 1.4$   |
|                                                       | 1958-59                          |                  |
|                                                       | September 2, 1958                | $9.8 \pm 1.1$    |
|                                                       | September 2, 1958 (snow<br>melt) | $10.0 \pm 1.1$   |
|                                                       | November 1, 1958                 | $10.5 \pm 1.1$   |
| ANTARCTIC SNOWS                                       |                                  |                  |
| <i>Ross Ice Shelf near Ross Island; 78° S, 170° E</i> | 1941 layer                       | $17 \pm 3$       |
| (Collected Jan. 15 and 17, 1955.)                     | 1960-61                          |                  |
| SHEN <i>et al.</i> (1963).                            | September-December 1960          | $82 \pm 8$       |
| <i>Little America III; 78° S, 165° W</i>              | 0-8 in.                          | $7.5 \pm 0.6$    |
| Collected Jan. 15 and 17, 1955.                       | 0-8 in.                          | $14.1 \pm 1.0$   |
| BEGEMANN & LIBBY (1957).                              | 1-2 ft. depth                    | $28.0 \pm 1.5$   |
|                                                       | 2-3 ft. depth                    | $13.0 \pm 1.5$   |
| <i>Admiral Byrd Bay; 70° S, 0° W</i>                  | 1 st ft.                         | $24.0 \pm 0.4$   |
| Collected February 19, 1955.                          | 2nd ft.                          | $12.5 \pm 0.8$   |
| BEGEMANN & LIBBY (1957).                              | 3 rd ft.                         | $13.5 \pm 0.7$   |
|                                                       | 5 th ft.                         | $9.6 \pm 1.0$    |
|                                                       | 6 th ft.                         | $5.2 \pm 1.0$    |
| <i>Atka Bay; 71° S, 8° W</i>                          | 0-8 in.                          | $19.2 \pm 0.8$   |
| Collected February 20, 1955.                          |                                  |                  |
| BEGEMANN & LIBBY (1957).                              |                                  |                  |
| Collected March 29, 1956.                             | 0- 6 in.                         | $15.8 \pm 1.0$   |
| BEGEMANN (1958 B).                                    | 6-12 in.                         | $13.0 \pm 2.0$   |
|                                                       | 12-18 in.                        | $18.2 \pm 2.0$   |
|                                                       | 18-24 in.                        | $20.7 \pm 1.5$   |
|                                                       | 24-30 in.                        | $21.0 \pm 1.5$   |
| <i>Mc Murdo Sound, 78°S, 167°E</i>                    | 0- 5 in.                         | $19.6 \pm 1.5$   |
| Collected March 4, 1956.                              | 5-11 in.                         | $22.5 \pm 1.5$   |
| BEGEMANN (1958 B).                                    | 11-14 in.                        | $11.5 \pm 1.0$   |
|                                                       | 17-23 in.                        | $15.4 \pm 1.0$   |
|                                                       | 23-29 in.                        | $17.5 \pm 1.0$   |
|                                                       | 29-35 in.                        | $13.5 \pm 1.0$   |
|                                                       | 35-41 in.                        | $18.8 \pm 2.0$   |
|                                                       | 41-47 in.                        | $19.5 \pm 1.5$   |
|                                                       | 47-53 in.                        | $20.5 \pm 1.5$   |
|                                                       | 53-59 in.                        | $16.0 \pm 1.5$   |
| <i>Kainan Bay, 78°S, 162.5°W</i>                      | 0- 6 in.                         | $13.1 \pm 1.0$   |
| Collected January 1, 1956.                            | 6-12 in.                         | $15.5 \pm 1.0$   |
| BEGEMANN (1958 B).                                    | 12-18 in.                        | $17.8 \pm 1.5$   |
|                                                       | 18-24 in.                        | $19.5 \pm 1.5$   |
|                                                       | 24-30 in.                        | $18.7 \pm 1.5$   |
|                                                       | 30-36 in.                        | $11.5 \pm 1.0$   |
|                                                       | 36-42 in.                        | $17.2 \pm 2.0$   |
|                                                       | 42-48 in.                        | $14.4 \pm 1.0$   |
|                                                       | 48-54 in.                        | $11.0 \pm 1.5$   |
|                                                       | 54-60 in.                        | $12.2 \pm 1.0$   |

| Source, Sample, Reference and Comments | Date and Precipitation<br>in mm | Tritium<br>Units |
|----------------------------------------|---------------------------------|------------------|
| <i>Byrd Station; 80° S, 120° W</i>     | top 3 in. January 1964          | 112 $\pm$ 7      |
| <i>South Pole; 90° S</i>               |                                 |                  |
| Snow mine sample. Taken January 1964.  | 45 ft. depth                    | 40.5 $\pm$ 2.4   |
| Surface snow.                          | January 1964                    | 200 $\pm$ 12     |

Further collections were made in November, 1964, to endeavour to find the true nature and cause of thermonuclear contamination in the deeper layers of this snow mine.

samples, and signalling the approach of the still larger concentrations for August, September and October of that year, demands that the commencement of the "fallout year" be taken within that month. The disposition of the collection periods with which this paper deals has influenced the choice of July 19 as being the date of commencement of the "fallout year". Thus samples collected between July 19, 1960, and July 18, 1961, are tabulated together under the "fallout year" 1960-61. Where the collection period for a sample straddles the date July 19, that result has been included amongst the data for the later "fallout year".

In the case of certain samples measured at this laboratory it was necessary to mix together separate consecutive rainwater samples before processing, because of insufficiency of individual sample volume. In most of such cases the samples were mixed together in exact proportion to the recorded precipitations. In a few cases the proportions were approximate.

The measured tritium concentrations are listed in the third column. 1 tritium unit (1 T.U.) represents 1 atom of tritium per  $10^{18}$  atoms of protium.

The blank at this laboratory is less than 0.1 T.U. All errors quoted for measurements at this laboratory represent 1 standard deviation over the entire process of electrolytical enrichment followed by conversion to hydrogen and Geiger counting (TAYLOR *et al.*, 1963; TAYLOR, 1963). Errors for other measurements are given as they appear in the sources of the data.

Acute difficulty has been experienced with the calibration of this laboratory's tritium standard. Two successive calibrations by the supplier of the original standard used have proved to be 54 % and 27 % too high respectively, and the correct calibration has only recently been confirmed by two reliable overseas laboratories. This means that the absolute concentrations

listed in two previous papers (BAINBRIDGE & O'BRIEN, 1962; TAYLOR, 1964), are not accurate. The conclusions expressed in these papers remain substantially unaltered however.

This laboratory has recently acquired a vial of NBS tritium standard No. 4926 (MANN *et al.*, 1964), and two diluted standards supplied by the International Atomic Energy Agency, Vienna.

The results from this laboratory presented here are based on the agreeing activities between separate dilutions of NBS No. 4926 performed at Vienna and this laboratory.

### The latitude variation of Southern Hemisphere THO fallout

The latitude profile for cosmic-ray THO fallout has been estimated by LAL & PETERS (1962), and is shown in Fig. 1. The magnitude of the fallout is based on theoretical calculations of stratospheric and tropospheric tritium production (CRAIG & LAL, 1961), while the form of the profile they deduced from fallout profiles for fission products such as those reported by MACHTA (1959). This profile of Lal and Peters is for the case of rainwater diluted in THO by admixture with oceanic moisture of negligible tritium concentration only. An annual pattern of concentration variations will be imposed on this average profile because of seasonal variations in stratospheric-tropospheric exchange. Year by year changes should follow the cosmic-ray cycle.

For thermonuclear THO, the fallout will not remain the same year by year. At any time the THO fallout at a given location may be divided into two components; early fallout and delayed (or stratospheric) fallout. For this purpose in this paper the early component is considered to include all THO deposited within the pre-

TABLE 2. *Latitude variation of fallout, Southern Hemisphere, 1957-58.*

| Station         | $\sin \lambda$ | Estimated average T. U. | Rainfall (mm)     | Tritium fallout (atoms/cm <sup>2</sup> sec) |
|-----------------|----------------|-------------------------|-------------------|---------------------------------------------|
| Campbell Island | 0.84           | 9.0                     | 1630              | 3.1                                         |
| Wellington      | 0.71           | 7.7                     | 1600 <sup>a</sup> | 2.9                                         |
| Raoul Island    | 0.52           | 3.4                     | 1030              | 0.75                                        |
| Fiji            | 0.375          | 2.5                     | 2480              | 1.25                                        |
| Rarotonga       | 0.36           | 3.1                     | 2025              | 1.3                                         |
| Hollandia       | 0.1            | 15.0                    | 2000 <sup>b</sup> | 6.4                                         |

<sup>a</sup> Mean value for New Zealand annual rainfall.<sup>b</sup> Estimated approximate value.

viously mentioned 1-2 month fallout period for thermonuclear tritium, which will generally include some fallout from the low stratosphere. The delayed or stratospheric component is considered to be that fallout, delayed in time by months or years within the stratosphere, which penetrates to the troposphere preferentially at mid-latitudes after mixing within the stratosphere. Thus the stratospheric components of THO fallout at any location in the Southern Hemisphere can comprise not only downwards injections from the southern stratosphere at mid-latitudes, but also a component from sim-

ilar injections at northern mid-latitudes which penetrates across the equator within the troposphere before reaching the earth's surface.

An opportunity exists to investigate the latitude profile for stratospheric fallout in the Southern Hemisphere in the "fallout year" 1957-58 for which tritium measurements exist at Campbell Island, Wellington, Raoul Island, Fiji, Rarotonga and Hollandia. All these stations of widely spread latitude lie within a narrow longitude belt and may be considered to receive rainfall diluted with oceanic moisture of tritium concentration close to 1 T.U. in those years, with the possible exception of Hollandia whose rainfall may contain small quantities of reevaporated surface moisture. Unfortunately the measurements at each station are only very few and the average tritium concentrations over 1957-58 can only be estimated roughly. The measured concentrations which appear to be due in part to early fallout from 1958 Pacific thermonuclear tests have been ignored, so that any penetration of THO from the Northern Hemisphere may be considered to approximate to that originating as delayed stratospheric THO fallout.

The tritium fallout estimates for the six stations are listed in Table 2, and the resulting latitude profile is shown as the upper curve of Fig. 1. It should be realised that this curve represents the sum total of delayed fallout of stratospheric thermonuclear THO and total cosmic-ray THO fallout. Some idea of the stratospheric thermonuclear fallout may be obtained from a subtraction of the lower from the upper curve of Fig. 1, although this is not an accurate assessment in view of the approximate nature of both curves.

Because of the approximate nature of the

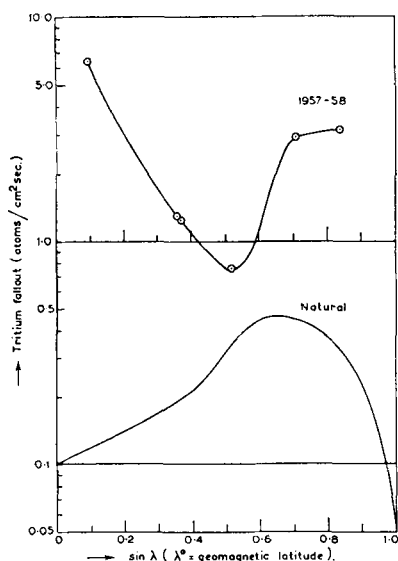


FIG. 1. Latitude variation of THO precipitation fallout in the Southern Hemisphere. Lower curve is total cosmic ray fallout deduced by LAL & PETERS (1962). Upper curve represents sum of delayed stratospheric fallout and total cosmic-ray fallout of THO for 1957-58.

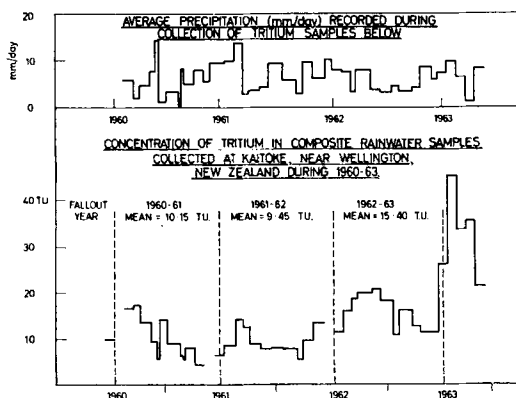


FIG. 2. Average precipitation and tritium concentration of rainwater samples at Kaitoke, Wellington, 1960-63.

1957-58 tritium fallout data, the upper curve of Fig. 1 cannot be used to give an exact division into northern and southern stratospheric components, as has been accomplished by MACHTA (1959) for  $\text{Sr}^{90}$  deposition. However the profile is of importance in that it demonstrates that mid-latitude concentrations of tritium in rainwater arise predominantly from fallout from the southern stratosphere, and may consequently be used to reach conclusions on such fallout. Results from later years also support this finding. The effectiveness of the intertropical convergence zone as a barrier preventing Northern Hemisphere THO of stratospheric origin from reaching southern mid-latitudes by trans-equatorial penetration within the troposphere is also shown. The two effects of the downwards flux of THO across the air-ocean interface, and of the finite concentrations of the ocean surface layers are not relevant to this conclusion. Their importance to the latitude variation of THO fallout will be discussed in a subsequent paper.

## Tritium fallout from the Southern Stratosphere

### a) THERMONUCLEAR THO IN MID-LATITUDE RAINWATERS AFTER THE MORATORIUM

Minimum South Pacific concentrations of tritium in oceanic rainwaters appear to occur about the latitude of Fiji, so that if one considers the steep fall-off of southern stratospheric THO fallout occurring between New Zealand and Raoul Island (Fig. 1), it is apparent that

the contribution of Northern Hemisphere THO, penetrating across the equator within the troposphere, to rainwater concentrations measured in New Zealand is quite negligible compared with THO falling out from the southern stratosphere.

In the pattern of increasing THO fallout measured at Kaitoke, the dominant features are the yearly spring peak of rainwater tritium concentration and the large increase in concentrations recorded at the beginning of the "fallout year" 1963-64 (Fig. 2). From the data of this paper it has been found to be possible to relate the observed increases to the history of thermonuclear testing. Fig. 3 (after MACHTA, 1962) shows zonal divisions of the stratosphere which may be used to predict the fallout of stratospheric nuclear debris. Broadly speaking, the fallout history of long-residence debris injected into each zone is typical of that zone only, and has been roughly delineated by worldwide fallout measurements. In Table 3 an attempt has been made to estimate the fusion energy of thermonuclear clouds injected into the various stratospheric zones of Fig. 3. For these estimates the fusion energy has been assumed to be distributed uniformly within, and in the immediate vicinity of, the thermonuclear clouds, which have dimensions varying according to the estimates of MACHTA *et al.* (1962). The test yields are as given in the most recent U.S.A.E.C. releases. In cases where the estimated fusion yields have not been specifically released, the ratio fusion yield/fission yield has been assumed to be 2 for U.S.S.R. tests and 1 for U.S.A. and U.K. tests. Included in the 1957-58 equatorial upper stratosphere estimates are the yields from the Teak and Orange tests detonated at 77 km

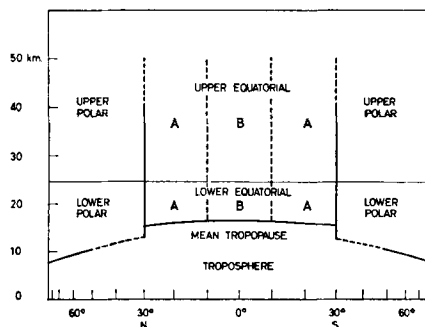


FIG. 3. Stratospheric zones of injection for prediction of history of stratospheric THO fallout, after MACHTA, (1962).

TABLE 3. *Estimated fusion yields of thermonuclear devices, 1952-62.*

| U.S.A. and U.K.    |                                        |                                | U.S.S.R.                                 |                                        |            |
|--------------------|----------------------------------------|--------------------------------|------------------------------------------|----------------------------------------|------------|
| Region             | Fusion yield (MT)<br>into stratosphere |                                | Region                                   | Fusion yield (MT)<br>into stratosphere |            |
|                    | Lower                                  | Upper                          |                                          | Lower                                  | Upper      |
| 1952-54            |                                        |                                |                                          |                                        |            |
| North Equatorial A | 20 approx.                             | 3 approx.<br>(mainly<br>Bravo) | Polar (?)                                | Not more<br>than 1                     | —          |
| 1955-56            |                                        |                                |                                          |                                        |            |
| North Equatorial A | 8 approx.<br>(Cherokee)                | 2 approx.<br>(Cherokee)        | Polar                                    | 3 approx.                              | Negligible |
| 1957               |                                        |                                |                                          |                                        |            |
| North Equatorial B | 3 approx.                              | Probably<br>negligible         | Polar (about<br>10 tests in<br>MT range) | 30 approx.                             | 3 approx.  |
| 1958               |                                        |                                |                                          |                                        |            |
| North Equatorial A | Less than 5                            | 2 approx.                      |                                          |                                        |            |
| North Equatorial B | 4 approx.                              | Less than 0.7                  |                                          |                                        |            |
| 1961               |                                        |                                |                                          |                                        |            |
| —                  | —                                      | —                              | Polar                                    | 64                                     | 30         |
| 1962               |                                        |                                |                                          |                                        |            |
| North Equatorial A | 2.7                                    | 0.3                            | Polar                                    | 60                                     | 60         |
| North Equatorial B | 6.3                                    | 0.7                            |                                          |                                        |            |

and 30 km respectively. The 1962 U.S.A. estimates do not include the yield of the Starfish high altitude test (1.4 MT fission yield) of July 9, detonated at about 300 km altitude, but the upper stratospheric yields almost certainly include some THO that is injected at mesospheric levels.

An attempt has been made to estimate the amounts of fusion yield from equatorial tests which are effective for the mixing of thermonuclear THO in a southwards direction within the stratosphere. Stratospheric concentrations of the tracer isotope  $W^{185}$  injected into the north equatorial A region of the lower stratosphere during the U.S.A. Pacific test series of 1958 indicated that the amounts of fallout mixing northwards and southwards within the lower stratosphere from this region are in the approximate ratio 4 to 1 (STEBBINS, 1961). From the north equatorial B region of the lower stratosphere the mixing is assumed to be approximately equal in each direction. The later fallout of the  $Rh^{102}$  tracer isotope of the 1958 Orange test (GUSTAFSON, 1962) indicated that distribution of fallout originating in *all* regions of the high equatorial stratosphere was probably about equal between the two Hemispheres.

The resulting effective yields for southwards mixing are shown in Table 4.

It seems reasonable to assume that the production of THO by a thermonuclear explosion is closely related to the fusion yield, although this was not apparently the case for the first U.S.A. Ivy test of 1952, in which smaller quantities of THO appear to have been generated than in subsequent tests. BEGEMANN, (1963) and MARTELL, (1963) have reported that all the test series after Ivy produced amounts of THO roughly proportional to the total fusion energy

TABLE 4. *Estimated effective fusion yields for southwards mixing of equatorial THO within the Stratosphere.*

| Year | Effective fusion yield (MT)      |                                  |
|------|----------------------------------|----------------------------------|
|      | Lower equatorial<br>stratosphere | Upper equatorial<br>stratosphere |
| 1952 | —                                | —                                |
| 1954 | 4                                | 1.5                              |
| 1956 | 2                                | 1.0                              |
| 1957 | 1.5                              | —                                |
| 1958 | 3.5                              | 1.2                              |
| 1962 | 4                                | 0.5                              |



released. From Tables 3 and 4 it is seen that, unless tritium production per MT of fusion yield of the series was unusually high, the U.S.A. Pacific tests of 1962 should not have resulted in an increase in southern stratospheric THO fallout any greater than the increases already experienced after earlier series. Some new THO may have been present in the 1963-64 spring influx from the Starfish test whose tracer isotope,  $\text{Cd}^{109}$ , was already falling in Wellington rainwater in April 1964 (WOODWARD, 1964), but the fusion energy of this test is not likely to have exceeded 1 MT effective for mixing into the southern stratosphere. Thus the only possible source of the increased spring peak of THO recorded in the Southern Hemisphere in 1963 (Fig. 2) appears to lie in the large amounts of THO injected into the polar stratosphere by the U.S.S.R. tests of 1961 and 1962 (Table 3).

Measurements of  $\text{Sr}^{90}$  in the northern stratosphere below 21 km during January-March 1962 (MACHTA, 1962) indicated a small spread of Soviet polar debris equatorwards. Further measurements of the equatorial and southern mid-latitude stratosphere below 21 km during December 1962-January 1963 indicated that the  $\text{Sr}^{90}$  concentrations in the lower southern mid-latitude stratosphere were much the same as during the same period in 1959 (STEBBINS, 1961) which is to be expected from comparison between the 1958 and 1962 Pacific test series. Thus it seems unlikely that large quantities of Soviet THO could mix southwards at these levels, particularly in view of the finding that the lower northern polar stratosphere has a residence half-time of about six months only, resulting in the loss to the troposphere of nearly all fallout from this region through the northern mid-latitude tropopause in the first spring season following each series (MACHTA, 1962). The upper north polar stratosphere, or mesosphere, with their known longer residence times for fallout debris, appear to be the only sources for southwards mixing, and it is important that the meteorological patterns at levels above 24 km should be investigated to determine whether southwards mixing can occur either seasonally or at irregular intervals. The spring maximum of THO fallout from the southern stratosphere occurring in 1963, as evidenced by the concentrations in New Zealand rainwaters, must be ascribed to such southwards mixing of Arctic THO, unless an unusually large subsidence of

equatorial upper stratospheric air occurred in this year. The two results for 1964 spring rainwaters at Kaitoke (Table 1), which have just come to hand, show a still greater spring maximum and provide clear evidence that THO in the equatorial upper stratosphere or mesosphere is building up due to further southwards mixing of high altitude THO from the north polar regions.

It is interesting to note that the rises in Kaitoke rainwater tritium concentrations recorded during 1962-63 and 1963-64 (Table 1), are in proportion respectively to the fusion yields for the upper polar stratosphere in 1961 and 1962 (Table 3). If these rises are attributable to THO from the upper north polar stratosphere, then the first fraction appearing in 1962-63 appears at Kaitoke at the same time after detonation as if it had been injected into the upper equatorial stratosphere. The rise of 1963-64 probably represents a second fraction of 1961 upper north polar THO plus a first fraction of 1962 THO from the same region. In view of the magnitude of both rises it is deduced from the fusion yields listed in Table 3 that southwards mixing of THO within the stratosphere from the north polar regions prior to 1961 could not have appreciably affected the concentrations of tritium in southern mid-latitude rainwaters.

#### b) THERMONUCLEAR THO FROM THE SOUTHERN STRATOSPHERE IN SOUTHERN HEMISPHERE PRECIPITATION UP TO THE END OF THE MORATORIUM

Since the equatorial stratosphere appears to be the only significant source prior to 1962 of thermonuclear THO in mid-latitude rainwaters of the Southern Hemisphere, the rise of THO fallout in mid-latitude regions between 1953 and 1961 may be analysed against the background of equatorial thermonuclear testing indicated in Tables 3 and 4. Unfortunately the number of rainwaters measured for tritium in this period is very few. Between the rainwater measurement of October 1, 1953 (VON BUTTLAR & LIBBY, 1955) and the first Kaitoke measurements of 1960 there is a gap of six years for which no Wellington rainwaters have been reported. Fortunately part of this gap can be bridged by making use of the Hutt River measurements of 1956-58, and the Wairakei rainwater concentrations of 1959-60. These meas-

urements are discussed in an appendix to this section of the paper.

The experimental and deduced values of mean "fallout year" concentrations representing THO fallout for the Wellington area are listed in the comments on Wellington measurements in Table 1, and may now be compared against the background of thermonuclear testing described in Tables 3 and 4.

In any "fallout year" prior to 1961, the mean tritium concentration experienced at Kaitoke can be divided into three separate components.

$$\text{Thus} \quad T_M = T_L + T_U + T_0,$$

where

$T_M$  is the mean concentration of tritium in rainwater falling at Kaitoke during the "fallout year" in question;

$T_L$  is the contribution to  $T_M$  arising from THO originally injected into the lower equatorial stratosphere;

$T_U$  is the contribution to  $T_M$  arising from THO originally injected into the upper equatorial stratosphere and mesosphere;

$T_0$  is the mean concentration due to cosmic-ray tritium.

The concentrations  $T_L$  and  $T_U$  arise only from long-term stratospheric fallout entering the troposphere at mid-latitudes after stratospheric mixing from equatorial regions. For the purposes of estimation of the relative contributions of  $T_L$  and  $T_U$  in each year the model adopted was as follows.

The stratosphere was considered to be zoned as indicated in Fig. 3 and the fusion yields effective for southwards stratospheric mixing were as indicated in Table 4. The amounts of THO produced in each year after 1952 were considered to be proportional to the total fusion yield for that year. Thus if  $F_L$  is the fusion yield for the lower stratosphere,  $F_U$  is the fusion yield for the upper stratosphere, both effective for southwards mixing, the total number of THO molecules effective for southwards mixing from these regions are  $k_L F_L$  and  $k_U F_U$  respectively, where  $k_L$  and  $k_U$  are the proportionality constants. The mean tritium concentration actually appearing at Kaitoke in each year will be proportional to the number of THO molecules falling out of the southern stratosphere in that year. This number will be represented by the products of the contributing THO yields

and time factors representing the lapse rates of THO from each stratospheric region. It is necessary to introduce a further proportionality factor for each region,  $x_L$  and  $x_U$ , to represent the fractions of  $k_L F_L$  and  $k_U F_U$  which eventually take part in the delayed stratospheric fallout, by passing into the southern troposphere after southwards stratospheric mixing. The factor  $x_L$  will take account of any early loss of THO by gravity fallout across the equatorial tropopause as a constituent of ice crystals, or frozen down upon other quick-falling debris, or by any other gravity process, while  $x_U$  will account for loss of THO due to radioactive decay before the first fraction reaches the tropopause, and possible loss of THO to the mesosphere. Thus the contributions of any year's testing to tritium concentrations measured in a subsequent year at Kaitoke are proportional to:

$$x_L k_L F_L \times (\text{time factor})_L + \\ + x_U k_U F_U \times (\text{time factor})_U$$

or equal to

$$A x_L k_L F_L \times (\text{time factor})_L + \\ + A x_U k_U F_U \times (\text{time factor})_U,$$

where  $A$  is the proportionality constant. The time factors for the lower stratospheric components are deduced by assuming an exponential decay of THO from the lower stratosphere whose decay factor is determined by a half-residence time of 1 year and a radioactive decay constant for tritium ( $0.055 \text{ year}^{-1}$ ). For the upper stratospheric components the exponential decay is based on a 3-year half-residence time for fallout to the troposphere and the same radioactive decay constant of tritium, but is considered to commence in the third "fallout year" following upon the year of testing. In the first "fallout year" following upon testing no fallout from the upper stratosphere takes place. For the second "fallout year" the component is taken to be half that defined for the third year. The basis for this decay model is much as described by MACHTA (1962). Fig. 4 shows the fallout of  $\text{Rh}^{102}$  from the upper equatorial stratosphere (after GUSTAFSON, 1962) and Fig. 5 is a reproduction of Machta's deduced time history of the Northern Hemisphere fallout from equal injections into upper and lower equatorial stratosphere (MACHTA, 1962). The time history of

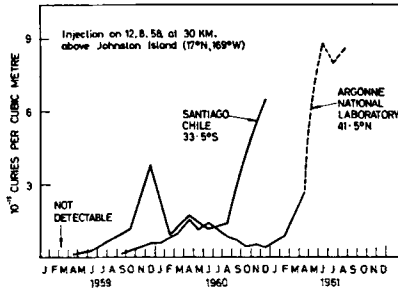


FIG. 4. Fallout of  $\text{Rh}^{102}$  tracer of 1958 Orange test, Hardtack series, after GUSTAFSON (1962).

stratospheric deposition of fallout from such injections has appeared from fallout measurements to be essentially the same for the Southern Hemisphere.

The deduced values of  $T_L$  and  $T_U$  for each year have been tabulated in Table 5. The pattern emerging is one of steadily increasing  $T_U$  from 1955 to 1961, and of decreasing  $T_L$  from 1954 to 1961 except for temporary increases recorded in 1956–57 and 1958–59. In order to determine whether this pattern was not imposed primarily by the choice of model, reasonable alterations to the model were made and the end results compared with those of Table 5. Details of the model undergoing alteration were the fusion yields of Table 4, the half-residence times for fallout from lower and upper stratosphere, and the magnitude of the second-year upper stratospheric fallout by comparison with the third-year component. From all these variations the same pattern emerged, although the absolute magnitudes of the components  $T_L$  and  $T_U$  were somewhat altered and the relative diffe-

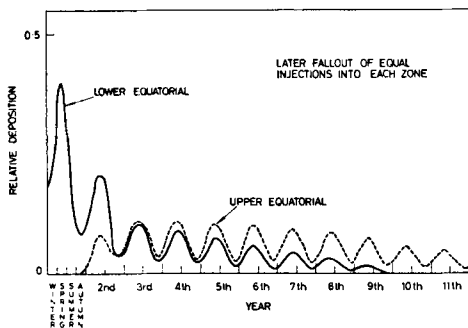


FIG. 5. Fallout history of isotopes subjected to normal stratospheric mixing processes from the upper and lower equatorial stratosphere, after MACHTA (1962).

TABLE 5. *Stratospheric components of fallout of Tritium in Kaitoke rainwater based on test yields.*

(All detonations considered effective)

| Fallout year | $T_L$            | $T_U$            | Measured $T_M$ (T. U.) |
|--------------|------------------|------------------|------------------------|
| 1954–55      | $1.90 A x_L k_L$ | —                | —                      |
| 1955–56      | $0.90 A x_L k_L$ | $0.13 A x_U k_U$ | —                      |
| 1956–57      | $1.39 A x_L k_L$ | $0.27 A x_U k_U$ | 5.0                    |
| 1957–58      | $1.38 A x_L k_L$ | $0.29 A x_U k_U$ | 7.7                    |
| 1958–59      | $2.35 A x_L k_L$ | $0.33 A x_U k_U$ | —                      |
| 1959–60      | $1.11 A x_L k_L$ | $0.36 A x_U k_U$ | 7.7–10.15              |
| 1960–61      | $0.52 A x_L k_L$ | $0.40 A x_U k_U$ | 10.15                  |

rences between successive values of  $T_U$  were found to be particularly sensitive to the variations.

From Table 5 it is deduced that the contribution of  $T_L$  to  $T_M$  is very small by comparison with the contribution of  $T_U$ , provided always that the model used for the time behaviour of stratospheric THO fallout is accepted as valid. However it may be considered likely that the value of  $x_L$  is not invariable for all types of thermonuclear detonation. A surface detonation (e.g. the Bravo test of the 1954 U.S.A. Castle series) may carry aloft large quantities of sea-water which would be expected to exert a considerable influence upon the early fallout of THO from the lower equatorial stratosphere (BEGEMANN & LIBBY, 1957). This influence would not be effective for air bursts. An analysis equivalent to that summarised in Table 5 has been performed to determine the relative magnitudes of  $T_L$  and  $T_U$  in the years 1954–61, using an alteration to the previous model which stipulates that  $x_L = 0$  for surface detonations. It is difficult to categorize the Cherokee deto-

TABLE 5 a. *Stratospheric components of Tritium in Kaitoke rainwater, based on test yields.*

(Air bursts only considered effective)

| Fallout year | $T_L$            | $T_U$            | Measured $T_M$ (T. U.) |
|--------------|------------------|------------------|------------------------|
| 1954–55      | —                | —                | —                      |
| 1955–56      | —                | $0.13 A x_U k_U$ | —                      |
| 1956–57      | $0.97 A x_L k_L$ | $0.27 A x_U k_U$ | 5.0                    |
| 1957–58      | $1.19 A x_L k_L$ | $0.29 A x_U k_U$ | 7.7                    |
| 1958–59      | $2.50 A x_L k_L$ | $0.33 A x_U k_U$ | —                      |
| 1959–60      | $1.17 A x_L k_L$ | $0.36 A x_U k_U$ | 7.7–10.15              |
| 1960–61      | $0.55 A x_L k_L$ | $0.40 A x_U k_U$ | 10.15                  |

nation of the 1956 U.S.A. Redwing series. The approximate statement of the yield of this test does not allow a definite decision as to whether the fireball reached far enough down to draw significant quantities of sea-water aloft into the stratosphere. For the present analysis Cherokee has been treated as an air burst. The resulting components  $T_L$  and  $T_U$  are listed in Table 5a. As with the earlier analysis, it appears that the contribution of  $T_L$  to  $T_M$  must be very small.

Thus 
$$x_L k_L < x_U k_U.$$

The concentrations measured at Kaitoke appear therefore to be due mainly to THO falling out from the upper stratosphere. Increased spring concentrations may generally be expected in the second spring season following upon tests conducted within the upper stratosphere, or from tropospheric detonations of several MT yield which push their clouds up to the higher levels of the stratosphere. It becomes important to determine whether the small contributions to long-term THO fallout from the lower equatorial stratosphere could be postulated as a consequence of a fractionation effect at the time of formation in a thermonuclear test ( $k_L < k_U$ ), or else of a meteorological removal of THO from this region within a period short by comparison with the meteorological processes which mix other debris polewards towards the mid-latitude region where mixing to the troposphere has been found to occur ( $x_L < x_U$ ). Since there have been many tests of about 1 MT yield whose clouds have penetrated the lower stratosphere only, considerable quantities of thermonuclear THO must have been created in this region. Thus the second interpretation, i.e.  $x_L < x_U$ , must be favoured.

The early removal of thermonuclear THO from the lower equatorial stratosphere now appears to be a characteristic feature of all types of detonation. At the present time the reason for this behaviour is not known. The ice-crystal mechanism suggested by BEGEMANN & LIBBY (1957) cannot be extended to include air bursts.

In the foregoing treatment the effect of the slowly increasing concentrations of surface oceanic water has been neglected. It has been evaluated that the contribution of rising surface ocean concentration to increase of  $T_M$  is only a fraction of 1 T.U. in southern mid-latitudes be-

tween 1953 and 1961, and hence is not of great significance.

The only tritium measurements of stratospheric water vapour so far reported (HAGEMANN *et al.*, 1959; BROWN *et al.*, 1961) have been made in the northern mid-latitude region of descending tritium fallout, where THO may be expected to be found at all sampling altitudes and in varying quantities with season. No measurements of tritium in equatorial stratospheric water vapour have been reported.

It is clearly important to consider the fallout of THO from the Arctic stratosphere. Here the testing season has generally occurred in winter. The temperature distribution of the Arctic stratosphere and height of the tropopause provide circumstances somewhat different than is the case for Pacific testing. Some Northern Hemisphere stations (Reykjavik, Trysil, Saskatchewan, Darmstadt, Ottawa) have concentrations of tritium in their rainwaters (*I.A.E.A. Tritium Lists*) during late 1961, which suggest that THO from the Soviet Arctic tests of that year was appearing during the testing period. However most of the THO from Soviet Arctic tests appears to be deposited, in the manner of other fallout products, by influx to the troposphere through the mid-latitude tropopause in the first spring season following a series. Thus conditions for removal of THO from the lower stratosphere appear to differ between the polar and equatorial regions.

#### c) FURTHER DISCUSSION OF HISTORY OF THERMONUCLEAR THO FALLOUT IN THE SOUTHERN HEMISPHERE

It has been shown above that the measured tritium concentrations of mid-latitude Southern Hemisphere rainwaters are to be interpreted by assuming that the predominant fallout of thermonuclear THO from the southern stratosphere originates at upper stratospheric levels. The first fraction of this fallout after a particular series appears in the second spring season after detonation and is followed in the next "fallout year" by a slightly larger fraction. In "fallout years" subsequent to the third after detonation the amounts of fallout decrease within the compass of an upper stratospheric half-residence time of something like three years. Thus the first significant increase over the cosmic-ray level of tritium in New Zealand rainwaters is thought

to have occurred in the spring season of the "fallout year" 1955–56, the originating thermonuclear tests being the Castle series of 1954. The estimate of LAL & PETERS (1962), depicted in Fig. 1, predicts an average concentration of about 1.5 T.U. for cosmic-ray tritium fallout in New Zealand rainwater. Beside this estimate the measurement of  $1.7 \text{ T.U.} \pm 0.2$  for Wellington rainwater on October 1, 1953 (VON BUTTLAR & LIBBY, 1955), is a reasonable result for a spring rainwater containing negligible thermonuclear tritium, and provides confirmation that delayed stratospheric fallout of THO from the Ivy test series of 1952 did not appreciably affect southern mid-latitude rainwater concentrations. Hydrological investigations of tritium in New Zealand groundwaters (GRANT-TAYLOR & TAYLOR, 1966) have indicated that the theoretical estimate of Lal and Peters is very close to the mark. In contrast it is clear that by 1956 there was thermonuclear tritium in the Hutt River samples (Fig. 4). Furthermore, the single value for the Hutt River concentration on April 29, 1956, suggests that the mean rainwater tritium concentration at Kaitoke during 1955–56 was not much lower than that for 1956–57. It seems most likely that stratospheric THO from the Castle series raised the tritium concentration of New Zealand rainwaters by a factor of about 3, and that the mean natural concentration was less than 2 T.U.

The 1954–55 Antarctic snow measurements average 16.5 T.U. in concentration (BEGEMANN & LIBBY, 1957) and do not show a decrease in the deeper layers which might suggest decreased THO deposition in earlier years. This average is confirmed by the later results quoted by BEGEMANN (1958B) for snows collected during early 1956. This is in marked contrast to the findings on Sr-90 and gross  $\beta$ -activity in Antarctic snows (LIBBY, 1956; PICCIOTTO and WILGAIN 1963; PICCIOTTO *et al.*, 1964), which show fission product concentrations in the snows of late 1954—early 1955 clearly originating as stratospheric fallout from the Castle series. It seems therefore to be definitely confirmed that no delayed fallout of THO accompanied the stratospheric fallout of fission products in the first spring season following Castle. In the case of BEGEMANN's 1956 samples any thermonuclear THO present would comprise only a small fraction of the topmost sample at each location from the spring snow immediately pre-

ceding collection, and it is not surprising that thermonuclear THO could not be identified. Many more measurements of THO in fresh Antarctic snows are needed to establish the pattern of seasonal deposition and its variation over the Continent. Tritium could well prove to be a most potent tool in the investigation of the Antarctic meteorological cycle. It is clear from available measurements that, although the fallout of THO is much less in the Antarctic than it is in mid-latitudes, the low amounts of diluting tropospheric vapour present in the Antarctic atmosphere make concentrations several times higher. From the latest available data it appears that the average concentration of natural tritium in coastal Antarctic snows was about 10 times that in New Zealand rainwaters (cf. TAYLOR, 1964).

On the present interpretation of stratospheric fallout the increase of almost 3 T.U. experienced in New Zealand rainwaters in 1957–58 is due to the first appearance of THO from the U.S.A. Redwing series of 1956. Thereafter the next significant change occurred in 1959–60 with the arrival of the first fraction of the Hardtack THO of 1958, to be followed by a somewhat greater second fraction in 1960–61.

The tritium results for Kaitoke rainwaters (Fig. 2) also give some evidence of a minor peak in the autumn months of 1961–62. It is tempting to assign this increase to the tropospheric exchange of THO across the equator at the time of the Northern Hemisphere spring peak. However data in Table 1, and the comparison between various collection stations listed in Table 6, suggest that significant tropospheric exchange of THO did not take place southwards across the equator in 1962, by comparison with 1961, when Northern Hemisphere concentrations were very much lower. Also the concentrations at Funafuti Atoll (Fig. 6) and Western Samoa (June 1962) do not indicate that quantities of THO significant enough to affect southern mid-latitude rainwater concentrations were moving southwards within the lower troposphere during the autumn and winter periods of 1962. If THO were crossing the equator at higher tropospheric levels before reaching lower levels at southern mid-latitudes then it would be reasonable to expect quite startling rises in rainwater tritium concentrations at Kaitoke in the autumn periods of 1962 and 1963 by comparison with the same periods in 1961. This is

TABLE 6. Latitude comparison of Tritium fallout during 1961, 1962 and 1963.

| Locality                                | Rainwater T. U.<br>1961 | Rainwater T. U.<br>1962 | Rainwater T. U.<br>1963 |
|-----------------------------------------|-------------------------|-------------------------|-------------------------|
| Saskatchewan                            | 21.3 (April)            | 968 (April)             | 7600 (May)              |
| Tokyo                                   | 26 (April)              | 131 (April)             | No Result               |
| Valentia, Ireland                       | 46.6 (May)              | 339 (April)             | 1089 (April)            |
| Entebbe, Uganda (Equator)               | 13.2 (April)            | 15 (April)              | 33.6 (April)            |
| Kaitoke, New Zealand                    | Mean 1960-61 = 10.2     | Mean 1961-62 = 9.5      | Mean 1962-63 = 15.4     |
| Data taken from I.A. E.A. tritium lists |                         |                         |                         |

not the case. A more likely explanation of the tritium concentrations recorded during May-June 1962 would seem to be the presence of some THO, injected into the lower stratosphere by the U.S.A. Pacific tests at Christmas Island, which has time to mix a little way south of the equator within the stratosphere before falling out within the 1-2 month period for early THO fallout.

The Funafuti measurements shown in Fig. 6 show clear evidence of the 1-2 month early fallout residence time observed in the Northern Hemisphere after earlier Pacific test series.

A most interesting comparison can be made between the tritium concentrations in Kaitoke rainwaters and those reported by the Stockholm Radioactive Dating Laboratory (*I.A.E.A. Tritium Lists*) for two other southern mid-latitude oceanic stations. Fig. 7 shows the data for Kaitoke, Marion Island and Gough Island during 1961-63. There is a closely similar time trend at all three stations and the concentrations are also very similar. This comparison yields further

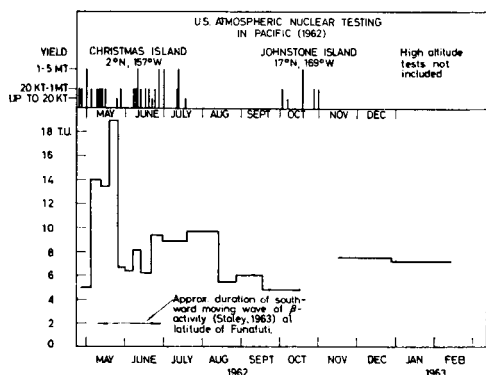


Fig. 6. Tritium concentrations of rainwater samples collected at Funafuti Atoll, 9°S, 179°E, during and immediately after the U.S. Pacific test series of 1962.

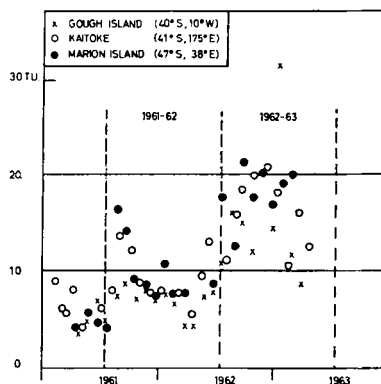


Fig. 7. Comparison of tritium in rainwaters at Kaitoke, Gough Island and Marion Island, 1961-63.

confidence in the tritium measurements at both the Stockholm and New Zealand laboratories.

Monthly composite rainwater samples for tritium measurements at this laboratory are currently being collected at Rarotonga, Kaitaia, Kaitoke, Invercargill, Melbourne and Campbell Island.

#### d) THE "FALLOUT YEAR"

The fallout pattern for  $\text{Rh}^{102}$ , depicted in Fig. 4, indicates that the Southern Hemisphere "fallout year" for THO should more strictly be taken to commence in April. However it is clear that the initial appearance of any new thermonuclear THO in the months April-July is not significant enough to affect the ambient concentrations of the previous "fallout year" unless a very much larger spring influx is anticipated.

#### e) APPENDIX. DEDUCTION OF THO IN KAITOKE RAINWATERS FOR THE "FALLOUT YEARS" 1956-57, 1957-58 AND 1959-60

The upper reaches of the Hutt River drain a hilly area of subtropical rain forest (Kaitoke) where the average annual rainfall of 250 cm is

rather well distributed. The hold-up time of surface moisture draining into the river is likely to be several weeks at the most. Thus the mean of a series of regularly repeated tritium measurements of riverwater over a "fallout year" should represent closely the concentration of tritium in the sum total of all rainwater which fell in the area during that year. If a simultaneous set of rainwater samples is collected in the area the mean river tritium concentration should be equal to the mean concentration of the set of rainwater samples, when each sample is weighted by the precipitation recorded during its collection period. Tritium measurements of samples from the Hutt River at Kaitoke are depicted in Fig. 8. The "fallout year" 1960-61 is the only year during which such a series of measurements is presently available for both rain and riverwater samples at Kaitoke. In this year the rainwater tritium mean (weighted by precipitation) was  $8.85 \text{ T.U.} \pm 0.3$ , in excellent agreement with the river mean of  $8.83 \text{ T.U.} \pm 0.3$ . It will be assumed therefore that the mean river concentrations for 1956-57 and 1957-58 are equal to the precipitation-weighted means for rainwater in the area over these two "fallout years".

The precipitation-weighted mean of tritium concentration in a series of rainwater samples, representing as it does the tritium concentration of all water which has fallen in the immediate area of the collection station throughout the series, is much influenced by local topography and both seasonal and yearly variations in precipitation. A clearer assessment of the year by year changes of the THO fallout from the southern stratosphere is given by the changes in the mean tritium concentration of atmospheric vapour in the lower troposphere in the area. Over a yearly period this mean concentration of tritium in atmospheric vapour should be proportional to the mean concentration of a series of regularly repeated rainwater samples collected throughout the entire period. If the collection periods for the rainwater samples are not closely equal, as has been the case at Kaitoke, then the mean rainwater concentrations must be obtained by weighting each concentration by the time of its collection period. It is desirable to express the mean rainwater tritium concentrations in this manner to achieve the best indication of the year by year variations of the stratospheric THO fallout.

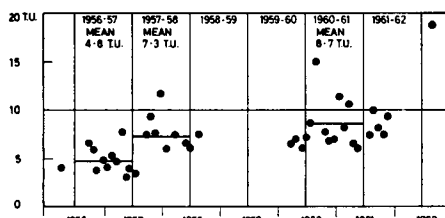


FIG. 8. Tritium concentrations of water from the upper reaches of the Hutt River, near Wellington.

To deduce such a mean for 1956-57 and 1957-58 at Kaitoke, the only figures available are the precipitation-weighted means derived from the Hutt River data. To derive the unweighted means the following procedure has been adopted. An idealised quarterly distribution of rainwater THO concentrations was assumed for each year to correspond with that measured at Kaitoke during 1960-61. During this "fallout year" the quarterly mean concentrations were approximately 1.5, 1.0, 0.8 and 0.7 times the time-weighted mean rainwater concentration over the entire "fallout year". If this distribution is assumed for the "fallout years" 1956-57 and 1957-58, then knowledge of the seasonal distribution of precipitation in the area during these years enables conversion of the precipitation-weighted mean rainwater concentration to the time-weighted mean which reflects the amount of THO in water vapour over the area and may be compared safely with the means derived from the later rainwater measurements.

A further part of the gap in the rainwater tritium measurements in the Wellington area can be bridged by making comparisons between Kaitoke and Wairakei measurements. As shown by the comments on the Wairakei measurements detailed in Table 1, the Wairakei concentrations are a close parallel to those at Kaitoke. This enables the 1959 Wairakei data to be used as an indication of Kaitoke tritium concentrations in that year. The 1959-60 mean tritium concentration in Kaitoke rainwater is deduced thereby to have been lower than that of 1960-61 but higher than that of 1957-58, i.e. between 7.7 and 10.15 T.U.

### The effect of the continental environment

Comment on the effects of the continental environment on the concentrations of tritium

in rainwater at individual stations have been made in the tabulated data of Table 1.

Ideally the effect of the continental environment can only be examined profitably if one first has a basis of a comprehensive set of measurements of tritium in oceanic rainwaters, and in the surface ocean affecting each environment that receives these rainwaters. From such measurements knowledge of several features may be obtained which have similar effect also in the continental environment. These features broadly are:

(a) Latitude variation of THO fallout in each Hemisphere caused by localised mixing of stratospheric THO into the troposphere and its subsequent meteorological dispersal throughout the troposphere.

(b) Longitudinal and latitudinal variation of tritium concentrations in rainwater due to dry or wet precipitation areas.

(c) Seasonal variations of the stratospheric-tropospheric exchange of THO.

(d) The effect of local climate in varying the tritium concentration of rainwater by the evaporation of falling raindrops and other micro-meteorological processes involved in the formation and deposition of precipitation (BOLIN, 1958; FRIEDMAN *et al.*, 1962). In this respect studies of the variation of DHO in natural waters will prove to be of considerable assistance (EHHALT *et al.*, 1963).

These studies should be based on measurements of delayed stratospheric THO fallout. Thermonuclear weapons testing introduces a 1–2 month perturbation whose THO will not be deposited globally, but may provide useful short term studies in isolated localities.

Once such a basis has been established it becomes necessary to study carefully selected continental localities, each of which is chosen to demonstrate one particular aspect of the continental effects only. For example to study the altitude effect a relatively sharp line of mountains set in a land environment receiving oceanic rainwaters would be required. The Southern Alps of New Zealand, rising to heights of over 12,000 ft., provide such a locality. It would be interesting to compare the altitude effect of, say, Mt. Cook with that of Mt. Erebus, Ross Island, which appears to be an Antarctic mountain very suitable for such a study. The influence of large bodies of inland water on the

tritium balance of the surrounding regions could also be examined. The North American Great Lakes, the Caspian Sea, Lake Victoria and even the Baltic Sea may be particularly suitable for this purpose. Such problems may be made more complicated in the Northern Hemisphere because of the confused background of tritium in surface moisture and oceanic waters due to past THO fallout. In certain cases, however, high surface moisture concentrations may make continental effects more readily detectable. A locality such as that of Pretoria represents a complicated situation. Pretoria lies within a large continental area at considerable altitude and in close proximity to desert areas. Evaporation of falling raindrops is an important effect in certain seasons (EHHALT *et al.*, 1963).

A recent measurement of tritium in snow has suggested that snow may not represent an ideal storage medium for the investigation of previous history of precipitation of oxygen and hydrogen isotopes. In an attempt to obtain a blank tritium water to decide upon the validity of many measurements of hydrothermal waters below 0.2 T.U. conducted at this laboratory, a quantity of snow was collected from a snow mine at the South Pole. The sample should have represented snow at least 100 years old and was taken during January, 1964, 1 metre in from the face of the mine. Its tritium concentration was 40.5 T.U., while that of a surface snow was 200 T.U. The deep snow was clearly heavily contaminated with thermonuclear tritium. This mine and its vicinity were sampled again in November, 1964 in order to determine whether contamination results from the exposure of the face of the mine over several years to atmospheric vapour of high tritium concentration, or whether contamination is more generally apparent at depths well below the snow surface. The experiments were designed to investigate the possibility of sub-surface snow acting as a cold-trap for atmospheric vapour. In this respect the smoothing out of deuterium variations in lower layers in two stratigraphic snow studies (BOTTER *et al.*, 1960; GONFIANTINI *et al.*, 1963) has already been noticed, but this may be mainly due to percolation of summer melt through the top layers.

## Conclusions

The following conclusions may be drawn from the data.



1. Tritium concentrations of Southern Hemisphere precipitation have been rising since the onset of thermonuclear weapons testing. The concentrations in mid-latitude spring rainwaters in 1964 are about 30 times the natural level.

2. Between 20° S and the equator, the tritium concentrations of post-thermonuclear rainwaters are determined mainly by the concentrations pertaining in the Northern Hemisphere, where tritium concentrations have been far higher than in the Southern Hemisphere. The penetration of more highly tritiated water vapour across the equator does not appear to affect concentrations southwards of about 20° S. In the Pacific area, rainwater concentrations in the region 0°–20° S can be increased significantly for about 1 month immediately following upon Pacific equatorial thermonuclear test series. In some cases such increased concentrations may be recorded in lesser degree farther south than 20° S.

3. At southern latitudes greater than 20° S the tritium concentrations of rainwaters are determined by the amount of thermonuclear THO fallout from the southern stratosphere. There is a marked seasonal dependence of spring maximum and autumn minimum in mid-latitude oceanic rainwaters, and the magnitude of the rainwater concentrations is closely related to the recent history of thermonuclear testing. The seasonal dependence of tritium in Antarctic snows has not yet been established. The latitudinal variation of thermonuclear THO fallout (expressed as tritium atoms per cm<sup>2</sup> per sec.) appears to resemble closely the pattern deduced by LAL & PETERS (1962) for cosmic-ray THO fallout.

4. Thermonuclear THO created in the lower equatorial stratosphere appears to be removed to the troposphere by some gravitational process within a 1–2 month mean residence period, and does not take part in the delayed fallout exhibited by particulate fission products which undergo slow polewards stratospheric mixing followed by mid-latitude stratosphere-troposphere exchange with a mean stratospheric residence time of 1 year approx. This behaviour is independent of the nature of the thermonuclear detonation (surface or air burst) and suggests strongly that some natural process must exist which results in a 1–2 month mean residence period for *all* water vapour in the lower equatorial stratosphere. It appears that this

unknown process does not operate in the winter lower north polar stratosphere.

5. Thermonuclear concentrations of THO in precipitation southwards of 20° S appear to arise entirely from THO which has been created in the upper stratosphere and mesosphere. The fallout of this THO appears in southern mid-latitude rainwaters in the second spring season following creation and exhibits a mean stratospheric residence time of several years. The data do not suggest an overall mechanism for transport from the upper stratosphere to ground-level but two points about the mechanism are clear. Firstly, the transfer of thermonuclear THO from the upper to the lower stratosphere is not a continuous process. It appears that a fixed fraction of the upper stratospheric reservoir of thermonuclear THO is transferred abruptly to the lower southern stratosphere prior to the spring season. This fixed fraction then becomes an available lower stratospheric reservoir for stratosphere-troposphere exchange on the previously mentioned annual pattern. It is not clear whether the abrupt transfer of THO from upper to lower stratosphere takes place over the poles or at lower latitudes. Secondly, the main stratosphere-troposphere exchange of delayed THO fallout takes place in mid-latitudes, as has been already determined for other fallout products.

6. Prior to 1961 the equatorial stratosphere was the only significant source of thermonuclear THO in Southern Hemisphere precipitation southwards of 20° S. Since 1961 thermonuclear THO from Soviet arctic testing has been spreading southwards at high stratospheric or mesospheric levels, and is building up THO as a long-residence reservoir in the upper stratosphere or mesosphere. This reservoir is contributing to greatly increased amounts of THO in Southern Hemisphere precipitation.

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