THE MEASUREMENT OF TRITIUM ACTIVITY IN
NATURAL WATERS

Part II. Characteristics of Global Fallout of H³ and Sr⁹⁰

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ABSTRACT

The concentrations of tritium have been determined in wet precipitations occurring over the Indian subcontinent during 1961–64, using a sensitive method for counting of tritium activity discussed in Part I* of this paper.

The tritium concentrations varied significantly during the period of observation; highest concentrations were observed during 1963. An analysis of the data reported here, in conjunction with those available for concentrations of H³ and Sr⁹⁰ in rains at higher latitudes, reveals that these nuclides which were originally placed at high altitudes in the polar regions during late 1962, were deposited chiefly at 30°–90° latitudes during 1963 and 1964 respectively in relative proportions of 1 and 0.6. The data show that the largest gradients in their zonal deposition occur at about 35°–40° N latitude and that to a first approximation, their deposition per unit area in 1963 or 1964 was practically uniform, separately in the 30°–90° and 0°–30° latitude regions. This observation suggests the existence of two well-defined cells, which are internally well mixed: the meridional transport to low latitudes occurs as a result of interaction between these cells. The annual deposition rates of Sr⁹⁰ as observed during 1963 and 1964 suggest a mean time of 3 months for exchange of air between the two cells, in good agreement with the values deduced for mid-months of the year on the basis of analysis of bomb produced C¹⁴ data.

The tritium and strontium data for the inland, coastal and island stations are analysed to evaluate the importance of (i) the re-evaporation of tritium from continents, and (ii) the molecular exchange of atmospheric tritium with oceanic water. Process (i) probably plays a significant role over the continents throughout the year; its effect, however, is experimentally visible only during June to September. The

estimated concentration of $\text{H}^3$ in evaporated water suggests that the precipitated water mixes very slowly with that in the soil; limits on the equivalent amount of exchangeable soil water are given.

It is shown that the relative wet deposition of $\text{H}^3$ and $\text{Sr}^{90}$ at island and coastal stations is similar to their estimated concentration ratio in upper level tropospheric air. Furthermore, the relative concentrations of $\text{H}^3$ and $\text{Sr}^{90}$ at continental and oceanic stations differ only to the extent expected due to reinjection of $\text{H}^3$ over continents. Thus, if one takes into account the recycling of $\text{H}^3$ at continental stations (which results in about a 50% higher apparent deposition on an annual basis), one is led to the conclusion that process (ii) is rather unimportant; an upper limit of 30% on the fraction of tritium removed over oceans by molecular exchange is deduced.

The mean annual concentration of $\text{Sr}^{90}$ in wet precipitation is lower at oceanic stations compared to that at continental stations. This could be due to meteorological effects peculiar to oceanic areas, e.g., higher rainfall and quick recycling of evaporated water. Otherwise, one must postulate a significant removal of $\text{Sr}^{90}$ (and $\text{H}^3$) by ocean spray and jet action.

1. INTRODUCTION

Several major injections of man-made tritium have occurred since 1953, particularly at high northern latitudes. Peak concentration values during spring or summer of 1954, 1956, 1958 and 1959 were around 700, 300, 1000 and 1500 T.U.† respectively at latitudes north of 30° N. (Buttlar, 1963; Brown, 1964). In contrast, the pre-bomb levels north of 30° N., arising due to cosmic ray production of $\text{H}^3$, were around 10 T.U. (Kauffman and Libby, 1954). As early as 1954, Begemann and Libby realised the value of man-made $\text{H}^3$ in the study of water cycle (Begemann and Libby, 1957). Since then, several papers have appeared discussing the potentialities of man-made tritium as a tracer for meteorological and hydrological investigations (Eriksson, 1965; Payne et al., 1964; Libby, 1963; Libby, 1961).

In 1961, we started a programme to measure tritium in precipitations occurring over the Indian subcontinent with a view to study meteorological processes as well as to determine the tritium levels in the waters that feed the various ground water reservoirs. Because of the expected seasonal and

† One T.U. signifies a ratio of $\text{H}^3$/H$^1 = 10^{-14}$; this method of specifying tritium concentrations, proposed by W. F. Libby, is now commonly adopted.
geographical variations in the fallout of tritium, and our interest in the hydro-
logical problems of the country, covering practically all regions from north
to south, we undertook measurements of the tritium concentrations in rains
at selected grid stations. The results of these measurements, till the end of
1964, are presented here.

Tritium levels were found to be the highest yet observed during 1963.
The available data for 1963 (during which period one can ignore the contri-
butions from previous injections) and the subsequent year have been analysed
for their geographical variations. These results, when combined with data
available for tritium levels at higher latitudes and for Sr\(^{90}\) in the northern
hemisphere, lead to very useful information pertaining to the large-scale
meridional tropospheric circulation, role of molecular exchange of atmos-
pheric tritium with oceanic water and the re-evaporation of tritium from
continents.

2. EXPERIMENTAL PROCEDURE AND DATA

Water samples were collected on a monthly basis from 17 grid stations
(see Fig. 1) by the rain-gauge method. All collections amounting to more
than 100 c.c. per month were preserved in a polyethylene stoppered bottle.
Tritium concentrations were measured in the annual “composite” samples,
prepared by mixing individual month’s collections in proportion to rainfall
during those months. At some stations, individual month’s collections
have been analysed to study seasonal effects. A few isolated samples of
“single rains” have also been analysed.

Methane gas was directly synthesised from the “sample” water and
its tritium activity determined with a sensitive gas-proportional counting
system. No electrolytic enrichment of tritium was necessary for the rain
samples as the tritium concentrations in rains during the period of measure-
ment were sufficiently large. The techniques for methane synthesis and the
counting of tritium have been described in detail in Part I of this paper (Lal
and Athavale, 1966).

The measured tritium concentrations for annual composite samples
are summarised in Table I, which also give details with respect to the geo-
ographical location of the station and the rainfall during the period of sampling
months. All composite samples in this table represent essentially the total
annual rainfall, except for the case of Srinagar (1963) and Minicoy (1964)
collection. The months of rainfall are June–September at most stations.
and this has to be borne in mind in comparing our data on tritium concentrations with those at higher latitude stations, where rainfall is not so peaked in certain months. The results of $\text{H}^3$ measurements in monthly composite samples, and a few individual rain samples are summarised in Table II. Tritium concentrations are given in T.U. (1 T.U. signifies a ratio of $\text{T/\text{H}} = 10^{-18}$). With this nomenclature, one finds that water having a concen-

Fig. 1. Annual mean (weighted) tritium concentrations in precipitation at 17 stations in the Indian subcontinent during 1962, 1963, and 1964.
Measurement of Tritium Activity in Natural Waters—II

The concentration of 1 T.U. has $7.2 \times 10^{-3}$ d.p.m. $\text{H}^3$ (or $6.7 \times 10^4$ atoms $\text{H}^3$) per cubic centimetre. The half-life of tritium has been taken to be 12.3 years (Jones, 1955).

**Table I**

<table>
<thead>
<tr>
<th>Sampling site (Lat., Long.)</th>
<th>Period of collection (Months of the year)</th>
<th>Rainfall (cm.)</th>
<th>Tritium concentration (T.U.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Srinagar 34° 05' N, 74° 50' E</td>
<td>7, 9, 10, 11, 12 (1962)</td>
<td>29.1</td>
<td>480±12</td>
</tr>
<tr>
<td>Amritsar 31°37' N, 74° 53' E</td>
<td>7, 8, 9 (1962)</td>
<td>19.1</td>
<td>200±12</td>
</tr>
<tr>
<td>Delhi 28°35' N, 77°12' E</td>
<td>6, 7, 8, 9 (1964)</td>
<td>119</td>
<td>510±17</td>
</tr>
<tr>
<td>Bikaner 28°00' N, 73°18' E</td>
<td>7, 8, 9 (1962)</td>
<td>23.7</td>
<td>160±16</td>
</tr>
<tr>
<td>Patna 25°37' N, 85°10' E</td>
<td>6, 7, 8, 9 (1964)</td>
<td>81.6</td>
<td>410±10</td>
</tr>
<tr>
<td>Shillong 25°34', N, 91°53' E</td>
<td>6, 7, 8, 9, 10 (1962)</td>
<td>113</td>
<td>280±18</td>
</tr>
<tr>
<td>Ahmedabad 23°04' N, 79°07' E</td>
<td>6, 7, 8, 9 (1963)</td>
<td>75.4</td>
<td>94±12</td>
</tr>
<tr>
<td>Calcutta 22°39' N, 11°54' E</td>
<td>6, 7, 8, 9 (1962)</td>
<td>81.1</td>
<td>170±16</td>
</tr>
<tr>
<td>Nagpur 21°09' N, 79°07' E</td>
<td>6, 7, 8, 9, 12 (1962)</td>
<td>107</td>
<td>110±12</td>
</tr>
<tr>
<td>Sampling site (Lat., Long.)</td>
<td>Period of collection (Months of the year)</td>
<td>Rainfall (cm.)</td>
<td>Tritium concentration (T.U.)</td>
</tr>
<tr>
<td>----------------------------</td>
<td>------------------------------------------</td>
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<td>----------------------------</td>
</tr>
<tr>
<td>Bhubaneshwar 20°48' N, 85°56' E</td>
<td>6, 7, 8, 9 (1964)</td>
<td>118</td>
<td>270±17</td>
</tr>
<tr>
<td>Bombay 18°54' N, 72°49' E</td>
<td>6, 7, 8, 9 (1962)</td>
<td>192.2</td>
<td>60±12</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1963)</td>
<td>250.2</td>
<td>490±35</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1964)</td>
<td>184.8</td>
<td>170±17</td>
</tr>
<tr>
<td>Khandala 18°46' N, 73°22' E</td>
<td>6, 7, 8, 9 (1962)</td>
<td>494</td>
<td>77±17</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1963)</td>
<td>503</td>
<td>550±22</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1964)</td>
<td>350</td>
<td>220±16</td>
</tr>
<tr>
<td>Hyderabad 17°27' N, 78°28' E</td>
<td>7, 8, 9, 10, 11, 12 (1962)</td>
<td>105</td>
<td>110±20</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1963)</td>
<td>75.9</td>
<td>680±30</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1964)</td>
<td>52.8</td>
<td>340±10</td>
</tr>
<tr>
<td>Madras 13°00' N, 80°11' E</td>
<td>6, 7, 8, 9, 10, 11, 12 (1962)</td>
<td>115.6</td>
<td>100±10</td>
</tr>
<tr>
<td></td>
<td>1, 4, 6, 7, 8, 9, (1963)</td>
<td>53.8</td>
<td>270±10</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9, 10, 11, 12 (1964)</td>
<td>130.4</td>
<td>140±10</td>
</tr>
<tr>
<td>Bangalore 12°57' N, 77°38' E</td>
<td>6, 8, 9, 10, 12 (1962)</td>
<td>75.9</td>
<td>110±16</td>
</tr>
<tr>
<td></td>
<td>4, 5, 6, 7, 8, 9 (1963)</td>
<td>75.0</td>
<td>470±16</td>
</tr>
<tr>
<td></td>
<td>10, 11, 12 (1963)</td>
<td>30.7</td>
<td>280±17</td>
</tr>
<tr>
<td></td>
<td>6, 7, 8, 9 (1964)</td>
<td>72.1</td>
<td>290±10</td>
</tr>
<tr>
<td>Trivandrum 08°29' N, 76°57' E</td>
<td>6, 7, 8, 9 (1962)</td>
<td>84.3</td>
<td>72±12</td>
</tr>
<tr>
<td></td>
<td>1 to 12 (1963)</td>
<td>175.5</td>
<td>180±18</td>
</tr>
<tr>
<td></td>
<td>2, 6, 7, 8, 9 (1964)</td>
<td>96.1</td>
<td>150±16</td>
</tr>
<tr>
<td>Minicoy 08°18' N, 73°00' E</td>
<td>7, 8, 9, 10, 11, 12 (1963)</td>
<td>113.4</td>
<td>170±16</td>
</tr>
<tr>
<td></td>
<td>1, 3, 4 (1964)</td>
<td>12.8</td>
<td>98±18</td>
</tr>
</tbody>
</table>

During 1959–61, we have only a few measurements of H² concentrations in rains at Delhi, Bombay and Ootacamund (Table II). The levels during 1961 are in the neighbourhood of 50 T.U., being lower than during the preceding years 1960 and 1959. This would be expected also as the period January 1959–September 1961 marked the “moratorium era”. Tritium levels were continuously decreasing till late 1961 when the resumption of testing by U.S.S.R. shot up the levels again. The artificial tritium injected at higher latitudes significantly raised the tritium concentrations in 1962 rains in India. As can be seen from Table I, the mean concentrations reached

**Table II**

*Tritium Levels in Individual Rains and Monthly Composite Samples*

<table>
<thead>
<tr>
<th>Sampling site (Lat., Long.)</th>
<th>Collection period</th>
<th>Rainfall (cm.)</th>
<th>Tritium concentration (T.U.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Delhi</td>
<td>July 1961</td>
<td>19.8</td>
<td>65±12</td>
</tr>
<tr>
<td>28°35' N, 77°12' E</td>
<td>August 1961</td>
<td>58.3</td>
<td>75±18</td>
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<td></td>
<td>September 1961</td>
<td>7.4</td>
<td>30±12</td>
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<td></td>
<td>October 1961</td>
<td>8.6</td>
<td>30±12</td>
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<td></td>
<td>December 1961</td>
<td>0.8</td>
<td>40±12</td>
</tr>
<tr>
<td></td>
<td>January 1962</td>
<td>3.3</td>
<td>460±20</td>
</tr>
<tr>
<td></td>
<td>February 1962</td>
<td>1.3</td>
<td>900±28</td>
</tr>
<tr>
<td></td>
<td>March 1962</td>
<td>0.8</td>
<td>750±20</td>
</tr>
<tr>
<td></td>
<td>June 1962</td>
<td>2.8</td>
<td>340±21</td>
</tr>
<tr>
<td></td>
<td>July 1962</td>
<td>20.1</td>
<td>200±18</td>
</tr>
<tr>
<td></td>
<td>August, 1962</td>
<td>8.5</td>
<td>200±19</td>
</tr>
<tr>
<td></td>
<td>September 1962</td>
<td>18.3</td>
<td>120±18</td>
</tr>
<tr>
<td></td>
<td>June 1963</td>
<td>11.8</td>
<td>1180±22</td>
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<td></td>
<td>September 1963</td>
<td>27.8</td>
<td>295±18</td>
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<td></td>
<td>June 1964</td>
<td>2.5</td>
<td>450±17</td>
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<td></td>
<td>July 1964</td>
<td>53.8</td>
<td>600±20</td>
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<tr>
<td></td>
<td>September 1964</td>
<td>18.2</td>
<td>390±18</td>
</tr>
<tr>
<td>Ahmedabad</td>
<td>September 1962</td>
<td>17.0</td>
<td>110±12</td>
</tr>
<tr>
<td>23°04' N, 72°38' E</td>
<td>June 1963</td>
<td>3.9</td>
<td>660±16</td>
</tr>
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<td></td>
<td>July 1963</td>
<td>13.8</td>
<td>800±24</td>
</tr>
<tr>
<td></td>
<td>September 1963</td>
<td>35.1</td>
<td>360±24</td>
</tr>
<tr>
<td>Sampling site (Lat., Long.)</td>
<td>Collection period</td>
<td>Rainfall (cm.)</td>
<td>Tritium concentration (T.U.)</td>
</tr>
<tr>
<td>---------------------------</td>
<td>----------------------</td>
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<td>-----------------------------</td>
</tr>
<tr>
<td>Bombay</td>
<td>Rain on 23–6-1961</td>
<td>..</td>
<td>170±20</td>
</tr>
<tr>
<td>18°54' N, 72°49' E</td>
<td>Rain on 22–7-1961</td>
<td>..</td>
<td>70±12</td>
</tr>
<tr>
<td></td>
<td>Rain on 16–8-1961</td>
<td>..</td>
<td>12±10</td>
</tr>
<tr>
<td></td>
<td>June 1961</td>
<td>39·3</td>
<td>45±10</td>
</tr>
<tr>
<td></td>
<td>July 1961</td>
<td>111·0</td>
<td>20±8</td>
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<td>September 1961</td>
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<td>1±10</td>
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<td></td>
<td>October 1961</td>
<td>10·1</td>
<td>20±8</td>
</tr>
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<td></td>
<td>June 1962</td>
<td>43·3</td>
<td>90±18</td>
</tr>
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<td></td>
<td>July 1962</td>
<td>75·7</td>
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<td>August 1962</td>
<td>39·7</td>
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<td>45±18</td>
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<td></td>
<td>December 1962</td>
<td>5·1</td>
<td>70±16</td>
</tr>
<tr>
<td></td>
<td>June 1963</td>
<td>37·0</td>
<td>545±23</td>
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<td>July 1963</td>
<td>79·7</td>
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<td>100·4</td>
<td>345±18</td>
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<td></td>
<td>September 1963</td>
<td>33·0</td>
<td>170±17</td>
</tr>
<tr>
<td></td>
<td>October 1963</td>
<td>1·8</td>
<td>300±19</td>
</tr>
<tr>
<td></td>
<td>June 1964</td>
<td>65·5</td>
<td>160±22</td>
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<tr>
<td></td>
<td>July 1964</td>
<td>47·1</td>
<td>230±16</td>
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<td></td>
<td>August 1964</td>
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<td>130±16</td>
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<td>September 1964</td>
<td>25·8</td>
<td>110±16</td>
</tr>
<tr>
<td></td>
<td>July 1963</td>
<td>4·1</td>
<td>570±30</td>
</tr>
<tr>
<td>Madras</td>
<td>November 1959</td>
<td>11·3</td>
<td>290±32</td>
</tr>
<tr>
<td>13°00' N., 80°11' E</td>
<td>October 1960</td>
<td>16·7</td>
<td>250±20</td>
</tr>
<tr>
<td>Bangalore</td>
<td>July 1963</td>
<td>2·3</td>
<td>420±24</td>
</tr>
<tr>
<td>12°57' N, 77°38' E</td>
<td>November 1959</td>
<td>11·3</td>
<td>290±32</td>
</tr>
<tr>
<td>Ootacamund</td>
<td>October 1960</td>
<td>16·7</td>
<td>250±20</td>
</tr>
<tr>
<td>11°24' N, 76°44' E</td>
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</table>
Subsequently, very large injections of artificial \( H^3 \) were made during late 1962, primarily as a result of U.S.S.R. nuclear detonations. As no further testing was continued, the tritium levels dropped during 1964. We will examine these data, particularly for the periods 1963 and 1964, in relation to meteorological parameters relevant to the dispersion and removal of \( H^3 \).

To obtain a clearer picture of the geographical variation, the results in Table I have been plotted on a map of India in Fig. 1, for all the grid stations during 1962, 1963 and 1964. Several features immediately become apparent:

(i) The fallout during 1963 is appreciably higher than that in the preceding and subsequent years. The average tritium level during 1964 was about sixty per cent. of the corresponding value in 1963.

(ii) An inland gradient exists in \( H^3 \)-concentrations; tritium concentrations increase as one moves eastwards from the western coastline of India.

(iii) The tritium concentrations at Delhi are significantly lower than those of Srinagar; the subsequent decrease south of Delhi is much slower.

3. DISCUSSION OF RESULTS

Since artificial tritium was injected at high latitude and above 10 km. altitude, it is apparent that the observed features in the fallout of tritium discussed above are a consequence of several meteorological parameters, important amongst which are: (a) the stratosphere-troposphere exchange, (b) the meridional transport of air, (c) scavenging of \( H^3 \) from the atmosphere by wet precipitations and (d) reintroduction of \( H^3 \) in the atmosphere as a result of re-evaporation of precipitated water. There are several other factors which should also be considered, such as climatic fluctuations which may produce local anomalies and removal of tritium from air by molecular exchange occurring across the air-sea interface. In order to see whether the features depicted by our data represent a world-wide pattern or are merely peculiar to the Indian subcontinent, we have compared our data with those available from other latitudes. Furthermore, to assess the roles of re-evaporation and molecular exchange of \( H^3 \), we have also examined the world data on the fallout of \( Sr^{90} \), which was also injected in late 1962 in amounts far exceeding those present in the atmosphere due to previous injections. The fallout pattern of \( Sr^{90} \) is governed by most of the meteorological parameters discussed above except for one important difference; \( Sr^{90} \) is not reintroduced in the atmosphere by evaporation. Furthermore, it is not expected to be removed by molecular exchange across the air-sea interface. In contrast, these two factors are considered to be very important in case of tritium which
becomes a constituent of the gaseous and liquid water (Bolin, 1959; Eriksson, 1965).

We shall now examine the global data on H³ and Sr⁹⁰ to delineate the importance of the meteorological parameters in bringing about the observed fallout patterns.

A. Latitudinal Variation in the World-wide Fallout of H³ and Sr⁹⁰ during 1963

In considering the global fallout patterns, we will consider the data separately for three types of stations: Inland, Coastal and Island. This classification rests on the foregoing discussions and is made to evaluate the importance of variability of meteorological parameters and the role of molecular exchange (for H³ alone). The available data on the mean annual concentrations of H³ (Tables I and II; I.A.E.A. Tritium Lists, 3, 4 and 5) and Sr⁹⁰ (Hardy and Rivera, 1965; Vohra, 1965) during 1963 are plotted as a function of geographic latitude for the three station types in Figs. 2, 4, 6 and 7. The corresponding latitudinal variation in the deposition of H³ and Sr⁹⁰ during 1963 at inland stations, obtained by multiplying the mean concentration figures by the mean annual precipitation in different latitude belts (as given by Junge, 1963), is presented graphically in Figs. 3 and 5. We have not given the latitudinal variation in the deposition of Sr⁹⁰ at island or coastal stations since it has not yet been established whether it is meaningful to obtain representative zonal deposition by multiplying the mean concentration values by the corresponding annual zonal rainfall figures, in view of the very variable local meteorological conditions expected for such stations. In any case, the relative deposition of Sr⁹⁰ at the three station types can be assessed from Figs. 4, 6, and 7 which give the observed mean annual concentrations of Sr⁹⁰.

The data presented in Figs. 2 to 7 clearly show a general decrease in the concentration/deposition of both H³ and Sr⁹⁰ as a function of latitude. A normal practice so far has been to describe the latitude variation of fallout of H³ and other precipitable fission products by a continuous curve. However, as we see from Figs. 2, 3, 4 and 5 which give the latitudinal variation in the concentration/deposition of H³ and Sr⁹⁰ at inland stations, all values separately within the 70°–30° and 30°–0° belts seem to centre around certain mean values, within factors of less than two. The magnitude of scatter in these belts is of the same order as observed for stations lying only a few degrees apart. The scatter is thus presumably of a meteorological origin, and we therefore infer from the H³ and Sr⁹⁰ data that, separately in the 70°–30° and 30°–0° northern latitude belts, their deposition during 1963 was fairly uniform.
Fig. 2. Latitudinal variation of mean concentrations of H\(^{3}\) in precipitation at Inland Stations during 1963. The ordinate should be multiplied by 7.14 to obtain concentrations in d.p.m./litre.
Fig. 3. Latitudinal variation of mean deposition of H\(^{a}\) at Inland Stations during 1963. The ordinate should be multiplied by 6·7 x 10\(^6\) to obtain deposition values in atoms/cm\(^2\).
Fig. 4. Latitudinal variation of mean concentration of Sr$^{90}$ in precipitation at Inland Stations, during 1963. The ordinate should be multiplied by 33.9 to obtain concentrations in d.p.m./litre. Data plotted are from the following stations:

1. Fairbanks (Alaska, U.S.A.)
2. Argonne (Illinois, U.S.A.)
3. Intl. Falls (Minnesota)
4. Columbia (Missouri, U.S.A.)
5. Helena (Montana, U.S.A.)
6. Williston (N. Dakota, U.S.A.)
7. Vermillion (S. Dakota, U.S.A.)
8. Klagenfurt (Austria)
9. Milan (Italy)
10. Kikuyu (Kenya)
11. Nairobi (Kenya)
12. Salisbury (S. Africa)
13. Bogota (Colombia-S. America)
14. Adis Ababa (Ethiopia-Africa)
15. Columbia (S. Carolina, U.S.A.)
16. Formosa (Argentina)
17. Leopoldville (Congo-Africa)
18. Quito (Ecuador-S. America)
19. Teheran (Iran)
20. Florence (Italy)
21. Mexico City (Mexico)
22. Pretoria (S. Africa)
23. Srinagar (India)
24. Delhi (India)
25. Gangtok (Sikkim)
26. Nagpur (India)
27. Bangalore (India)
FIG. 5. Latitudinal variation of mean deposition of Sr\textsuperscript{90} at Inland Stations during 1963. The ordinate should be multiplied by 0.0339 to obtain deposition values in d.p.m./cm.\textsuperscript{2} "Key" for stations is the same as for Fig. 4.
Fig. 6. Latitudinal variation of mean concentration of Sr\(^{90}\) in precipitation at Coastal Stations during 1963. The ordinate should be multiplied by 33.9 to obtain d.p.m./litre. Data plotted are from the following stations:

1. San Francisco (California, U.S.A.)
2. New Orleans (Louisiana, U.S.A.)
3. Westwood (New Jersey, U.S.A.)
5. Medford (Oregon, U.S.A.)
6. Houston (Texas, U.S.A.)
7. Sterling (Virginia, U.S.A.)
8. Darwin (Australia)
9. Townsville (Australia)
10. Rio de Janeiro (Brazil)
11. New Foundland (Canada)
12. Wheelus AB (Libya)
13. Oslo (Norway)
14. Caracas (Venezuela)
15. Bangkoc (Thailand)
16. Miami (Florida, U.S.A.)
17. Palo Alto (California, U.S.A.)
18. Brisbane (Australia)
19. Belem (Brazil)
20. S. Jose des Campos (Brazil)
21. Colombo (Ceylon)
22. Beirut (Lebanon)
23. Ben Gashir (Libya)
24. Lagos (Nigeria)
25. Durban (S. Africa)
26. Calcutta (India)
27. Bombay (India)
Fig. 7. Latitudinal variation of mean concentration of Sr$^{90}$ in precipitation at Island Stations during 1963. The ordinate should be multiplied by $33.9 \times 10^{-6}$ to obtain d.p.m./litre. Data plotted are from the following stations:

A. Hilo (Hawaii, U.S.A.)
B. Lihue (Hawaii, U.S.A.)
C. Lajes Field (Azores)
D. Canton Island
E. Anderson AFB (Guam)
F. Keflavik (Iceland)
G. Iwo Jima
H. Misawa (Japan)
I. Tachikawa (Japan)
J. Johnston Island
K. Koror Island
L. Majuro Island
M. Main Island (Puerto Rico)
N. Wake Island
O. Wake Island
P. Tarawa (Gilbert Island)
Q. Kiliwik Island (Bermuda)
R. Singapore (Malaysia)
S. Cebu City (Philippines)
T. Yap Island
U. Palau Island
V. Eniwetok Atoll (Mili Atoll)
W. Hilo (Hawaii, U.S.A.)
X. Tainan (Taiwan)
Y. Taiwan (China)
Z. Hiroshima (Japan)

Fig. 6. Concentration of Sr$^{90}$ in precipitation (mc x mile$^{-2}$ x month$^{-1}$)
This interpretation is at variance with those of other investigators (cf. Machta, 1961; Thatcher and Payne, 1965) who have concluded that the deposition of precipitable fission products is well represented by a continuously variable function of latitude.

The above-mentioned features in the deposition of H\textsuperscript{3} and Sr\textsuperscript{90} are seemingly well supplemented by the data on nuclear weapon produced C\textsuperscript{14} during 1963 (Lal and Rama, 1966). Analogous to the case of H\textsuperscript{3} and Sr\textsuperscript{90}, the bomb-C\textsuperscript{14} also shows marked uniformity separately in the two latitude belts considered; stations situated within 5° or so around the 30° N latitude display both transitory and disturbed response, with a sudden marked decrease in the fallout as one proceeds south of about 30° latitude. The present analysis therefore seems to support the existence of a 30°-0° Hadley cell which receives fresh supply of H\textsuperscript{3} or Sr\textsuperscript{90} activities from the higher latitude air as a result of interactions (mixing) of air in the troposphere at about 30° latitude belt. We have not considered here the fallout data north of 70° N; the annual rainfall at 70°-90° latitude is fairly low and this probably explains why the observed deposition of Sr\textsuperscript{90} (Hardy and Rivera, 1965) is low in the polar regions. Our discussions of the global fallout are however not affected in any serious manner since the fractional area in the 90°-70° belt is quite small. Thus, we cannot comment on the extent or division of Hadley cells north of 30°. The data do not however contradict the existence of two Hadley cells comprising the 90°-60° and 60°-30° belts. In further discussions, we will however restrict to the latitudinal subdivisions, 70°-30° and 30°-0° for considering the global fallout characteristics of the radionuclides, H\textsuperscript{3} and Sr\textsuperscript{90}.

The observed values for the mean deposition of H\textsuperscript{3} and Sr\textsuperscript{90} during 1963 in the 70°-30°, and 30°-0° north latitudes are summarised in Table III, separately for the three station types considered. The geographical and latitudinal variation in their concentration/deposition derived from data in Table II are given in Tables IV and V; the figures on annual deposition of H\textsuperscript{3} and Sr\textsuperscript{90}, based on coastal and island stations are not given, for reasons discussed earlier.

We will now evaluate the rate at which air from high latitudes must exchange with the 30°-0° Hadley cell to account for the observed deposition of H\textsuperscript{3} and Sr\textsuperscript{90}, following a simple box-model treatment for the transport and mixing of air in the troposphere.

**B. Exchange of Air Across the 30° Latitude Belt**

Lal and Rama (1966) have carried out a detailed analysis of the data on global dispersion of the radiocarbon from the Russian tests in terms of a
model which invokes the existence of two internally well-mixed zones in the northern troposphere with boundary at 30° latitude, and have tried to evaluate the rate of mixing of air between them. Their analysis revealed that the mixing between the two tropical cells (30°-0° N and 0°-30° S) is slow compared to that between the two cells, 90°-30° and 30°-0°, in the same hemisphere. In view of this we will consider a simplified two cell tropospheric-model for the northern hemisphere as shown in Fig. 8. In this model, the exchange of air across the equator is not considered. This approach seems to be justified for precipitable nuclides, e.g., H³ and Sr⁹⁰, but is not applicable for the case of C¹⁴. The mixing parameter K₁-₂ denotes the fraction of air exchanged monthly from cell 1 (90°-30° N) to cell 2 (30°-0° N). Although the analysis of Lal and Rama (1966) reveals that K₁-₂ varies markedly with season, we shall concern ourselves only with the average annual value. The material which descends into cell 1 from the stratosphere is partly transported to the tropics by north-south mixing and is partly deposited in cell 1 itself by wet precipitation. In the case of H³, a part of the deposited amount is reinjected in the troposphere by re-evaporation (ocean water or the plant and soil water for the oceanic and continental areas respectively). Due to its increased life in the troposphere, a larger fraction, compared to that for Sr⁹⁰ and Cs¹³⁷, can thus be transported to the tropical cell (cell 2). For inland stations,

### Table III

*Annual Concentration/Deposition of H³ and Sr⁹⁰ during 1963*

<table>
<thead>
<tr>
<th>Latitude belt</th>
<th>Station type</th>
<th>Tritium Concentration (T.U.)</th>
<th>Tritium Deposition (d.p.m./cm²)</th>
<th>Strontium 90 Concentration (d.p.m./l)</th>
<th>Strontium 90 Deposition (d.p.m./cm²)</th>
<th>Concentration ratio H³ (T.U.)/Sr⁹⁰ (d.p.m./l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>70°-30° North</td>
<td>Inland</td>
<td>2790 (11)*</td>
<td>1640 (11)</td>
<td>50 (13)</td>
<td>4·4 (13)</td>
<td>56</td>
</tr>
<tr>
<td></td>
<td>Coastal</td>
<td>1460 (9)</td>
<td>38 (12)</td>
<td></td>
<td>38</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Island</td>
<td>1080 (2)</td>
<td>23 (6)</td>
<td></td>
<td>47</td>
<td></td>
</tr>
<tr>
<td>30°-0° North</td>
<td>Inland</td>
<td>630 (9)</td>
<td>470 (9)</td>
<td>10·5 (6)</td>
<td>1·0 (6)</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>Coastal</td>
<td>420 (6)</td>
<td>9·5 (9)</td>
<td></td>
<td>44</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Island</td>
<td>170 (5)</td>
<td>7·1 (17)</td>
<td></td>
<td>24</td>
<td></td>
</tr>
</tbody>
</table>

*Figures within parenthesis denote the number of stations on which the result is based.*
**Measurement of Tritium Activity in Natural Waters—II**

**TABLE IV**

*Relative Geographical Variation in the Annual Concentrations of $^{3}H$ and $^{90}Sr$ during 1963*

<table>
<thead>
<tr>
<th>Latitude belt</th>
<th>Station types compared</th>
<th>Relative concentration</th>
<th>Tritium</th>
<th>Strontium-90</th>
</tr>
</thead>
<tbody>
<tr>
<td>$70^\circ-30^\circ$ (North)</td>
<td>Inland: Coastal</td>
<td>1.9 *</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Inland: Island</td>
<td>(2.6)*</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>$30^\circ-0^\circ$ (North)</td>
<td>Inland: Coastal</td>
<td>1.5</td>
<td>1.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Inland: Island</td>
<td>3.7</td>
<td>1.5</td>
<td></td>
</tr>
</tbody>
</table>

* Based on insufficient data.

The observed values of the relative deposition in the $70^\circ-30^\circ$ N and $30^\circ-0^\circ$ N latitude belts are $4.4 \pm 1.0$ and $3.5 \pm 0.7$ for $^{90}Sr$ and $^{3}H$ respectively (see Table V). The lower value observed for $^{3}H$ seems to indicate the role of re-evaporation from the continents. The errors however are too large to establish the case conclusively.

**TABLE V**

*Relative Variations in the Concentration/Deposition of $^{3}H$ and $^{90}Sr$ during 1963 in the $70^\circ-30^\circ$ N and $30^\circ-0^\circ$ N Latitude Belts*

<table>
<thead>
<tr>
<th>Station type</th>
<th>Relative concentration/deposition ($70^\circ-30^\circ$ N: $30^\circ-0^\circ$ N)</th>
<th>Tritium</th>
<th>Strontium-90</th>
</tr>
</thead>
<tbody>
<tr>
<td>Concentration</td>
<td>Deposition</td>
<td>Concentration</td>
<td>Deposition</td>
</tr>
<tr>
<td>Inland</td>
<td>4.4</td>
<td>3.5</td>
<td>4.8</td>
</tr>
<tr>
<td>Coastal</td>
<td>3.5</td>
<td>..</td>
<td>4.0</td>
</tr>
<tr>
<td>Island</td>
<td>(6.4)*</td>
<td>..</td>
<td>3.2</td>
</tr>
</tbody>
</table>

* Based on insufficient data.
For precipitable nuclides like Sr\(^{90}\) and Cs\(^{137}\), the ratio of fallouts in the high latitude cell (cell 1) to that in the tropical cell (cell 2) can be shown to be given by:

\[
\frac{F(\text{cell 1})}{F(\text{cell 2})} = M_2^{-1} + \frac{K_F}{K_{1-2}}
\]  

where \(M_2^{-1}\) is the ratio of masses of air in cells 1 and 2, taken to be 0.85 from meteorological data; \(F\) and \(K_F\) respectively denote the fall out and the fraction removed per month by precipitation. \(K_F\) is assumed to be identical in the two cells. In case of H\(^3\), another variable \(K'_F\) can be introduced to account for the re-evaporation. It is generally believed that the mean removal time of activities like Sr\(^{90}\), Cs\(^{137}\), H\(^3\) from the troposphere by precipitation is of the order of 3–4 weeks, i.e., \(K_F = 1.3\) to 1 month\(^{-1}\). Taking the mean ratio of fallouts of Sr\(^{90}\) and H\(^3\) in cells 1 and 2 to be 4 \pm 1, equation (1) yields a value of 0.35\(^\pm\)0.1 (month\(^{-1}\)) for \(K_{1-2}\). This figure is in good agreement with the value deduced by Lal and Rama (1966) for \(K_{1-2}\) on the basis of observations of tropospheric dispersion of C\(^{14}\); their analysis reveals a strong seasonal dependence in the rate of meridional mixing across the 30° latitude belt. The value of the exchange coefficient, \(K_{1-2}\), is found to vary according to the relation:

\[
K_{1-2} = 1 + 0.7 \sin \left(\frac{2\pi t}{\tau} + \frac{\pi}{2}\right)
\]  

where \(t\) is expressed in months and \(\tau = 12\). The value of 0.35\(^\pm\)0.1 (month\(^{-1}\)) derived from the H\(^3\) and Sr\(^{90}\) data corresponds to the months of May to
August in equation (2). The agreement between the two analyses is therefore very good, considering that most of the fall out of $H^3$ and Sr$^{90}$ occurs during May–August.

In the above calculations we took the mean observed ratio of $H^3$ and Sr$^{90}$ fallout in the high latitude and tropical cells. As we will see in the following sub-section, the re-evaporation of water appreciably increases the lifetime of $H^3$ on the continents, thus implying that we should have taken the higher value of $4.4 \pm 1.0$ observed for Sr$^{90}$, for evaluating the mean value of $K_{1-2}$. This would yield about 15% lower value for $K_{1-2}$ and the conclusions remain unchanged.

C. Seasonal Variation in the Fallout of $H^3$ and Sr$^{90}$

Several authors have studied the seasonal effects in the wet fallout of $H^3$ and Sr$^{90}$. Reference may be made to the analysis carried out by Libby (1961), who examined the northern hemisphere $H^3$ and Sr$^{90}$ data during 1959. On the basis of the observed parallelism between concentrations of $H^3$ and Sr$^{90}$, he showed that both these nuclides resulted primarily from the October 1958 Soviet tests, and argued that the results also implied an essentially identical mechanism of removal of these activities from the stratosphere into the troposphere. A similar comparison of $H^3$ and Sr$^{90}$ fallout at Bedford, Massachusetts, however, yielded an apparently quite non-similar seasonal fallout pattern and Libby (1961) concluded that the result indicated a dissimilar tropospheric removal mechanism for the two isotopes.

In view of the largest yet observed concentrations of $H^3$ and Sr$^{90}$ in wet precipitations during 1963, we have examined the seasonal variations of these nuclides. Our analysis for this study was restricted to inland stations to avoid consideration of effects due to loss of $H^3$ by air-sea exchange, regarding which our knowledge is very limited at present. Also to further eliminate the day-to-day meteorological fluctuations, we have considered only the monthly averages for their concentrations at several inland stations situated in the 30°–70°N latitude band. The computed average values for $H^3$ (I.A.E.A. Tritium Lists Nos. 3, 4, and 5) and Sr$^{90}$ (Hardy and Rivera, 1965) are plotted in Fig. 9 for 1963; the inland stations selected for this purpose are listed in the caption of Fig. 9.

It can be seen from Fig. 9 that during the months February–May 1963, the $H^3$ and Sr$^{90}$ concentrations follow each other quite closely: 40 T.U. concentration of $H^3$ corresponds to 1 d.p.m./l Sr$^{90}$ during these months. Subsequently, however, the two curves depart markedly. During June to September 1963, there is a fair parallelism between the two curves but the
constant of proportionality is about 70 instead of 40. We wish now to examine the implications of these observations in terms of tropospheric removal mechanisms of these isotopes.

![Graph](image)

**Fig. 9.** Seasonal variation of mean monthly concentrations of $H^3$ and $Sr^{90}$ in wet precipitations at *Inland Stations* in $30^\circ$-$70^\circ$ N. belt, during 1963. Stations for $H^3$ and $Sr^{90}$ data are listed below separately:

- **$H^3$:** Stuttgart, Vienna, Ankara, Portsmouth, Edmonton, Chicago and Flagstaff.
- **$Sr^{90}$:** Fairbanks, Argonne, Intl. Falls, Columbia (Missouri), Helena, Williston, Vermillion, Klagenfurt, Milan, Columbia (S. Carolina) and Florence.

The fact that the seasonal variations in the fallout of $H^3$ and $Sr^{90}$ exhibit a remarkable parallelism till May straightaway rules out the hypothesis that any significant relative seasonal changes in their *monthly fallout* occur due to any dissimilarities in their wash-out mechanism. In individual showers, $H^3$ and $Sr^{90}$ ratios may vary due to the fact that removal (scavenging) efficiencies for $H^3$ which forms part of gaseous and liquid water may be different from that of aerosol borne $Sr^{90}$. However since the residence time of water...
and aerosols in the lower troposphere is of the order of 1 week only (Junge, 1963), in contrast to the mean residence time of 3-4 weeks in the troposphere (H³ or Sr⁹⁰), it is clear that any dissimilarities in the wash-out mechanism will not alter their mean monthly fallout rates which must equalise their steady state input due to mixing across the tropopause. This last-mentioned equilibrium must exist for the two isotopes within periods of the order of 1 month, though not necessarily simultaneously. Thus the parallelism observed during February–May would be expected for 1963 which represents H³ and Sr⁹⁰ released in the stratosphere during late 1962.

Clearly therefore the marked changes in H³ and Sr⁹⁰ concentrations following May 1963 must be ascribed to some other phenomenon which though operative during earlier months does not lead to any appreciable modifications in the seasonal march of wet fall out. The only process which seems to be important in the present context is the reinjection of H³ due to evaporation and plant transpiration. This process increases the apparent lifetime of H³ alone in the troposphere as Sr⁹⁰ is not expected to be reinjected once it is removed by wet precipitation. If this process produces the effect as observed in Fig. 9, one must be able to explain the lack of any influence during earlier months. Qualitatively this is quite in accord with expectations: during the months of March–May when the stratospheric injection is increasing rapidly, the contribution due to re-evaporation should be small compared to that introduced by downward mixing of stratospheric air, and therefore the fallouts of both H³ and Sr⁹⁰ may be expected to vary identically, or more precisely remain proportional to their concentration ratio in the lower stratospheric air in the high latitude cell which feeds the tropospheric air. The increase in H³ due to re-evaporation is expected to become appreciable only when the stratospheric input is declining or becomes small. The observed pattern of H³ and Sr⁹⁰ concentrations during October–December probably arises due to a marked decrease in the rate of evapotranspiration during these months (Benton and Estoque, 1954).

In order to obtain a more quantitative idea of the role of evaporation and plant transpiration to the injection of H³ in the troposphere, we have set up an oversimplified model as shown in Fig. 10. Here we consider the balance between the injection and removal of H³ and Sr⁹⁰ in a vertical column in the troposphere at an inland station. Neglecting any net loss of H³ or Sr⁹⁰ by horizontal transport, one obtains the following differential equations for the time changes in the total amounts T and S of H³ and Sr⁹⁰ respectively in the vertical air column:
Following earlier discussions, the fallouts of H\textsuperscript{3} and Sr\textsuperscript{90} are expected to be related to T and S by the relations:

\[
K_F(T)_t = P(Tp)_t
\]  
\[
K_F(S)_t = P(Sp)_t
\]

where,

- \(Q_T\) = net rate of introduction of H\textsuperscript{3} from aloft by turbulent mixing,
- \(Q_S\) = net rate of introduction of Sr\textsuperscript{90} from aloft by turbulent mixing,
- \(K_F\) = rate of removal of precipitable activities from the column,
- \(P\) = rate of precipitation of water,
- \(E\) = rate of total evaporation of water,
- \(T_P\) = concentration of H\textsuperscript{3} in precipitation,
- \(S_P\) = concentration of Sr\textsuperscript{90} in precipitation,
- \(T_E\) = concentration of H\textsuperscript{3} in evaporated water.

Equations (3) to (6) allow one to study the experimentally observed time variations in the concentrations of H\textsuperscript{3} and Sr\textsuperscript{90} in terms of contributions due to evapotranspiration.

---

**Fig. 10.** Model adopted for evaluating the importance of evapotranspiration in the balance of H\textsuperscript{3} in the troposphere.
to reinjected H$^3$; it may be noted here that but for the term ET in equation (3), one would not expect any relative seasonal changes in $T_P$ and $T_S$ on the simplified model adopted here. We will now consider the case when $Q_T$ and $Q_S$ are both changing exponentially with time as $e^{\kappa t}$ and that the ratio of concentrations of H$^3$ and Sr$^{90}$ in upper tropospheric air, which is assumed to remain constant, is $R$. For simplicity, we assume that the ratio $T_E/T_P$ remains invariant with time. Then at any sufficiently late time (i.e., a period $> 1/K_F$ after $Q_T$ and $Q_S$ start increasing or decreasing), both H$^3$ and Sr$^{90}$ concentrations are expected to change essentially as $e^{\kappa t}$ provided $K_F > K$ which is applicable in the present case, separately for the periods February to May and June to September. The ratio of H$^3$ and Sr$^{90}$ concentration for precipitation in either of the cases when $K$ is positive or negative is then given by:

$$\left[\frac{T_P}{S_P}\right]_t \approx \frac{R (K + K_F)}{K + K_F \left(1 - \frac{E}{P} \cdot \frac{T_E}{T_P}\right)} \tag{7}$$

and the delay in time, $\triangle t$, after which H$^3$ concentrations reach a value equal to RS$^0$ is given by:

$$e^{\kappa \triangle t} \approx \frac{K + K_F \left(1 - \frac{E}{P} \cdot \frac{T_E}{T_P}\right)}{(K + K_F)} \tag{8}$$

Now considering the period June–September, 1963, from Fig. 9 we observe that $K = -0.35 \pm 0.05$ and the factor by which the H$^3$/Sr$^{90}$ ratio is enhanced over previous months is about $2 \pm 0.5$; the time delay, $\triangle t$, is about 2 months. These observations are consistent with a value of about 0.35 for $(E/P \cdot T_E/T_P)$. Taking a plausible range of values of 0.5 to 1 for E/P, the corresponding values of $T_E/T_P$ are 0.7–0.35; the actual value would be higher because the ratio E/P, assumed to be constant in the present calculations, in fact decreases between the months of June and September, and also because loss of tritium by horizontal transport to the oceans has not been considered in the model.

It can therefore be concluded that the value of $T_E/T_P$ always remains close to unity pointing to a quick re-evaporation of the precipitated water. Further, as the atmospheric inventory of tritium reduces by the combined effects of precipitation and reduced stratospheric-tropospheric mixing during October–December months, the value of $T_P$ (and also $S_P$) decreases to significantly low values (cf. Fig. 9). This also indicates that the water which evaporates is that which precipitated a short while ago and had not mixed
with older water present in the soil. This result is in general agreement with the inferences drawn by Eriksson (1965) and Zimmermann et al. (1965). The downward movement of water in soil seems to be fairly well-layered with freshly precipitated water essentially pushing the older water downwards. From the data in Fig. 9, we estimate that the total effective depth of water to which the freshly precipitated water mixes in the soil lies somewhere between 10 and 50 cm.

The above analysis also yields that the ratio of $H^3$ and Sr$^{90}$ concentrations in upper tropospheric air (or the lower stratospheric air) is about 40 (T.U./d.p.m. litre$^{-1}$) since it is the ratio observed till the end of May when both $H^3$ and Sr$^{90}$ concentrations were rising simultaneously. During this period $K$ is positive, and $T_R/T_P$ is expected to be $<0.5$. The value of $E/P$ is expected to be $\leq 0.5$ (Benton and Estoque, 1954) during winter and spring months. An examination of equation (7) then clearly shows that $T_P/S_P$ is expected not to differ appreciably from $R$. As a further confirmation of this deduction we note that the ratio $H^3/Sr^{90}$ at island and coastal stations is observed to be about 40 (Table III), both in the 70°–30° and 30°–0° belts. The corresponding mean ratio at inland stations, on an annual basis (Fig. 9, Table III), is ~ 60 which indicates that reinjection of tritium due to evapotranspiration increases the “apparent” lifetime of tritium in the troposphere significantly; the apparent deposition of $H^3$ is larger by about 50%.

Observations similar to the one in Fig. 9 have been made earlier; see for example Libby (1961) who compared $H^3$ and Sr$^{90}$ fallout at Bedford. The mean ratio of deposition of $H^3$ and Sr$^{90}$, averaged over period of weeks, was found to be significantly higher during May–July 1959 compared to the values observed during February–April 1959. These differences were attributed by Libby (1961) to be due to non-similar removal mechanisms of the two isotopes. This may be partly true; our analysis however clearly shows that if monthly averages of several stations are considered, i.e., if day-to-day local meteorological factors are cancelled out, the fallout of $H^3$ differs from that of Sr$^{90}$ primarily because of reinjection of $H^3$ in the troposphere.

The model adopted (Fig. 10) is very oversimplified indeed. The troposphere must be considered to be made of at least two boxes as evidenced from the fact that even at the 30°–0° belt where direct stratosphere–troposphere exchange is negligible or absent, the island and coastal stations show a mean concentration ratio, $H^3$ (T.U.)/Sr$^{90}$ (d.p.m./l) of about 40, i.e., the same as at higher latitudes. Thus the data indicate that the residence time of air in the upper troposphere is comparable to or larger than the time scales of mean zonal mixing and that the lower tropospheric air, where mixing and
scavenging processes occur on time scales of the order of a week, should be
considered as a separate reservoir in the box-model approach. The data on
Bi$^{81}$ and Pb$^{82}$ in rains and tropospheric air are quite in accord with a two-
layer tropospheric mixing model (Bhandari, Lal and Rama, in preparation).

D. Environmental Variation in the Fallout of H$^3$ and Sr$^{90}$

So far we have examined the 1963 H$^3$ and Sr$^{90}$ data with a view to study
the characteristics of meridional tropospheric mixing in the northern hemi-
sphere and the role of evaporation and plant transpiration in the tritium-
balance. All data considered so far referred chiefly to inland stations. We
will now examine the H$^3$ data at island and coastal stations to evaluate the
importance of air-sea molecular exchange processes in scavenging the atmo-
spheric tritium. Again, inasmuch as Sr$^{90}$ is not reinjected in the troposphere
by evaporation, it is also not expected to be removed from the boundary air
layer by molecular exchange. We will therefore consider Sr$^{90}$ data as well
to eliminate any meteorological factors which may be important in deciding
the concentrations of the two isotopes at island, coastal or inland stations.

The available data for H$^3$ and Sr$^{90}$ concentrations/deposition for 1963
are summarised in Table III for the three station types. In order to bring
out the features in the environmental variation, the data of Table III are
re-arranged in Table IV in terms of ratios of concentrations at different station
types. These results lead to the following conclusions:

(i) Mean concentration of Sr$^{90}$ at inland stations is higher by a factor
of about 1.6 (values range between 1.1 and 2.2) compared to those at island
and coastal stations in the same latitude band.

(ii) Mean concentration of H$^3$ at inland stations is higher by a factor of
about 2.6 (values range between 1.5 and 3.7) compared to those at coastal
and island stations. The observed concentrations at island stations seem to
be smaller compared to coastal stations by about a factor of two. The available
data are however not precise enough to make a closer estimate.

Lower concentration of Sr$^{90}$ at island and coastal stations can probably
be understood in terms of local meteorological causes such as quick precipi-
tation of relatively large amounts of locally evaporated water in these areas.
The fact that environmental precipitation factors are significantly larger in
case of H$^3$ can be largely accounted for by the re-evaporation effect found
to be important at inland stations (see section C). We deduced that the
re-evaporation of H$^3$ from the continents enhances the mean annual concen-
tration at inland stations by a factor of about 1.5; this is sufficient to account
for essentially the entire difference observed between concentrations of H₃ and Sr⁹⁰ at the *island* (and *coastal*) and *inland* stations. Our analysis therefore does not support the hypothesis of any appreciable scavenging of H₃ from air by molecular exchange process; from the available data we conclude that less than 30% of tropospheric tritium is removed over oceans by this process.

The observed lower fallout of Sr⁹⁰ at *island* and *coastal* stations deserves some comments. Firstly, if we had considered the H₃ data only, we would have reached the conclusion that molecular exchange removes appreciable H₃ from the air over the oceans. (cf. Eriksson 1965). Secondly, it is of considerable interest to see the mechanism(s), outside of molecular exchange process, for the observed approximately 60% lower fall out of Sr⁹⁰ at *island* and *coastal* stations; the same mechanism would also deplete H₃ over oceanic areas. Two possibilities are (i) removal from the boundary air layer by sea-spray and jet action, (ii) relatively larger dilution of the precipitated water or more precisely a quicker turnover of evaporated ocean water. Apparently, the second process seems to be a more plausible one, but it is difficult to assess the relative importance of these processes. Furthermore, since it is quite clear from fundamental principles that molecular exchange must deplete the tritium in the boundary layer, it seems quite natural to deduce that either of these processes deplete the inventory of tritium sufficiently so that the role of molecular exchange is not discernible.

Another alternative which may be considered is to postulate a higher rainfall over oceans. In fact from the analysis of natural (pre-bomb) H₃ data, Libby has deduced a value of 250 cm./yr. for the mean precipitation over the Pacific and Atlantic Oceans in the 30°–50° N latitude belt; the corresponding excess rainfall of about 170 cm./yr. over oceans, compared to land, has been attributed by him to ocean spray (Libby, 1961). As higher rainfall would clearly reduce the concentrations of H₃ and Sr⁹⁰ correspondingly, this could well be the case. Eriksson (1965) has criticized this on the grounds that thousands of various measurements indicate a much lower rainfall figure, about 90 cm./yr. Nevertheless the fact is that the concentrations of both H₃ and Sr⁹⁰ are significantly lower at *island* stations due to a non-molecular transfer process, and if the mean annual rainfall is deduced from any balance considerations, a higher value will certainly result. Thus, if direct precipitation measurements over oceanic areas do yield values comparable to that over land, either of the two possibilities discussed in the preceding paragraph must be considered.
E. Comparison of Global Fallout during 1963 and 1964

It was discussed in Section 2 that during 1964, the H₃ concentrations in the Indian subcontinent reduced to a value of about 60 per cent. of those in 1963. If the large-scale weather patterns do not change from year to year, one should expect a similar reduction in all regions, not only for H₃ but for other precipitable nuclides also, e.g., Sr⁹⁰. That in fact is roughly the case is evident from Table VI which lists the mean annual relative 1964: 1963 H₃ and Sr⁹⁰ figures for the northern hemisphere for the three station types considered. (Sources of data for 1964 are the same as quoted in the text for the year 1963.) A reduction of 40% over one year's duration, in the absence of new weapon tests, indicates an effective stratospheric storage time of about two years. This figure is in good agreement with the value concurrently believed to hold for the bulk stratosphere on the basis of previous studies of fission products (cf. Bjornerstedt et al., 1963).

<table>
<thead>
<tr>
<th>Latitude belt</th>
<th>Station type</th>
<th>Relative concentration (1964: 1963)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Tritium</td>
</tr>
<tr>
<td>70°–30°</td>
<td>Inland</td>
<td>0.64</td>
</tr>
<tr>
<td>(North)</td>
<td>Coastal</td>
<td>0.52</td>
</tr>
<tr>
<td></td>
<td>Island</td>
<td></td>
</tr>
<tr>
<td>30°–0°</td>
<td>Inland</td>
<td>0.70</td>
</tr>
<tr>
<td>(North)</td>
<td>Coastal</td>
<td>0.44</td>
</tr>
<tr>
<td></td>
<td>Island</td>
<td></td>
</tr>
</tbody>
</table>

* Based on insufficient data.

4. Conclusions

The features of H₃ fallout observed for the Indian subcontinent form a part of the general global features and are a consequence of many meteoro-
logical variables. The observed sharp transition in the fallout at about 30°
latitude is interpretable in terms of weak interaction between the 90°–30°
and 30°–0° tropospheric Hadley cells during the mid-months of the year,
when most of the stratosphere-troposphere exchange occurs.

The present analysis of the relative variations in the fallout of H³ and
Sr⁹⁰ at continental and oceanic stations in the northern hemisphere has led
to several useful conclusions:

1. The apparent lifetime of H³ at inland stations is considerably
increased due to evapotranspiration; on an annual basis, about 50% of the
tritium is redeposited by precipitation. The implications of the data to
“mixing” of precipitated water with the soil water are discussed.

2. The mean concentrations of Sr⁹⁰ and H³ at island and coastal stations
are lower than those observed at inland stations by factors of 1.6 and 2.6
respectively. If one takes into account the apparent increase in the fallout
of H³ at inland stations due to evapotranspiration, then the relative environ-
mental depletion factors, (oceanic, vs. land area) for the two isotopes are
nearly the same. [The concentrations of (H³ and Sr⁹⁰) are lowest at island
stations; values at coastal stations seem to lie in the middle.] The data
indicate that over the oceans, less than 30% fallout of tritium is due to air-sea
molecular-exchange processes. This result is in disagreement with the
conclusions arrived at earlier (Eriksson, 1965; Bolin, 1959). According
to Eriksson, about 66% of the tritium is removed by the molecular-exchange
process occurring in the boundary layer (Eriksson, 1965).

The implications of the observed lower concentrations of Sr⁹⁰ at island
(and coastal) stations are discussed.

3. During 1963–64, the apparent stratospheric residence time of H³
and Sr⁹⁰, injected by U.S.S.R. nuclear weapons’ testing in the stratosphere
during late 1962, was about 2 years.

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