

HEALTH AND SAFETY LABORATORY  
ENVIRONMENTAL QUARTERLY

June 1, 1977 through September 1, 1977

Prepared by  
Edward P. Hardy, Jr.  
Environmental Studies DivisionNOTICE

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ENVIRONMENTAL QUARTERLY  
October 1, 1977ABSTRACT

This report presents current information from the HASL environmental programs, the Australian Radiation Laboratory, the Air Resources Laboratories of NOAA, the Air Monitoring Section of the Bhabha Atomic Research Centre in Bombay, India and the National Radiation Laboratory in New Zealand. The initial section consists of interpretive reports and notes on background corrections for  $^{90}\text{Sr}$  in ion-exchange resin used in the Australian fallout network, corrections to previously reported  $\text{N}_2\text{O}$  concentrations in the stratosphere, trace metal concentrations in a marine sediment as measured by five laboratories, an estimate of maximum credible atmospheric radioactivity concentrations from nuclear tests, strontium-90 concentrations in human bone in New York City and San Francisco through 1976, and worldwide deposition of  $^{90}\text{Sr}$  through 1976. Subsequent sections include tabulations of radionuclide and stable lead concentrations in surface air; strontium-90 in deposition, milk, diet and tapwater; fallout and atmospheric radioactivity measurements in India and environmental radioactivity measurements in New Zealand. A bibliography of recent publications related to environmental studies is also presented.

Preceding reports in this series:

<u>Year</u>	<u>HASL Reports Nos.</u>
1958	42, 51
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1960	77, 84, 88, 95
1961	105, 111, 113, 115
1962	117, 122, 127, 131
1963	132, 135, 138, 140
1964	142, 144, 146, 149
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1977	315, 318, 321

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## INTRODUCTION

Every three months, the Health and Safety Laboratory issues a report summarizing current environmental data obtained at HASL. This report, the latest in the series, contains information that became available during the period from June 1, 1977 to September 1, 1977. The next report is scheduled for publication January 1, 1978. Preceding reports in the series, starting with HASL-42, "Environmental Contamination from Weapons Tests", and continuing with HASL-328 (this report), may be purchased from the National Technical Information Service, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia 22161. A complete listing of these quarterly reports is given on the abstract page of this report.

To give a more complete picture of the current fallout situation and to provide a medium for rapid publication of radionuclide and non-nuclear pollutant data, these quarterly reports often contain information from other laboratories and programs, some of which are not part of the general ERDA program. To assist in developing, as rapidly as possible, provisional interpretations of the data special interpretive reports and notes prepared by scientists working in the environmental field are also included from time to time. Many of these scientists are associated in some way with the general ERDA program. Information developed outside HASL is identified as such and is gratefully acknowledged by the Laboratory. In this report, data from the Australian Radiation Laboratory, the Air Resources Laboratories of NOAA, the Air Monitoring Section of the Bhabha Atomic Research Centre in India and the National Radiation Laboratory in New Zealand, are presented.

A portion of the analyses either have been or are being carried out by commercial laboratories under contract to the HASL Environmental Studies Division. The results of these analyses are reported as part of HASL's regular environmental program. The contractor analytical laboratories which provided data are Nuclear Science and Engineering Corp., Pittsburgh, PA; Isotopes, Inc., Westwood, NJ; Radiochemistry, Inc., Louisville, KY; LFE Environmental Analysis Labs., Richmond, CA; Controls for Radiation, Inc., Cambridge, MA; Hazleton-Nuclear Science Corp., Palo Alto, CA (now Teledyne Isotopes Palo Alto Labs.); Food Chemical & Research Labs., Inc., Seattle, WA; Custom Nuclear Co., Mountainview, CA; Ledoux and Co., Teaneck, NJ; and U. S. Testing Co., Richland, WA.

This report is divided into four main parts:

1. Interpretive Reports and Notes
2. HASL Environmental Program Data
3. Data from Sources Other than HASL
4. Recent Publications Related to Environmental Studies

PART I  
INTERPRETIVE REPORTS AND NOTES



STRONTIUM-90 IN ION-EXCHANGE RESIN USED IN THE  
AUSTRALIAN FIEFS NETWORK

A statistical analysis of strontium-90 data from measurements reported by HASL on unexposed ion-exchange resin from Australian fallout stations and on HASL quality control blanks.

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1. Estimation of the Strontium-90 Activity in Ion-Exchange Resin before Exposure to Fallout

In order to determine monthly strontium-90 fallout deposited at the eight Australian FIEFS\* stations, account must be taken of the level of strontium-90 contamination of the ion-exchange resin as prepared for use in the FIEFS. This procedure has always been important in monitoring strontium-90 fallout deposit in Australia because the level of strontium-90 contamination of ion-exchange resin, supplied by manufacturers in the Northern Hemisphere, has remained of the same order of magnitude as the monthly fallout deposit in the Southern Hemisphere.

Each shipment of ion-exchange resin received from the manufacturer\*\* is treated to reduce ionic contamination, mixed thoroughly and then divided into separate lots for use month-by-month over the ensuing years. The separate monthly lots are protected against introduction of strontium-90 contamination before use. Estimates of the strontium-90 activity of the ion-exchange resin, as prepared for use in the FIEFS, are obtained by

- analysis of samples from the resin bulk, extracted before separation into monthly lots, to gain a measure of the level of strontium-90 activity before the shipment is introduced into service and
- analysis by HASL of one or more samples of unused resin taken from each monthly lot in turn, as this is brought into use with the FIEFS, and submitted as blinds to HASL with the monthly set of eight fallout samples.

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\*FIEFS is an acronym for the Funnel Ion-Exchange Fallout Sampler employed in the Australian fallout monitoring network (Devlin et al, 1971).

\*\*Permutit cation exchange resin, Zerolit 225 (H) commercial grade; 100 kg of resin, as a narrow column, was washed with 500 kg of 3N HCl and 400 kg of demineralized water.

Surveillance of the analytical procedures is exercised by HASL by the addition of quality control materials, including blanks, with the monthly sets of fallout samples.

Throughout use of ion-exchange resin from a shipment, a single value is adopted as the currently available best estimate of the strontium-90 activity of the unexposed resin. At the outset, this value derives from the measurements made before the resin was brought into service; after that, the value is regularly reviewed in the light of the accumulating data from the analyses by HASL. Revision of the value leads to recalculation of all previous results on monthly fallout deposits obtained using resin from that particular shipment. The procedure is continued until the entire shipment of resin is exhausted. A final review is then made of all data on the unexposed resin and final results are calculated, and reported, for all of the monthly fallout deposits determined using the resin.

The present paper records the statistical analysis of accumulated data on strontium-90 activities reported by HASL

- for unexposed ion-exchange resins measured with the monthly sets of FIEFS samples for the period March 1973 to October 1976, all derived from the one shipment of resin, and
- for HASL quality control blanks, measured with the same monthly sets of FIEFS samples.

The measurement results are summarized in Table 1 and their frequency distributions are depicted in Figure 1. These results include the activity of chemical reagents used in the analyses (Harley, 1977).

## 2. Statistical Tests

Standard statistical tests - the one-way analysis of variance to test the equality of group means and Bartlett's test for the equality of group variances - were applied to the data for unexposed ion-exchange resin, discussed in section 3, and to the results for HASL quality control blanks, discussed in section 4.

Statistical arguments were applied to the frequency distributions of the measurement results for the unexposed ion-exchange resin, and for the HASL quality control blanks, to show that the lower tails of both distributions are anomalous; this is discussed in section 5. Correlation between the two sets of measurement results is discussed in section 6.

All statistical tests were made at the 5% significance level.

### 3. Unexposed Ion-Exchange Resin from the Australian FIEFS Network

Inspection of the measurement results on the unexposed ion-exchange resin indicates that

- all results are reported as positive and
- results for the six-monthly period January to June, 1974, are systematically lower than for any other six-monthly period.

For convenience in the analysis, the data were divided, as far as possible into six-monthly groups as follows

<u>Group</u>	<u>Number of Results</u>
March to June, 1973	4
July to December, 1973	6
January to June, 1974	6
July to December, 1974	6
January to June, 1975	6
July to December, 1975	6
January to June, 1976	6
July to October, 1976	4

One-way analysis of variance and Bartlett's test establish that both the group mean and variance for the period January to June, 1974, are different from those for the other periods.

Excluding the measurement results for January to June, 1974, the mean strontium-90 activity is  $0.55 \pm 0.06$  dpm; the uncertainty term is the standard deviation of the mean of the 38 results. A t-test shows that the mean activity is non-zero and positive.

### 4. HASL Quality Control Blanks

Inspection of the measurement results for the HASL quality control blanks indicates that

- All results are reported as positive.
- results for the six-monthly period January to June, 1974, are systematically lower than for any other six-monthly period and
- for nine monthly sets, a separate result for the HASL quality control blank was not available.

In order to compose data sets of sufficient size for statistical analysis, and because of the gaps in the monthly results, the data were divided into six groups as follows

<u>Group</u>	<u>Number of Results</u>
March to December, 1973	7
January to June, 1974	5
July, 1974 to June, 1975	6
July to December, 1975	6
January to June, 1976	6
July to October, 1976	4

One-way analysis of variance and Bartlett's test establish that both the group mean and variance for the period January to June, 1974, are different from those for the other periods.

Excluding the measurement results for January to June, 1974 the mean strontium-90 activity for the 29 measurements, and its uncertainty, are  $0.37 \pm 0.05$  dpm; by t-test, the mean activity is non-zero and positive.

#### 5. Examination of the Frequency Distributions

It is expected that the reported strontium-90 activities for the unexposed ion-exchange resin, and for the HASL quality control blanks, would each be distributed normally, with mean zero, or near-zero and positive; whereas, the frequency distributions of the two sets of measurement results, shown in Figure 1, indicate that

- the results for the unexposed resins are in two distinct groups; namely, a widely dispersed set of activities centered about 0.5-0.6 dpm and a much narrower group below 0.10 dpm,
- the results for the HASL quality control blanks are in two groups; namely, a broad distribution centered about 0.4-0.5 dpm and a narrow group below 0.15 dpm.

Neither set of measurement results, as reported, would appear to be normally distributed in the region of lowest activity. This can be demonstrated by comparing the confidence limits for the frequency of reported results with the frequency that would be expected. Using the normal approximation of the binomial distribution for a series of n repetitions of an event, with probability p of the event occurring with relative frequency h, the confidence limits for p are given by

$$p' = \frac{hn + K^2/2 \pm K (h(1-h)n + K^2/4)^{1/2}}{n + K^2}$$

where  $100(1 - \alpha)\%$  are the confidence limits and  $K$  is defined by

$$\frac{1}{\sqrt{2\pi}} \int_{-K}^K \exp\left(-\frac{t^2}{2}\right) dt = 1 - \alpha$$

For 95% confidence limits,  $K$  is 1.96.

The confidence limits for the frequencies of reported results are given in Table 2 and compared with the expected frequencies assuming that the reported activities are normally distributed. It is evident from Table 2 that far more results are reported in the regions of lowest activity than would be expected; that is, unless it is accepted that the low activity results represent the entire lower tail of the distribution, including negative values.

#### 6. Correlation between Pairs of Results Reported for Unexposed Ion-Exchange Resins and HASL Quality Control Blanks

The pairs of measurement results are plotted as a scatter diagram in Figure 2.

The plot reveals that for the lower activities of HASL quality control blanks, the results on unexposed ion-exchange resin range from near zero up to 1.0 dpm; whereas, at the lower values of activity of unexposed resin, the reported dpm of the HASL blanks range from zero only up to 0.1 dpm. This is inconsistent with the expectation that both sets of results would have a similar spread.

Therefore, before testing the pairs of measurement results for correlation, a pair was deleted if the unexposed resin activity was below 0.10 dpm or the HASL blank was below 0.15 dpm. The correlation coefficient for the remaining 22 pairs of observations is 0.44; this is non-zero and positive at the 5% significance level and would appear to be due to the retention, in all reported results, of the activity of the chemical reagents used in the analyses.

#### 7. Re-estimation of Mean and Standard Deviation

From the foregoing analysis, it is evident that the reported measurement results are anomalous in the lower activity region. Therefore, in order to re-estimate population mean and standard deviation for the strontium-90 activities of the unexposed



resin, the lower tail of the distribution of reported results, including this region, is to be truncated.

For a normal distribution truncated at  $x_1$ , the proportion of observations above  $x_1$  is

$$1 - F\left(\frac{x_1 - \bar{x}}{\sigma}\right)$$

and the mean of observations above, and including,  $x_1$  is given by

$$\bar{x} + \sigma \exp \left\{ -\frac{(x_1 - \bar{x})^2}{2\sigma^2} \right\} / \sqrt{2\pi} \left\{ 1 - F\left(\frac{x_1 - \bar{x}}{\sigma}\right) \right\}$$

$$\text{where } F\left(\frac{x_1 - \bar{x}}{\sigma}\right) = \frac{1}{\sigma \sqrt{2\pi}} \int_{-\infty}^{x_1} \exp \left\{ -\frac{(x - \bar{x})^2}{2\sigma^2} \right\} dx$$

$\bar{x}$  is the mean of the normally distributed sample that should have been observed

$\sigma$  is the standard deviation

$\bar{x}$  and  $\sigma$  are obtained from these expressions by use of tables of the area under the standard normal distribution.

For the unexposed ion-exchange resin, the re-estimated mean of the measurement results is 0.5 dpm and the standard deviation is 0.4 dpm. The values for the results on HASL quality control blanks are 0.3 dpm and 0.4 dpm, respectively. The t-test indicates that both mean values are non-zero and positive.

#### 8. Lower Limit of Detection of Strontium-90 Fallout Deposit

Contamination of unexposed ion-exchange resin with strontium-90 imposes a lower limit on detectability of strontium-90 fallout deposit when the resin is employed in fallout monitoring with the eight Australian FIEFS stations. A lower limit of detection (LLD) for strontium-90 fallout deposit in the monthly fallout samples, measured by HASL can be established by applying the reasoning of section D-08 of the HASL Procedures Manual (Harley, 1972) and assuming that the measurement standard deviation for unexposed resin,  $s$ , also applies to exposed resin at near-zero activities of strontium-90 fallout deposit. Then,  $LLD = 4.65s$  at a confidence level of 95% for detecting

strontium-90 fallout deposit and at a 5% risk of falsely concluding that strontium-90 fallout activity is present, when it is not.

As  $s$  is estimated to be 0.4 dpm for the unexposed ion-exchange resin being employed with the Australian FIEFS station, the LLD for strontium-90 activity in the fallout samples for these stations is 1.9 dpm; this corresponds to a strontium-90 fallout deposit of 0.012 mCi/km<sup>2</sup>.

## 9: Conclusions

From the statistical analysis of the monthly measurement results reported by HASL for the unexposed ion-exchange resin used in the Australian FIEFS network from March 1973 to October 1976, and for HASL quality control blanks analyzed with the unexposed resin, it is concluded that

- (i) The means of both sets of measurement results, for unexposed resin and for quality control blank, are non-zero and positive.
- (ii) The measurement results for the period January to June, 1974, are different to those reported for other periods. The reason for this difference is not known (Harley, 1977).
- (iii) The lower tails of the frequency distributions of both sets of measurement results are anomalous; however, the anomalies are consistent with negative results being reported as positive, near-zero activities.
- (iv) If (iii) is accepted, then re-estimation of the mean and standard deviation of the results on the unexposed resin gives 0.5 dpm and 0.4 dpm, respectively; the corresponding values for the results on the HASL blank are 0.3 dpm and 0.4 dpm, respectively.
- (v) The two sets of reported measurement results are correlated with each other evidently due to the retention, in all reported results, of the activity of the chemical reagents used in the analyses.
- (vi) The lower limit of detection for strontium-90 fallout deposit, at the eight Australian FIEFS stations employing the ion-exchange resin, is 0.012 mCi/km<sup>2</sup>.

## References

- Devlin, B. A., Moroney, J. R., Nunn, R. O. and Stewart, F. M. (1971)  
Defence Standards Laboratories Report AWTSC No. 1, 6-10
- Harley, J. H., ed. (1972) HASL Procedures Manual, USERDA Report HASL-300
- Harley, J. H. (1977) Private Communication

Table 1

Summary of Monthly Measurement Results Reported by HASL for Australian  
Unexposed Ion-Exchange Resins and for HASL Quality Control Blanks<sup>(3)</sup>

Monthly Set	1973		1974		1975		1976	
	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm
January			0.07	0.07	0.06	0.09	0.50	0.47
February			0.02	0.02	0.37		0.64	0.23
March	1.13	0.11	0.02	0.01	0.46	0.27	1.04	0.09
April	0.06		0.02	0.02	0.50	0.06	0.85	0.76
May	0.64 <sup>(1)</sup>	≤ 0.16 <sup>(2)</sup>	0.06	0.02	0.05		0.63	0.37
June	0.80	0.61	0.02		0.32	0.44	0.95	0.56
July	0.75		0.82	0.02	0.52	0.32	0.73	0.66
August	0.39	0.52	0.02		0.11	0.08	0.56	0.88
September	0.67	0.20	0.65	0.37	0.74	0.13	1.18	0.75
October	0.72	1.01	0.27		1.43	0.70	0.70	0.24
November	0.16	0.11	0.37		0.50	0.48		
December	0.10	0.11	0.29		0.10	0.10		

(1) Used only in analysis of variance

(2) Not used in any statistical analysis

(3) All results include the activity of chemical reagents used in the analyses.

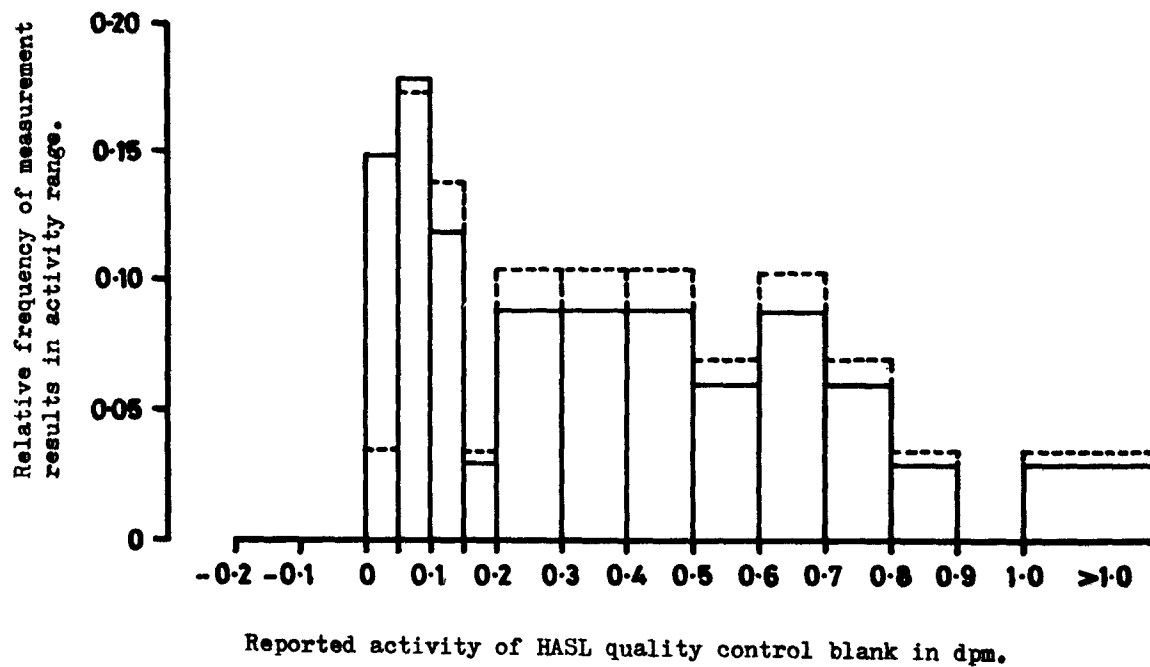
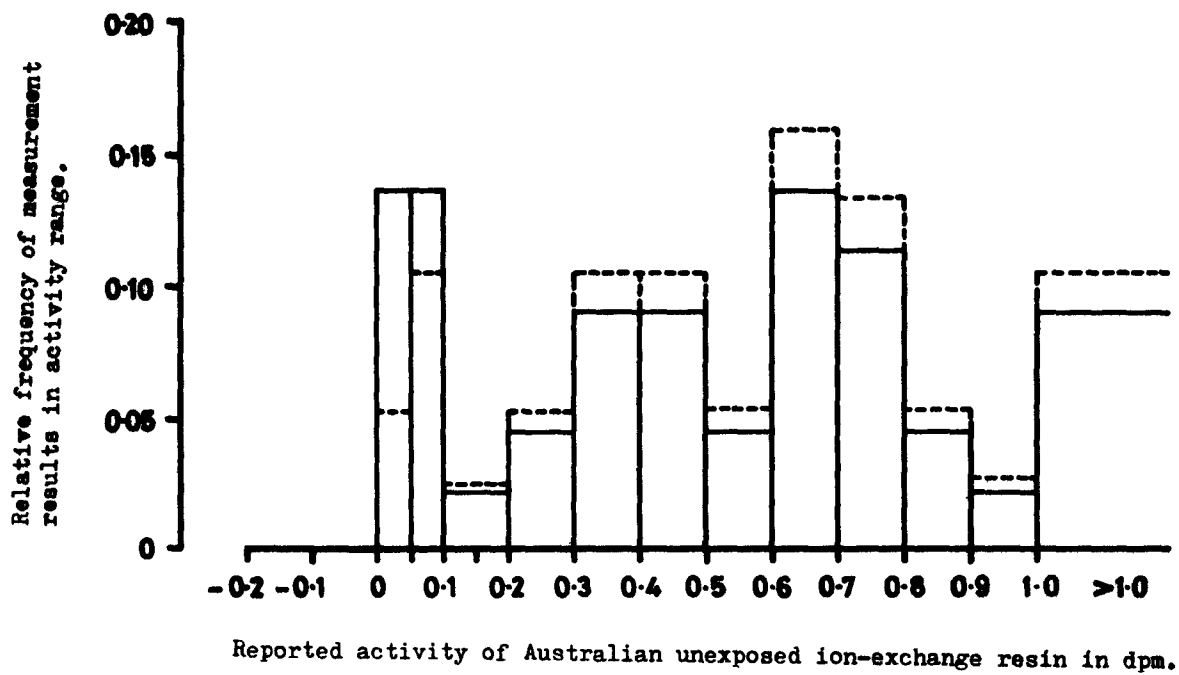


FIGURE 1: Frequency distributions of the monthly measurement results, from March 1973 to October 1976, reported by HASL for Australian unexposed ion-exchange resins and HASL quality control blanks

—— including results for January to June, 1974  
 ---- excluding results for January to June, 1974.

Table 2

Comparison of Frequencies in the Lower Activity Range

	Unexposed ion-exchange resin activity range: 0 to 0.10 dpm		HASL quality control blank ac- tivity range: 0 to 0.15 dpm	
	Jan-June 1974 included	Jan-June 1974 excluded	Jan-June 1974 included	Jan-June 1974 excluded
<u>Reported measurement results</u>				
total number of measurement results	44	38	34	29
number of measurement results in the range	12	6	15	10
frequency of measurement results in the range	0.27	0.16	0.44	0.35
95% confidence limits on frequency	0.16, 0.42	0.07, 0.30	0.29, 0.61	0.20, 0.53
<u>Expectation for normally distributed activities</u>				
mean activity*	0.48 dpm	0.55 dpm	0.32 dpm	0.38 dpm
unbiased standard deviation*	0.36 dpm	0.35 dpm	0.28 dpm	0.28 dpm
expected frequency in the activity range	0.05	0.04	0.15	0.12
expected frequency for the entire lower tail of the distribution below the upper limit of the activity range	0.15	0.10	0.27	0.21

\* Estimated from Table 1; see sections 2 & 3 of the text

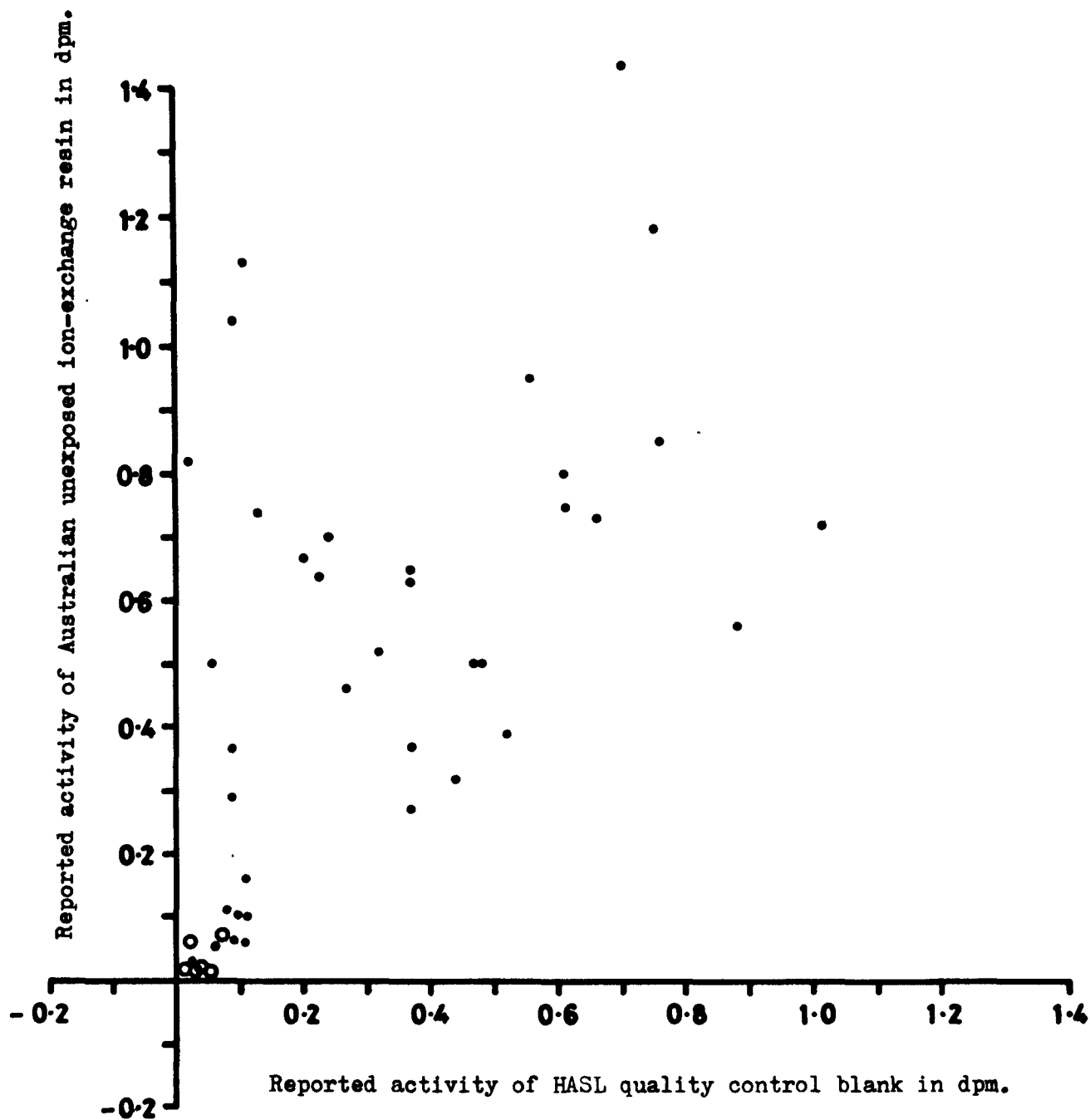


FIGURE 2: Scatter diagram of the pairs of measurement results, from March 1973 to October 1976, reported by HASL for Australian unexposed ion-exchange resin and HASL quality control blanks. The pairs of results for the period January to June, 1974, are distinguished as ○.



CORRECTION OF: N<sub>2</sub>O CONCENTRATIONS IN THE STRATOSPHERE IN 1975

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ABSTRACT

Due to an error in data reduction, stratospheric concentrations of N<sub>2</sub>O which were published earlier are retracted and the correct concentrations are reported. The distribution of the stratospheric concentrations of N<sub>2</sub>O is similar to those of CCl<sub>3</sub>F and SF<sub>6</sub>, although N<sub>2</sub>O is more stable. The inventory of N<sub>2</sub>O in the stratosphere of the Northern Hemisphere in July 1975 is 136 Tg against which future observations can be compared.

In an earlier HASL report<sup>(1)</sup>, we presented the concentrations of N<sub>2</sub>O in the stratosphere in July 1975. Because of a miscalculation in the reduction of the analytical data, a systematic error was introduced in the results. We retract the earlier results and present the corrected data in Table 1 and Figure 1 of this report.

The collection of the samples by the WB-57F aircraft, the N<sub>2</sub>O separation by gas chromatography and the N<sub>2</sub>O assay by electron capture detection have been described in the earlier report<sup>(1)</sup>. Each sample is analyzed at least in duplicate, and the mean and its standard deviation are reported. The electron capture



detector has been calibrated with dilutions of  $N_2O$  standards available from Matheson Gas Products, East Rutherford, NJ. Four of the stratospheric samples (see Table 1) were shared with the Brookhaven National Laboratory (BNL) and Washington State University (WSU). On the average, HASL's results were 10% below WSU's results and 10% above BNL's.

These four interlaboratory samples were analyzed first, and the data reduced separately. The remaining 18 samples were subsequently analyzed, and their data reduced as a group. In this second group of calculations instead of dividing by a corrective factor of 0.86, we multiplied by 0.86 and incurred a constant error factor of 1.35.

#### DISCUSSION

We estimate from Figure 1 and HASL's standard method of planimetering<sup>(2)</sup> that the stratospheric inventory of  $N_2O$  in the Northern Hemisphere in July 1975 is 136 Tg.

Schmeltekopf et al.<sup>(3)</sup> collected air samples from a high altitude balloon launch over Laramie, Wyoming ( $41^\circ N$ ) in June 1975, and their  $N_2O$  concentrations are included in Figure 1. At 18 km, Schmeltekopf et al.'s  $N_2O$  concentration is in good agreement with

our results. The decrease in their  $N_2O$  concentrations above the maximum altitude of the aircraft is similar to the decrease we assumed in order to calculate the stratospheric inventory.

According to our planimetry method, 19% of the  $N_2O$  in the stratosphere of the Northern Hemisphere resides above the 240  $nl/l$  contour. If this estimate of the relative decrease in concentration above the 240  $nl/l$  contour were incorrect by as much as 50%, then our stratospheric inventory would be in error by about 10%. The precision of inventorying a trace material in the stratosphere of the Northern Hemisphere in terms of one standard deviation about the mean has been shown to be  $\pm 9\%$  from 17 individual Airstream missions from which the equilibrium inventories of cosmogenically produced Be-7 were measured (M. Schonberg, unpublished data, 1976). Therefore, excluding the problem of calibration of the electron capture detectors and propagating the above two sources of error, the precision of our estimates of the stratospheric inventory of  $N_2O$  is less than  $\pm 13\%$ . Future stratospheric inventories of  $N_2O$  will be evaluated in the light of this precision to determine what changes may be taking place.

The overall distribution of the  $N_2O$  concentrations in Figure 1 is compatible with a gas emanating from the earth's surface whose conversion to higher oxidation states increases with

altitude in the stratosphere. The  $N_2O$  diffuses upward in the equatorial stratosphere where formation of  $NO$  and  $NO_2$  is more rapid. The lower concentrations of  $N_2O$  then mix and are transported poleward along sloping surfaces parallel to the tropopause. This pattern is very similar to the behavior of  $CCl_3F$  and  $SF_6$  measured in these same samples<sup>(4)</sup>.

The ratios of  $N_2O$  to  $CCl_3F$  and to  $SF_6$  in the July 1975 mission are also given in Table 1 and are illustrated in Figures 2 and 3. These figures indicate that  $N_2O$  is much more stable in the stratosphere than  $CCl_3F$  and only slightly more stable than  $SF_6$ . Although the gradients in the  $N_2O/SF_6$  ratio in Figure 3 are shallow, both figures show an increase in the ratios with altitude and with latitude lending support to the transport pattern described above.

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TABLE 1

N<sub>2</sub>O CONCENTRATIONS AND RATIOS TO CCl<sub>3</sub>F AND SF<sub>6</sub> IN THE STRATOSPHERE  
IN JULY 1975

HASL No.	Collection Date	Alt. (km)	Latitude	Longitude	N <sub>2</sub> O (nl/l±σ)*	$\frac{\text{N}_2\text{O}}{\text{CCl}_3\text{F}}$ (10 <sup>3</sup> )**	$\frac{\text{N}_2\text{O}}{\text{SF}_6}$ (10 <sup>5</sup> )**
G-99†	7/17/75	12.2	71-75N	149-148W	295±1	3.7	9.8
G-100†	7/17/75	15.2	75-71N	148-148W	280±4	4.0	10
G-101	7/15/75	16.8	36-39N	100-100W	300±10	4.2	11
G-103	7/16/75	13.7	59-61N	146-150W	305±1	4.0	11
G-104	7/25/75	15.2	27-24N	92-90W	315±10	3.4	12
G-105	8/1/75	16.8	5-1N	80-80W	310±4	3.6	11
G-106	7/25/75	18.3	15-12N	83-81W	290±8	4.1	11
G-107	7/19/75	19.2	58-61N	139-147W	230±1	7.7	13
G-108†	7/18/75	18.3	75-71N	148-149W	220±1	6.1	12
G-109†	7/18/75	16.8	71-75N	148-149W	240±3	5.3	11
G-110	7/19/75	16.8	61-58N	150-139W	260±4	5.5	12
G-111	7/19/75	19.2	48-52N	134-134N	235±2	6.5	13
G-112	7/20/75	15.2	61-58N	150-142W	290±4	4.1	14
G-113	7/20/75	15.2	52-48N	132-122W	295±2	4.0	13
G-114	7/20/75	13.7	39-36N	102-100W	305±3	3.2	10
G-116	8/6/75	16.8	27-24N	95-95W	305±7	3.9	12
G-117	8/2/75	15.2	12-15N	81-83W	320±4	3.4	11
G-118	8/2/75	18.3	24-27N	90-93W	280±12	4.1	12
G-119	8/6/75	19.2	24-27N	95-95W	285±9	4.8	12
G-120	7/19/75	16.8	52-48N	134-134W	275±3	4.3	13
G-121	7/23/75	19.2	39-36N	94-95W	255±6	4.7	12
G-122	7/23/75	15.2	36-39N	95-94W	305±4	3.4	11

\*nl/l represents nanoliters per liter or 10<sup>-9</sup>/V. σ is the standard deviation of the concentrations.

\*\*Numbers should be read as 3.7×10<sup>3</sup>, 9.8×10<sup>5</sup>, etc.

†Shared with WSU and BNL for intercomparison analyses (see text).

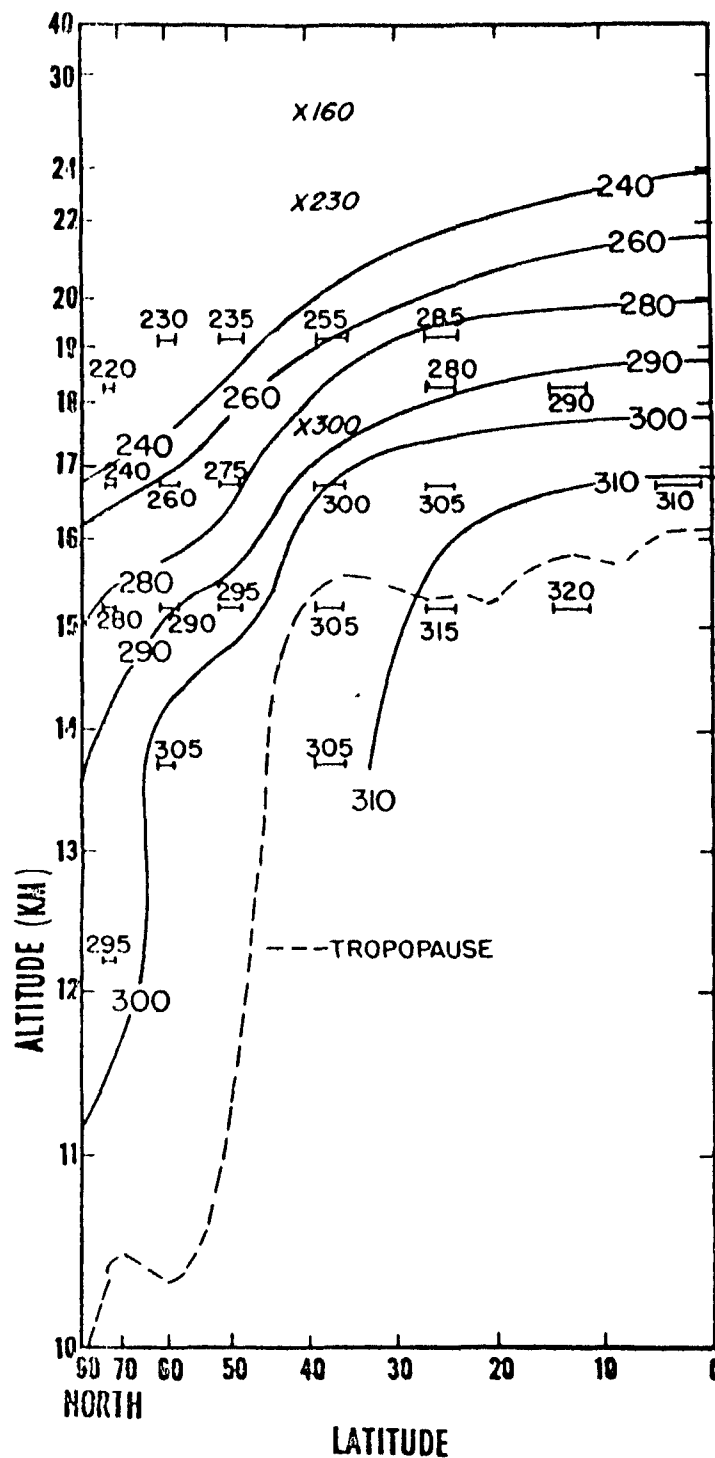


Fig. 1.  $N_2O$  concentrations,  $nl/l$  in July 1975. Short line segments reflect the altitude and latitude intervals of each sample, and the corresponding  $N_2O$  concentrations are given directly above or below this symbol. The x symbols locate Schmeltekopf *et al.*'s<sup>(3)</sup>  $N_2O$  measurements from balloon flights in June 1975. The solid lines are contours of equal concentration drawn from the aircraft data.

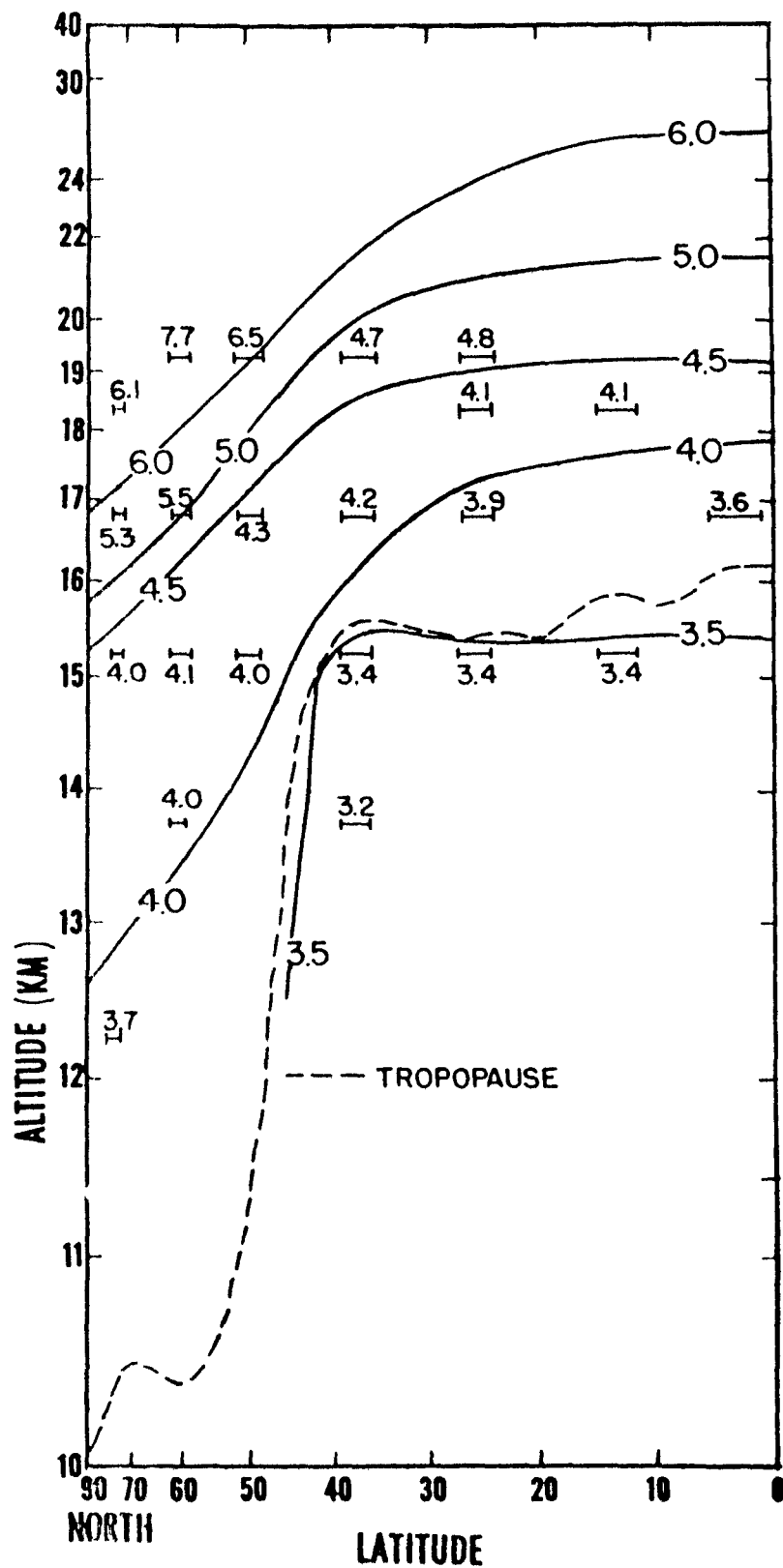


Fig. 2.  $N_2O/CCl_3F$  ratios  $\times 10^{-3}$ . The short line segments are the same as in Fig. 1. The solid lines are contours of equal ratio. Each value has been multiplied by  $10^{-3}$ , so that a value of 3 actually represents a ratio of  $3 \times 10^3$ .

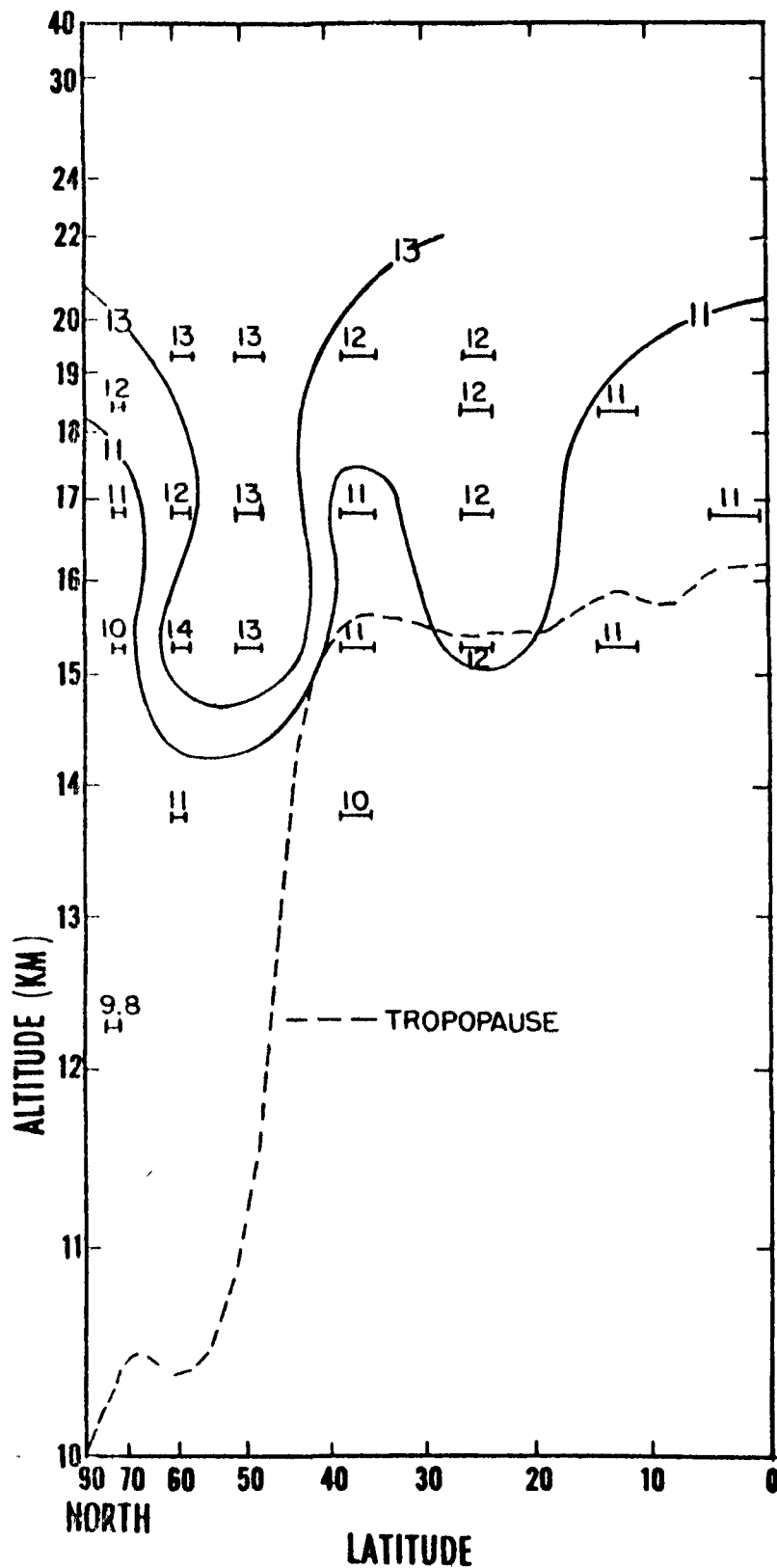


Fig. 3.  $N_2O/SF_6$  ratios  $\times 10^{-5}$ . The short line segments are the same as in Fig. 1. The solid lines are contours of equal ratio. Each value has been multiplied by  $10^{-5}$ , so that a value of 8 actually represents a ratio of  $8 \times 10^5$ .





# AN INTERLABORATORY COMPARISON OF TRACE ELEMENT ANALYSES OF A NEAR SHORE MARINE SEDIMENT

by Merrill Heit, HASL

## ABSTRACT

An interlaboratory comparison was made of the measurements by five laboratories of the concentrations of eighteen trace elements in an air dried, homogenized, near-shore marine sediment sample.

Average sediment concentrations in  $\mu\text{g/g}$  dry weight were measured for As,  $7.4 \pm 13.3$ ; Be,  $1.5 \pm 0.9$ ; Cd,  $1.1 \pm 0.3$ ; Cr,  $38 \pm 15$ ; Cu,  $24 \pm 2$ ; F,  $50 \pm 41$ ; Hg,  $0.15 \pm 0.9$ ; Ni,  $22 \pm 3$ ; Pb,  $37 \pm 10$ ; V,  $42 \pm 33$ ; and Zn,  $76 \pm 8$ . None of the laboratories detected Sn or Te in the sediment sample. The values reported for Ag, Ga, Sb, Se and Tl differed among laboratories by an order of magnitude so that realistic averages for these elements could not be calculated. A comparison of the concentrations of 10 of the elements in the sample is made with data reported on the trace element content of other coastal marine sediments.

## INTRODUCTION

As a preliminary study to our investigations of the input of trace pollutants associated with the combustion of fuels into aquatic systems<sup>(1)</sup>, the Health and Safety Laboratory has performed an interlaboratory comparison of eighteen trace elements present in a near-shore marine sediment. The aims of this study were: a) to determine the levels of detectability of these trace elements in a sediment matrix and b) to evaluate the

present "state of art" of trace element analyses in sediments based upon the performances of five recommended laboratories. Included in the group of laboratories were a university laboratory, a small laboratory associated with an oceanographic institute, two commercial laboratories, and a laboratory operated by a major corporation for a government agency.

The eighteen elements chosen for analysis are all present in fossil fuels and are expected to be released to the environment through combustion or related processes<sup>(2)</sup>. Seventeen of these elements are known to be hazardous, or are potentially hazardous to man or various ecosystems. These elements are Ag, As, Be, Cd, Cr, Cu, F, Ga, Hg, Ni, Pb, Se, Sb, Sn, Te, Tl, and Zn<sup>(3)</sup>. Vanadium was included as an eighteenth element because it may serve as a tracer for pollutants released to the environment by the combustion of fossil fuels<sup>(4)</sup>.

#### MATERIALS

The marine sediment used in the study was collected by John C. Burke of Woods Hole Oceanographic Institution using a steel Van Veen grab from Great Harbor, Woods Hole, MA, during September 1976 and shipped to HASL in a polyethylene container. The sediment was then air dried, passed through a U.S.G.S. No. 20 sieve and mixed by rotating for seven days. One hundred gram

aliquots of this air dried homogenized sediment was sent in polyethylene bottles, which had been washed with 4N  $\text{HNO}_3$ , to each of the five laboratories for analysis.

The specific sample preparations and analysis performed by each of the five laboratories were as follows:

Laboratory A - The sediment samples were digested in HF and  $\text{HNO}_3$ . The liquid fraction, after filtering, was analyzed for all of the elements. All of the elements except F, Hg, As and Sb were analyzed by graphite furnace or flame atomic absorption (A.A.). Arsenic and Sb were measured by DC arc emission spectroscopy, while Hg was analyzed by Cold Vapor A. A. Fluorine was measured by an ion specific electrode.

Laboratory B - The sediment was dissolved by two methods - by digestion in concentrated  $\text{HNO}_3$ , and by fusion with  $\text{Na}_2\text{CO}_3$ . The fusion method was abandoned after it was found to raise the limits of detection for Ag, Be, Cd, Ni, Pb, Sn, Te, Tl, and V. Several techniques were used by laboratory B for measurement of the elements and often more than one technique was used for an element. Induction coupled plasma emission was used for Be, Cd, Cr, Cu, Ni, Pb, V, and Zn. Graphite furnace A. A. was used for Ag, Cd, Pb, Sb, Se, Sn, Te, and Tl. Flame A. A. was used for Ag, Cd, Cu, Ni,

Pb, and Zn. Neutron activation analysis was used for As, Sb, Se, and Te. Nitrous oxide flame emission was used for Cr and V, while cold vapor A. A. was used for Hg. Finally, F was prepared by a separate dissolution, and measured by an ion specific electrode.

Laboratory C - The sediment was digested in concentrated  $\text{HNO}_3$ . Hg was analyzed by cold vapor A. A., while As and Se were measured using a carbon rod atomizer technique. Fluorine was analyzed with an ion specific electrode. The remaining elements Ag, Be, Cd, Cr, Cu, Ga, Ni, Pb, Sb, Sn, Te, Tl, and Zn were analyzed by either graphite furnace or flame A. A.

Laboratory D - For those trace elements analyzed by A. A., Be, Cd, Cu, Ni, Pb, and Tl, the sediment was first dried to constant weight in a dessicator over silica gel. Two different sample destruction procedures were then used, low temperature ashing with an oxygen plasma and  $\text{HNO}_3$  digestion in a Teflon bomb at  $90^\circ\text{C}$ . The samples analyzed for Ag, As, Co, Cr, Ga, Hg, Sb, Se, Sn, Te, and V underwent minimal preparation, and were analyzed by instrumental neutron activation analysis. Fluorine was not measured.

Laboratory E - The sediment was prepared by  $\text{HNO}_3$  digestion. Analysis was by flame A. A. for all of the elements except F which was measured by an ion specific electrode.

## RESULTS AND DISCUSSION

Because information was not available on the amounts of sample required to detect many of these elements as well as the expected concentrations of the elements in this near-shore marine sediment, a single relatively large sample, to be analyzed in triplicate, was sent to each of the laboratories rather than "spiked" samples which may have been prepared at unrealistic levels. Unfortunately, while the precision of the laboratories may be determined by this technique their accuracy can not.

The averages and range of concentrations found by the five laboratories for the eighteen trace elements are given in Table 1. Less-than values were not included in the calculations of the means in the last column.

Because of large variability in some of the data reported by the laboratories, a statistical test, Chauvenets criterion<sup>(6)</sup>, was applied to reject some of the individual laboratory results which may have been caused by calibration problems. These values

are indicated by brackets in Table 1 and include 52.7  $\mu\text{g/g}$  Cd reported by Laboratory E, 58  $\mu\text{g/g}$  Cu reported by Laboratory D, and 120  $\mu\text{g/g}$  Zn reported by Laboratory A. The averages shown in Table 1 for these 3 elements were calculated without these data.

The concentrations of Ag, Ga, Sb, Se, and Tl reported by the five laboratories often differed from each other by an order of magnitude. When averaged, the standard deviations for these elements were greater than their means. Thus, the averages reported in Table 1 for these elements remain questionable. In addition, no values could be given to Sn and Te as they were not detectable in the sediment.

Table 2 shows the precision of the measurements based upon triplicate analysis for all of the laboratories except E, which is based upon seven replicates. For many of these elements (Cd, Cr, Cu, Ni, Pb, V, and Zn), the precision of the values reported by at least three of the five laboratories was good ( $\leq 10\%$  standard deviation). It was fair ( $> 10 - < 20\%$  standard deviation) for As, Be, and Hg and poor ( $> 20\%$  standard deviation) for Se and Sb. Only two of the laboratories had acceptable precision for Ag, F, and Tl, while only one laboratory had acceptable precision for Ga. None of the laboratories could detect Sn or Te in the sediment. Thus, although a number of laboratories showed good

precision for an element, there may still be large variations among the values reported by the laboratories for that element.

It would appear that at present there is less difficulty with the analysis for As, Be, Cd, Cr, Cu, F, Hg, Ni, Pb, V, and Zn at their ambient levels in this marine sediment matrix than for Ag, Ga, Sb, Se, and Tl. Since Sn and Te could not be detected by any of these laboratories, it remains uncertain whether they occur in this sediment at presently unmeasurable concentrations or whether there are unresolved problems with their analysis.

Table 3 is a comparison of the average concentrations ( $\mu\text{g/g}$  dry weight) of 10 of the elements in the Woods Hole sample with marine sediment samples from other locations reported to have low, moderate and high levels of pollution.

From these data it would appear that the levels of Cu, Ni, Pb, and Zn in the Woods Hole sediment sample are similar to those reported for Galveston Bay, Texas, while the As and Cr levels appear similar to values reported for the Pamlico Estuary, North Carolina. Cadmium and V concentrations in the test sample appear similar to levels reported for the Santa Barbara Basin, California. The Hg concentration appears similar to that reported in surface sediment from Long Island Sound. In addition, the level of Be in



the experimental sample was within the range of values reported for the Seward Peninsula of Alaska, an area naturally high in Be. No comparative information was available for F.

All of these near shore marine environments with the exception of the Seward Peninsula, Alaska are areas which receive moderate amounts of anthropogenic pollution. Thus, it is not unreasonable for the levels of the trace elements in the Woods Hole sediment to be comparable to those found in sediment from other near shore locations of the United States in proximity to population centers or industrialized areas.

#### Acknowledgement

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TABLE 1. AVERAGE CONCENTRATIONS OF TRACE ELEMENTS IN THE SAMPLE REPORTED  
BY THE 5 LABORATORIES ( $\mu\text{g/g}$  dry weight)

Element	Laboratories <sup>a</sup>					Average
	A	B	C	D	E	
Ag	21.0 $\pm$ 3.4	0.7 $\pm$ 3.4	<1	<0.2	<100	11 $\pm$ 14
As	5.5 $\pm$ 0.1	11.7 $\pm$ 1.5	5.0 $\pm$ 1.0	<1.2	<200	7.4 $\pm$ 3.3
Be	1.1 $\pm$ 0.1	0.7 $\pm$ 0.1	<4	1.7 $\pm$ 0.6	2.8 $\pm$ 0.4	1.5 $\pm$ 0.9
Cd	0.8 $\pm$ 0.2	1.1 $\pm$ 0.2	1.1 $\pm$ 0.1	1.5 $\pm$ 0.3	[52.7 $\pm$ 3.8] <sup>B</sup>	1.1 $\pm$ 0.3
Cr	54.0 $\pm$ 1.0	32.0 $\pm$ 2.0	40.7 $\pm$ 1.2	46.0 $\pm$ 6.2	15.9 $\pm$ 2.6	38 $\pm$ 15
Cu	24.7 $\pm$ 3.2	25.3 $\pm$ 1.5	23.7 $\pm$ 0.6	[58.0 $\pm$ 3.0] <sup>B</sup>	21.2 $\pm$ 2.6	24 $\pm$ 2
F	>10	96.7 $\pm$ 5.9	19.7 $\pm$ 5.9	-	33.0 $\pm$ 3.0	50 $\pm$ 41
Ga	0.5 $\pm$ 0.2	<1	20.0	<1	<500	10 $\pm$ 14
Hg	0.13 $\pm$ 0.01	0.10 $\pm$ 0.01	0.19 $\pm$ 0.01	<0.02	0.19 $\pm$ 0.02	0.15 $\pm$ 0.05
Ni	20.0 $\pm$ 7.0	13.0 $\pm$ 0	14.3 $\pm$ 0.6	26.0 $\pm$ 6.0	38.4 $\pm$ 3.0	22 $\pm$ 3
Pb	34.7 $\pm$ 4.7	38.7 $\pm$ 3.2	35.3 $\pm$ 1.5	23.0 $\pm$ 5.0	51.2 $\pm$ 3.5	37 $\pm$ 10
Sb	0.7 $\pm$ 0.7	0.5	<20	8.7 $\pm$ 1.0	48.4 $\pm$ 5.6	15 $\pm$ 23
Se	79.0 $\pm$ 18.2	<0.5	2.7 $\pm$ 0.6	1.3 $\pm$ 0.2	237.0 $\pm$ 56	80 $\pm$ 56
Sn	<1	<30	<60	<20	<0.5	-
Te	<0.2	<0.5	<20	<130	<500	-
Tl	1.0 $\pm$ 1.0	<1	10.0 $\pm$ 0	<0.5	<0.1	6 $\pm$ 6
V	94.7 $\pm$ 3.5	5.4 $\pm$ 0.2	40 $\pm$ 0	42 $\pm$ 5.0	29.5 $\pm$ 6.4	42 $\pm$ 33
Zn	[120.0 $\pm$ 17.3] <sup>B</sup>	87.0 $\pm$ 1.0	74.3 $\pm$ 2.5	75.0 $\pm$ 10.0	66.8 $\pm$ 6.6	76 $\pm$ 8

<sup>a</sup> All laboratories remain anonymous.

<sup>B</sup> Data in brackets is rejected by Chauvenets Criteria.

TABLE 2. ANALYTICAL PRECISION ( $\pm\%$ )

Laboratory	E l e m e n t s									
	Ag	As	Be	Cd	Cr	Cu	F	Ga	Hg	
A	16	2	9	26	2	13	-	28	8	
B	<1	13	9	14	6	6	3	N.D.	11	
C	N.D.	20	N.D.	6	3	2	30	<1	5	
D	N.D.	N.D.	35	7	13	5	-	N.D.	N.D.	
E*	N.D.	N.D.	14	7	16	12	9	N.D.	11	
	Ni	Pb	Sb	Se	Sn	Te	Tl	V	Zn	
A	35	14	23	23	N.D.	N.D.	9	4	14	
B	<1	8	<1	N.D.	N.D.	N.D.	N.D.	4	1	
C	4	4	N.D.	22	N.D.	N.D.	<1	<1	3	
D	23	22	35	15	N.D.	N.D.	N.D.	12	13	
E*	8	7	12	24	N.D.	N.D.	N.D.	22	10	

N.D. = Non detectable

- = A triplicate analysis was not made

\* = The precision of laboratory E is based upon 7 replicates per element.

TABLE 3. COMPARISON OF TRACE ELEMENTS IN THE SAMPLE  
AND MARINE SEDIMENTS FROM OTHER AREAS

Element	Average Sample Concentration ( $\mu\text{g/g}$ dry wt.)	Average of Other Marine Sediments ( $\mu\text{g/g}$ dry wt.)	Relative Level of Pollution	Location of Other Sediments	Reference
As	7.4 $\pm$ 3.3	1.85	1	Trinity Bay, New Foundland	9
		3.5	1	Hawke Channel, Labrador	10
		6.86 <sup>a</sup>	2	Pamlico Estuary, NC	15
Be	1.5 $\pm$ 0.9	0.023	1	Deep Pacific Ooze	6
		0.037-2.7	1	Seward Peninsula, Alaska	6
Cd	1.1 $\pm$ 0.3	0.7	1	Haifa Bay, Israel	12
		1.4	2	Georgia, SC, Estuary	16
		1.2 <sup>B</sup>	2	Santa Barbara, Calif.	11
Cr	37.7 $\pm$ 14.6	6.8	1	Saldanha Bay, S. Africa	7
		3.7	1	Urr Estuary, England	8
		31.3	2	Pamlico Estuary, NC	15
Cu	23.7 $\pm$ 1.8	4.0	1	San Antonio Bay, Texas	13
		19.0	2	Galveston Bay, Texas	13
		46.0	3	Houston Ship Channel, Texas	13
Hg	0.15 $\pm$ 0.05	0.07	1	Urr Estuary, England	8
		0.24 <sup>C</sup>	2	Long Island Sound	14
		2.23	3	Mersey Estuary, England	17
Ni	22.3 $\pm$ 10.4	9.9	1	San Antonio Bay Texas	13
		22.1	2	Galveston Bay, Texas	13
		34	3	Houston Ship Channel, Texas	13
Pb	36.6 $\pm$ 10.1	9.5	1	San Antonio Bay, Texas	13
		24	2	Galveston Bay, Texas	13
		113	3	Houston Ship Channel, Texas	13
V	42.3 $\pm$ 32.7	16 <sup>B</sup>	1	Santa Pedro, California	11
		32 <sup>B</sup>	2	Santa Monica, California	11
		42 <sup>B</sup>	2	Santa Barbara, California	11
Zn	75.8 $\pm$ 8.4	8.7	1	San Antonio Bay, Texas	13
		51	2	Galveston Bay, Texas	13
		240	3	Houston Ship Channel, Texas	13

1 = relatively little pollution  
2 = relatively moderate level of pollution  
3 = relatively high level of pollution

<sup>a</sup> Calculation from data.

<sup>B</sup> Reducible fraction only.

<sup>C</sup> Top 2 cm of core.



An Estimate of Maximum Credible Atmospheric Radioactivity  
Concentrations from Nuclear Tests

by

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ABSTRACT

Based on aircraft radioactivity samples following each atmospheric Chinese nuclear test from 1964 to 1976, an estimate is made of maximum credible air concentrations from nuclear detonations. The calculated maximum credible air concentration is a function of the estimated total yield and fission yield, and estimates of the initial vertical radioactivity distribution and the rate of horizontal spread of the debris with time. A simple expression for horizontal diffusion appears to give good results for many days following a nuclear test in temperate latitudes.



## 1. Introduction

The injection of radioactive debris into the atmosphere by a nuclear explosion could result in exposure of aircraft to relatively high radioactivity concentrations. Air concentrations depend on the amount of radioactivity produced, its initial vertical distribution, and such meteorological factors as wind speed and the rate of dispersion of the debris as it travels. This paper presents a simple technique for estimating the maximum credible concentrations as a function of the energy yield of a nuclear device detonated well above the ground and time after detonation, based on past observations. For nuclear devices detonated at or near the ground, the radioactivity in the cloud would be depleted due to local fallout.

## 2. Air Concentration Data

Radioactivity concentration measurements in the atmosphere following the Chinese nuclear tests from 1964 to 1976 are given in Table 1. The approximate total yield (equivalent TNT energy) of each detonation is given with a reference to the data source. For detonations having a total yield of 1 megaton or less it was assumed that the fission yield was equal to the total yield. For the five high yield tests, each over 2 megatons, conducted prior to November 1976, the fission yield was estimated by Telegadas (1974, 1976). For the latest test, November 17, 1976, the fission yield is assumed to be one half the total yield since no data are yet available to provide a better estimate.

The altitude of sampling is shown for the highest concentrations found following each test. The gross beta radioactivity concentrations

at sampling times, determined by counting radioactivity collected on filter papers, are in units of picocuries per standard cubic meter of air (pCi/SCM; a SCM is computed at 76 cm Hg and 15°C or 1 SCM = 1.225 x 10<sup>3</sup>g air). The concentration in the last column has been adjusted to its expected value one day (D+1) after the detonation. This adjustment takes into account the decay of mixed fission products using the Way-Wigner law:  $R_t = R_1 t^{-1.2}$  where R is the radioactivity in a sample and t is time; the R<sub>1</sub> refers to the radioactivity at unit time (e.g., 1 hour or 1 day). A dilution factor is also included in the adjustment and will be discussed later.

### 3. A Simple Method for Calculating Radioactivity Concentrations

The determination of radioactivity concentrations produced by the detonation of an atmospheric nuclear explosion depends on the total radioactivity produced, its initial vertical distribution and the rate of dispersion of the debris as it travels.

A simple expression containing these parameters can be given, such that

$$\bar{X}_{h,t} = \frac{\gamma \cdot \left( \frac{1.45 \times 10^{23} \text{ fissions}}{\text{kiloton}} \right) \cdot \beta_t \cdot f_h \cdot \left( \frac{0.45 \text{ pCi}}{\text{dpm}} \right)}{\pi (2\sigma_H)^2} \quad (1)$$

where  $\bar{X}_{h,t}$  is the average gross  $\beta$  concentration, (pCi/m<sup>3</sup>), at altitude (h) and time (t) after the detonation.

$\gamma$  is the fission yield of the device, (kilotons, KT).

$\beta_t$  is the gross beta radioactivity per fission at time (t), (dpm/fission).

$f_h$  is the fraction of radioactivity per meter at altitude (h).

$\sigma_H$  is the horizontal standard deviation of the cloud radioactivity distribution (m).

$2\sigma_H$  is the assumed horizontal radius (r) of the debris cloud at time (t); therefore,  $\pi(2\sigma_H)^2$  is the area of the debris cloud over which the concentration is calculated.

Conversion factors used in Equation (1) are:

- a) 1 KT fission produces  $1.45 \times 10^{23}$  fissions, and
- b) 0.45 picocuries (pCi) are equivalent to 1 disintegration per minute (dpm).

#### Fission yield.

Estimates of fission yields for the five high yield Chinese tests prior to November 1976 are given in Table 1, while the November 17, 1976 high yield test is assumed to have a fission yield of 2000 KT. For all the other lower yield detonations the fission yield is assumed to be equivalent to the total yield.

#### Gross beta radioactivity.

The total quantity of gross beta remaining after decay of the initial amount from  $10^6$  fissions is shown in Figure 1. Harley et al. (1965) give the total beta radioactivity remaining at 10 days and later while Healy and Baker (1968) provide the decay starting several hours after the nuclear explosion. We have used the Healy-Baker curve for the period from 1 to 10 days and the Harley et al. curve after 10 days.

#### Initial vertical radioactivity distribution.

Telegadas (1974, 1976) estimated the initial vertical distributions of radioactivity for the first five high yield Chinese tests as given in Appendix A, Table A-1. Although these vertical distributions were based on radioactivity measurements made several weeks or months after

each event, they represent the best information available to assess the original distribution.

Stratospheric horizontal dispersion.

An expression for the horizontal standard deviation,  $\sigma_H$ , has been obtained by Heffter et al. (1975) from a compilation of many lateral dispersion measurements in the troposphere (Heffter, 1965). Heffter et al. (1975) give

$$\sigma_H \text{ (meters)} = 0.5t \text{ (sec)} \quad (2)$$

as a reasonable approximation for the growth of a cloud during several days travel time in the troposphere.

Very few measurements have been made of lateral dispersion in the lower stratosphere. Virtually no measurements after several days travel time have been reported. Equation (1) was used to calculate  $\sigma_H$  from the stratospheric concentration measurements for the six high yield tests given in Table 1. Calculated values of  $\sigma_H$  ranged from 0.25t to 1.1t for travel times from 1.5 to 17 days. This agrees well with the range of  $\sigma_H$  given by Bauer (1974) for these travel times in the lower stratosphere.

Since we are concerned with estimating the maximum credible concentrations, we will minimize the cloud growth, using

$$\sigma_H \text{ (meters)} = 0.25t \text{ (sec)} \quad (3)$$

for travel times of several days in the lower stratosphere.

Since vertical dispersion is very slow in the stratosphere, it will be neglected, and the vertical distribution given in Appendix A will be used for the first few weeks after detonation. The effects of wind shear are also neglected.

#### 4. Calculated Versus Observed Concentrations for the High Yield Chinese Tests.

Equation (1) was used to calculate an expected air concentration at the altitude and time of the observed maximum concentration for each of the Chinese high yield nuclear tests. To illustrate the procedure, on the June 17, 1967 test the observed gross beta concentration 15 days after the detonation at 20.1 km was  $0.65 \times 10^6$  pCi/SCM (Table 1).

Procedure:

- a) Fission yield (Y) = 1600 KT (Table 1).
- b) From Figure 1, at 15 days  $\beta_t$  is 7.7 dpm/ $10^6$  fissions or  $7.7 \times 10^{-6}$  dpm/fission. Multiplying this number by the fission yield and assuming  $1.45 \times 10^{23}$  fissions per KT yields a value of  $18 \times 10^{20}$  dpm or  $8 \times 10^{20}$  pCi of gross beta remaining in the atmosphere after 15 days.
- c) Layer 19.1 to 20.6 km (62.5-67.5 kft) contains 28% of the total activity (see Appendix A, Table A-1), hence  $f_h$  is 
$$\frac{0.28}{1.5 \text{ Km}} \approx 0.19/\text{Km} \approx 1.9 \times 10^{-4}/\text{m}.$$
- d) Assuming  $\sigma_H$  (meters) = 0.25t (sec), at 15 days  $\sigma_H = 3.24 \times 10^5 \text{ m}.$

The average concentration at an altitude of 20.1 km would therefore be:

$$\begin{aligned}\bar{X} &= (8 \times 10^{20} \text{ pCi}) (1.9 \times 10^{-4} \text{ m}^{-1}) \frac{1}{\pi (2 \times 3.24 \times 10^5)^2 \text{ m}^2} \\ &= 1.15 \times 10^5 \text{ pCi/m}^3 \text{ (ambient)}\end{aligned}$$

Finally, to convert ambient cubic meters of air at 20.1 km to standard cubic meters of air we multiply the volume by  $\rho/\rho_0 = 7.1 \times 10^{-2}$  at 20.1 km.

This results in a calculated concentration of  $1.6 \times 10^6$  pCi/SCM compared to the observed concentration of  $0.65 \times 10^6$  pCi/SCM.

In this instance the observed concentration is overestimated by about a factor of 2. This is encouraging considering all the uncertainties in the calculation. We must also bear in mind that the peak concentration at this altitude may not have been sampled.

The comparisons between observed and calculated concentrations for the six high yield tests are given in Table 2 and shown graphically in Figure 2. For the November 17, 1976 event no fission yield information was available and a fission yield of 2000 KT was assumed. Since no estimate has been made yet of the initial vertical activity distribution, as has been done for the previous five high yield events, it was assumed that 38% of the total activity was contained in a 1500 meter layer (5000 feet thick) centered at 20.4 km, altitude of the observed concentration.

This method of determining air concentrations overcalculates in all six cases. For five cases the overcalculation of the observed concentration was a factor of 5 or less. The October 1970 case was overcalculated by about a factor of 20. We note that this sample was collected well below the altitude of maximum concentration and the vertical distribution was estimated from very limited data.

Considering all the uncertainties, this simple technique appears to give acceptable results. The tendency to overestimate the measured concentration is not considered to be a serious drawback since the technique will be used to estimate a maximum credible concentration.

##### 5. Maximum Credible Air Concentrations.

The prediction of maximum credible air concentrations is based on the above procedure except that a mean vertical radioactivity distribution

is used and the altitude of the radioactivity maximum is estimated from the total yield of the explosion.

The assumption is made for air bursts detonated at a relatively low altitude, below about 3 km, with a total yield of 500 KT or less, the total radioactivity produced will remain initially in the troposphere. For larger yields (>500 KT) all the radioactivity is assumed to be injected into the stratosphere.

The cloud thickness as a function of yield is shown in Figure 3. Diagrams such as this have appeared previously (e.g., Quenneville and Nagler, 1959; Ferber, 1965; and Peterson, 1970). Figure 3 represents a composite of previous cloud height determinations and is intended to represent the vertical extent of nuclear clouds in temperate latitudes. Different atmospheric conditions produce variability in the height of nuclear clouds of the same yield. The variability about the cloud top curve in Figure 3 is about  $\pm 2$  km as estimated by Quenneville and Nagler (1959) and Ferber (1965).

Also shown in the inset of Figure 3 is an assumed initial vertical distribution of radioactivity in the nuclear cloud. This parameter is less well documented by actual data than the visual cloud characteristics. Ferber (1965) was able to determine from measurements that, for an airburst, the stem portion of the nuclear cloud contained less than 1% of the total radioactivity. Early time measurements in the mushroom cap of a nuclear cloud are few, particularly for air bursts. Ferber (1965) and Peterson (1970) have estimated the initial vertical activity distribution based on very limited data.

Telegadas (1974) presented an initial mean vertical activity distribution for a 3 MT nuclear detonation in temperate latitudes. This estimate was based on the calculated vertical activity distributions for the first four high yield Chinese tests conducted between 1967 and 1970 (see Appendix A). The calculated initial vertical distribution estimated by Telegadas (1976) for the June 27, 1973 high yield test had a similar distribution as the composite for the first four events. For lack of better data, it is assumed that the initial mean vertical distribution as given by Telegadas (1974) is valid for all air bursts. This distribution, shown in the inset diagram of Figure 3, is divided into seven layers of equal thickness and is similar to the distribution proposed by Ferber (1965) and Peterson (1970). The altitude of the maximum concentration is assumed to be mid-way between the cloud top and base, Figure 3. This altitude is assumed to be the midpoint of a layer containing 38% of the total activity (inset of Figure 3).

We assume that the horizontal dispersion expression given by Heffter et al. (1975) (Equation (2)) is valid for tropospheric injections (500 KT or less) and that Equation (3) is valid for stratospheric injections (>500 KT). Unless there is information to the contrary, detonations  $\leq 500$  KT are assumed to be 100% fission, all those >500 KT are assumed to be 50% fission. Vertical dispersion is neglected.

With the above assumptions, a maximum credible concentration at one day after detonation has been estimated from Equation (1) for nuclear yields from 1 KT to 10 MT. This is shown by the curve in Figure 4.

Since the horizontal standard deviation,  $\sigma_H$ , is assumed to increase linearly with time, the volume of a nuclear cloud layer (assuming no



vertical dispersion) increases as  $t^2$ . The gross beta radioactivity decreases according to  $t^{-1.2}$  decay. By combining these two factors, dispersion and decay, one can arrive at a reduction factor, shown graphically in Figure 5, which gives the decrease in air concentration with time after 1 day. As an example, if one had an observed concentration on D+1 and wished to estimate the concentration at D+4 one need only to multiply the observed concentration by the ratio of the reduction factor on day 4 ( $10^{-2}$ ) to the reduction factor on day 1 (1). The concentration would, therefore, be a factor of 100 lower after four days than after one day following a nuclear explosion.

The reduction factors in Figure 5 were applied to the observed concentration data in Table 1 to adjust all the data to D+1. The highest adjusted concentration for each detonation in Table 1 was plotted in Figure 4. All but one of the data points fall below the curve which represents the maximum credible concentration. The June 27, 1973 nuclear test, which had a reported total yield between 2000-3000 KT (see Table 1), lies above the maximum credible curve. Note that this concentration measurement ( $1 \times 10^6$  pCi/SCM) at 20 km was made 17 days after June 27, 1973 and then adjusted to a concentration of  $11,000 \times 10^6$  pCi/SCM at D+1. The adjusted concentration is about a factor of 2 higher than the estimated maximum credible concentration.

The highest adjusted concentrations for the nuclear tests greater than 2 MT were for the 3 events sampled at the highest altitude, about 20 km. These concentrations were within a factor of 2 of the predicted maximum credible concentration. For the other 3 high yield events, which were sampled at lower altitudes, the adjusted concentrations were a factor

of about 20 to 400 below the predicted maximum credible concentration.

There are four events with an estimated yield of 20 KT that were sampled at altitudes ranging from 6.4 to 10.7 km (see Table 1) from one to two and a half days after detonation. The concentrations were between  $0.8 \times 10^6$  and  $6.9 \times 10^6$  pCi/SCM at D+1. This indicates the variability in the maximum concentration encountered for any given yield.

The point denoted by an asterisk in Figure 4 represents radioactive debris encountered over the mid-western U.S. 4.3 days after a U.S. nuclear test over Christmas Island (List et al., 1964). The detonation occurred on May 4, 1962 and was reported to have a total yield in the intermediate range (200 KT - 1 MT). The highest concentration was measured at 15.2 km on May 8, 1962, and had a value of about  $10^6$  pCi/SCM at collection time. This observed concentration was adjusted to D+1 and plotted in Figure 4 as another valid data point.

The maximum credible concentration as a function of yield as depicted in Figure 4 seems to be a reasonable estimate. It is felt that the stratospheric horizontal dispersion ( $\sigma_H$ ), although an oversimplification of the real atmospheric dispersion, is a reasonable approximation of cloud radius growth up to about 5 days, possibly up to 10 days. One can use the reduction factor in Figure 5 to estimate a maximum credible concentration curve for any time up to 10 days.

#### 6. Visual Observation of a Nuclear Cloud.

The atmospheric nuclear test of November 17, 1976 conducted at Lop Nor, China ( $40^\circ\text{N}$ ,  $90^\circ\text{E}$ ) at 0600 GMT was reported to have a total yield of about 4 megatons. Aircraft sampling 4.4 days after this event at 20.4 km obtained

a maximum gross beta air concentration of  $65 \times 10^6$  pCi/SCM. This concentration was one of the highest measured for any Chinese test.

Approximately one-half hour before the aircraft sample was taken the pilot reported a visual description of a rust colored cloud at 20.4 km. The cloud was described as elliptical in shape with a major axis in a west-east direction approximately 330 km long; the minor axis about 110 km long. This cloud was estimated to have a vertical extent of 230 meters. It is believed that the rust color was due to the oxides of nitrogen formed by the nuclear explosion. Since the aircraft obtained the high gross beta concentration while sampling in and around the visible cloud, it is assumed that the dimensions of the visible cloud are also representative of the radioactive cloud. If this assumption is valid, a portion of the original nuclear cloud hung together for four or five days after detonation, undergoing rather slow horizontal dispersion.

If we assume that the rust colored cloud which covered an area of  $2.9 \times 10^{10} \text{ m}^2$ , was circular rather than elliptical in shape, then its radius would have been about  $9.5 \times 10^4$  meters. If we further assume that the radius (r) is equal to  $2\sigma_H$  then at 4.4 days the time of visual sighting,

$$\sigma_H = 4.8 \times 10^4 \text{ meters} = 0.12t \text{ (sec)} \quad (4)$$

which indicates a dispersion rate about half of that assumed for the lower limit in Equation (3).

It can be shown that if we used Equation (4) in the determination of an air concentration, we would have calculated a concentration a factor of 4 greater than when we used Equation (3). It was shown in Table 2, that for the six high yield Chinese events which injected the

bulk of its radioactive debris into the lower stratosphere, the calculated concentrations, using Equation (3) overcalculated the observed concentrations from a factor of 1.1 to 19.6. Using Equation (4), which is based on only one measurement, we would overcalculate the observed concentrations from a factor of 4.4 to 78. We, therefore, used Equation (3) as a reasonable estimate of horizontal dispersion in the lower stratosphere.

## 7. Conclusion.

Many of the parameters used to determine the maximum credible concentration as a function of yield and time after detonation, such as initial vertical extent of the cloud, initial radioactivity distribution with altitude, and assumed fission-fusion ratio are based on limited data. The use of the simple linear growth of the cloud radius with time (Eqs. (2) and (3)) oversimplifies real atmospheric dispersion. It must be stressed that the altitude of the stabilized nuclear cloud will vary with meteorological conditions. Figure 3 should, therefore, be used with caution.

In spite of the uncertainties involved, the calculated curve of maximum credible concentration is consistent with the limited data on maximum concentrations measured in nuclear clouds from 1 to 17 days after detonation.

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Table I.—Concentrations of Radioactive Debris From Chinese Atmospheric Nuclear Tests, 1964—1976.

Year	Date	Approximate Yield (Kilotons)		Sample Collection Altitude (Km)	Concentration Gross Beta (10 <sup>6</sup> pCi / SCM )	
		Total	Fission		At Collect Time	At D+1
1964	Oct. 16	20 <sup>(1)</sup>	20	9.0	1.8	1.8
1965	May 14	40 <sup>(1)</sup>	40	12.2	1.0	2.5
1966	May 9	300 <sup>(1)</sup>	300	11.6	1.4	6.1
	Oct. 28	20 <sup>(1)</sup>	20	8.8	0.48	6.9
	Dec. 28	300 <sup>(1)</sup>	300	9.8	4.3	10.7
1967	Jun. 17	3000 <sup>(1)</sup>	1600 <sup>(2)</sup>	12.2 20.1	0.02 0.65	0.05 4300
	Dec. 24	20 <sup>(1)</sup>	20	10.7	0.55	0.8
1968	Dec. 28	3000 <sup>(1)</sup>	1900 <sup>(2)</sup>	15.2	130	370
				9.5	1.7	1.4
1969	Sep. 29	3000 <sup>(1)</sup>	1800 <sup>(2)</sup>	16.2	35	125
				9.1	0.04	0.2
1970	Oct. 14	3000 <sup>(2)</sup>	1500 <sup>(2)</sup>	11.6	1.5	4.3
				14.9	4.7	16.8
1971	Nov. 18	20 <sup>(3)</sup>	20	6.4	0.06	1.2
1972	Jan. 7	< 20 <sup>(4)</sup>	< 20	7.6	0.08	2.3
	Mar. 18	20-200 <sup>(5)</sup>	20-200	13.7	2.9	8.3
1973	Jun. 27	2000- 3000 <sup>(6)</sup>	1400 <sup>(7)</sup>	20.0	1.0	11000
1974	Jun. 17	200-1000 <sup>(6)</sup>	200-1000	11.5	0.07	0.2
				16.5	0.10	10.6
1976	Jan. 23	< 20 <sup>(6)</sup>	< 20	2.7	5 x 10 <sup>-6</sup>	0.003
	Sep. 26	20-200 <sup>(6)</sup>	20-200	10.4	1.0	2.9
	Nov. 17	4000 <sup>(6)</sup>	2000 <sup>(8)</sup>	10.7 20.4	1.6 65	4.0 7600

(1) Hardy, 1970 ; (2) Telegadas, 1974; (3) USAEC News Release, O- 213;  
(4) USAEC News Release, P-6; (5) USAEC News Release, P-77; (6)  
Information received from Office of Public Affairs, USERDA; (7) Tele-  
gadas, 1976; (8) Assumed for computation purposes.

Table 2. Comparison of the Observed and Calculated Gross Beta Concentrations at Day of Collection.

Date	Observed ( $10^6$ pCi/SCM)	Calculated ( $10^6$ pCi/SCM)	Calc./Observed
June 17, 1967	0.65	1.6	2.5
Dec. 28, 1968	130	650	5.0
Sept. 29, 1969	35	84	2.4
Oct. 14, 1970	4.7	92	19.6
June 27, 1973	1.0	1.1	1.1
Nov. 17, 1976	65	140	2.2



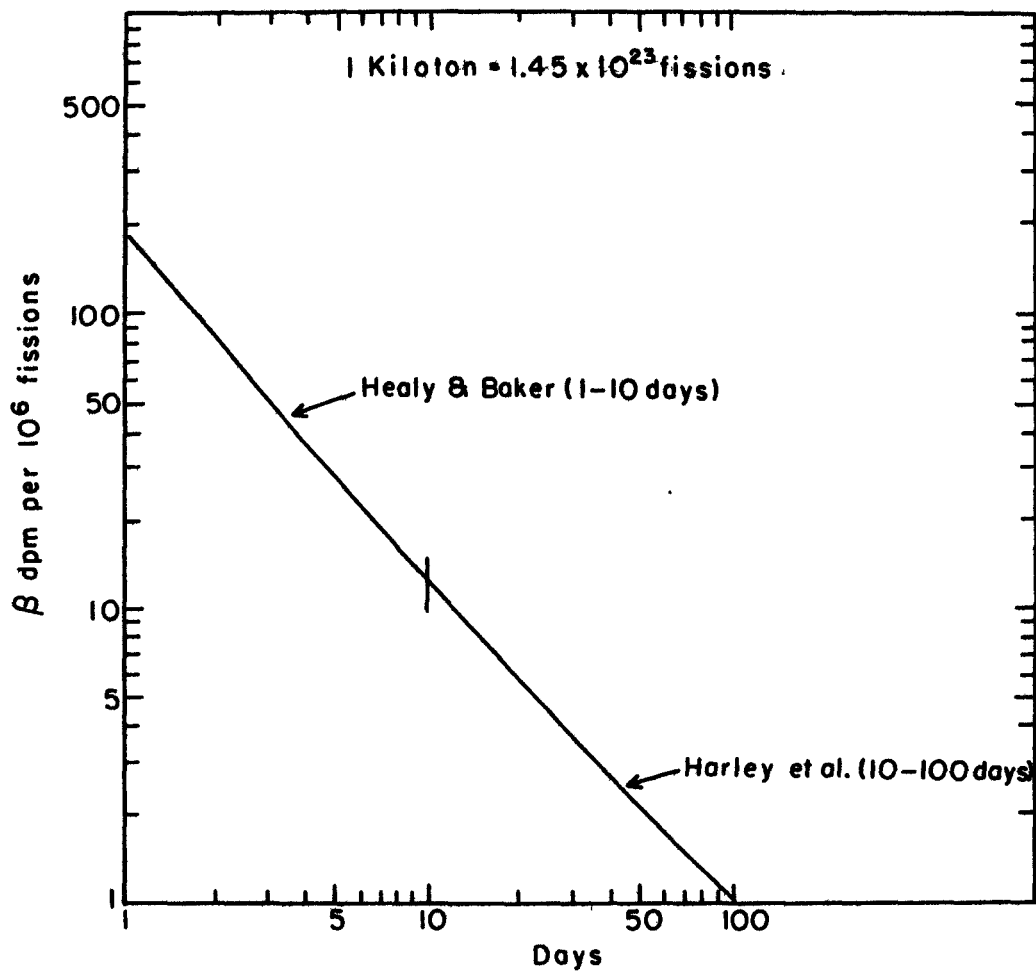


Figure 1. Total beta disintegrations per minute per  $10^6$  fissions remaining after a nuclear detonation.

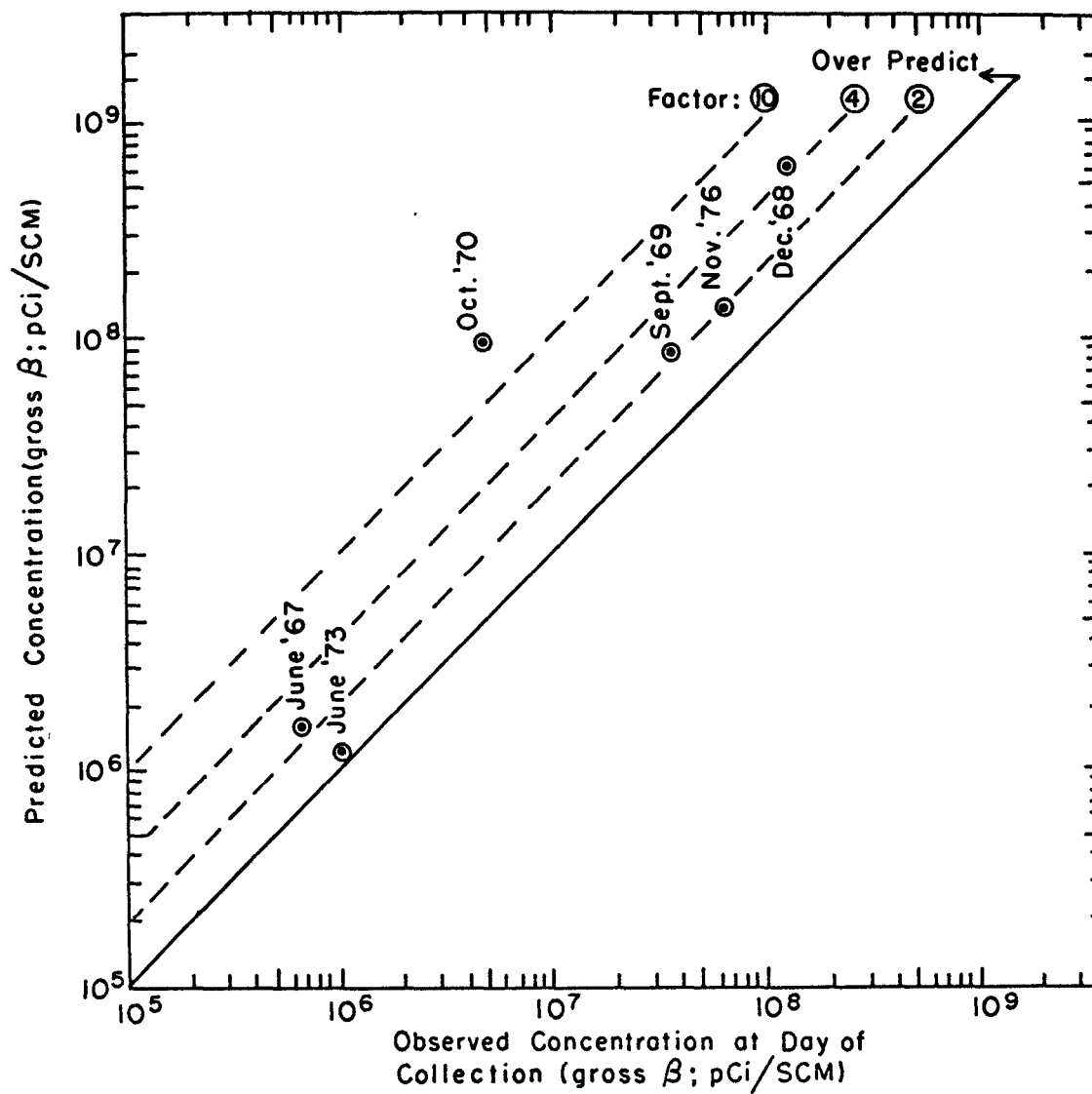


Figure 2. Comparison of observed and predicted concentrations at day of collection (gross beta, picocuries per standard cubic meter of air).

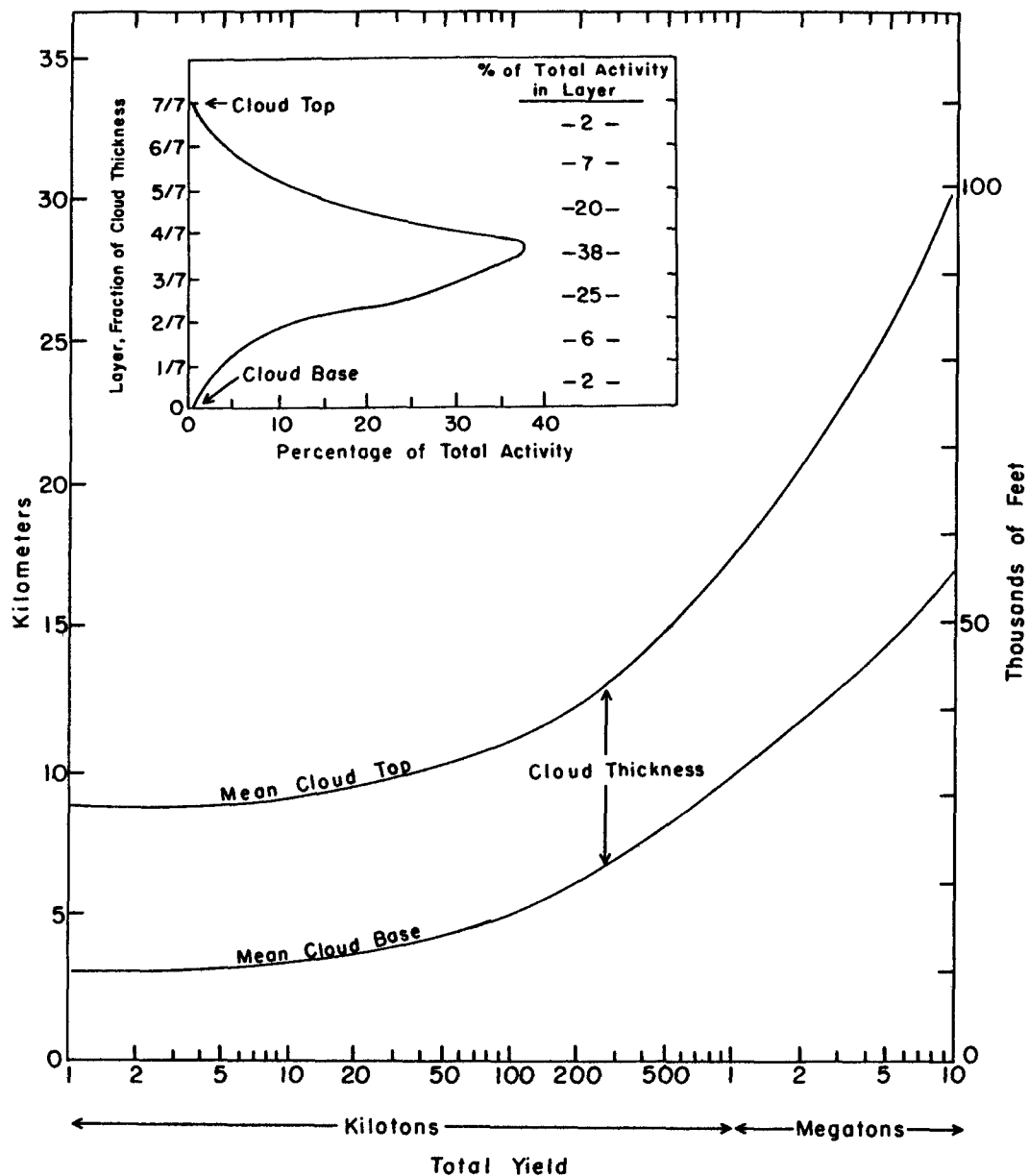
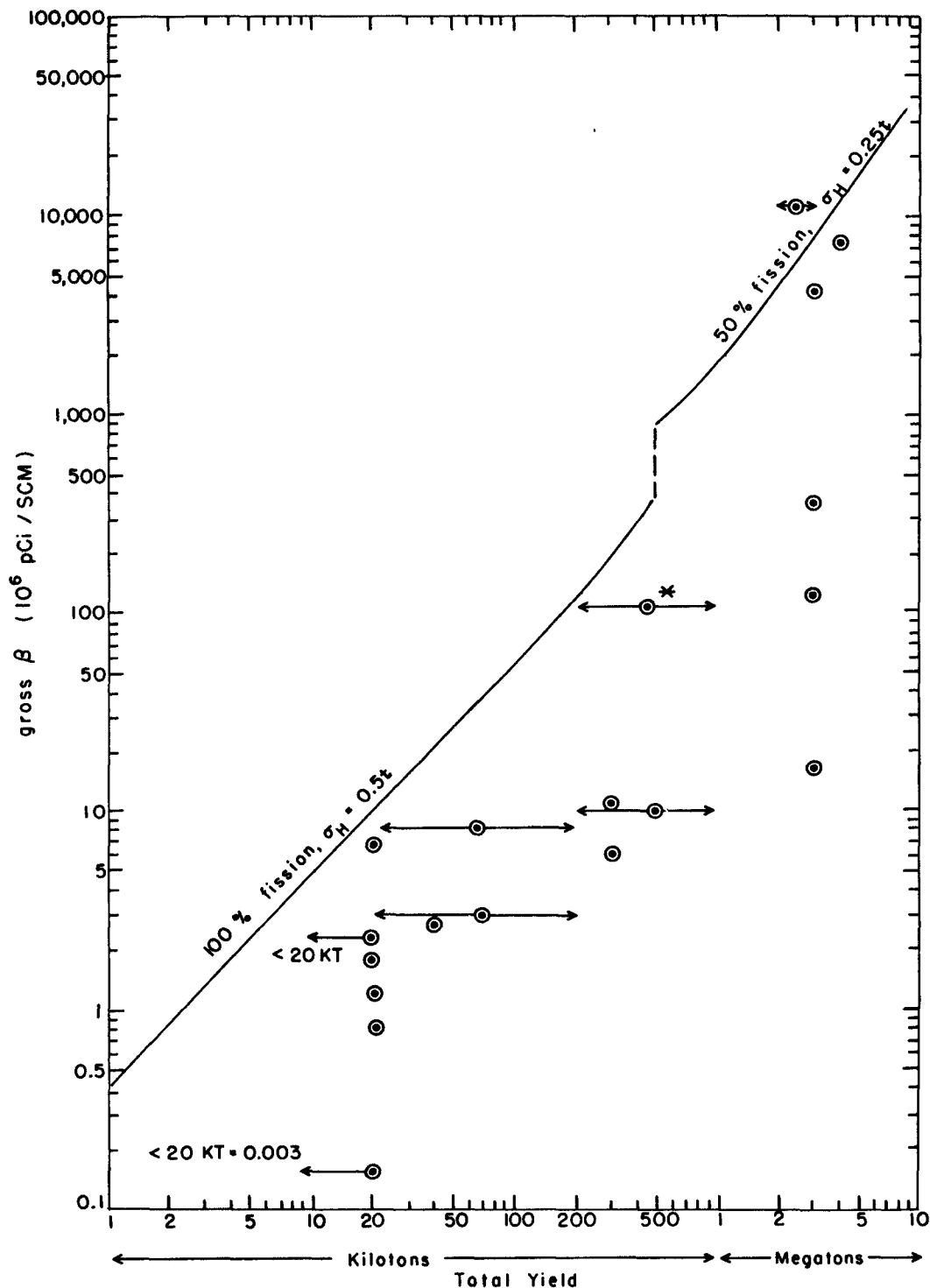


Figure 3. Mean cloud top and base as a function of total yield. Inset diagram presents the assumed initial vertical activity distribution within the cloud.



**Figure 4.** Observed concentrations (pCi/SCM) adjusted to one day after detonation. Solid curves denote calculated maximum credible concentration at one day assuming tropospheric dispersion (Eq. 2) for yields 500 KT or less and stratospheric dispersion (Eq. 3) for yields greater than 500 KT. (Asterisk - see text for explanation.)

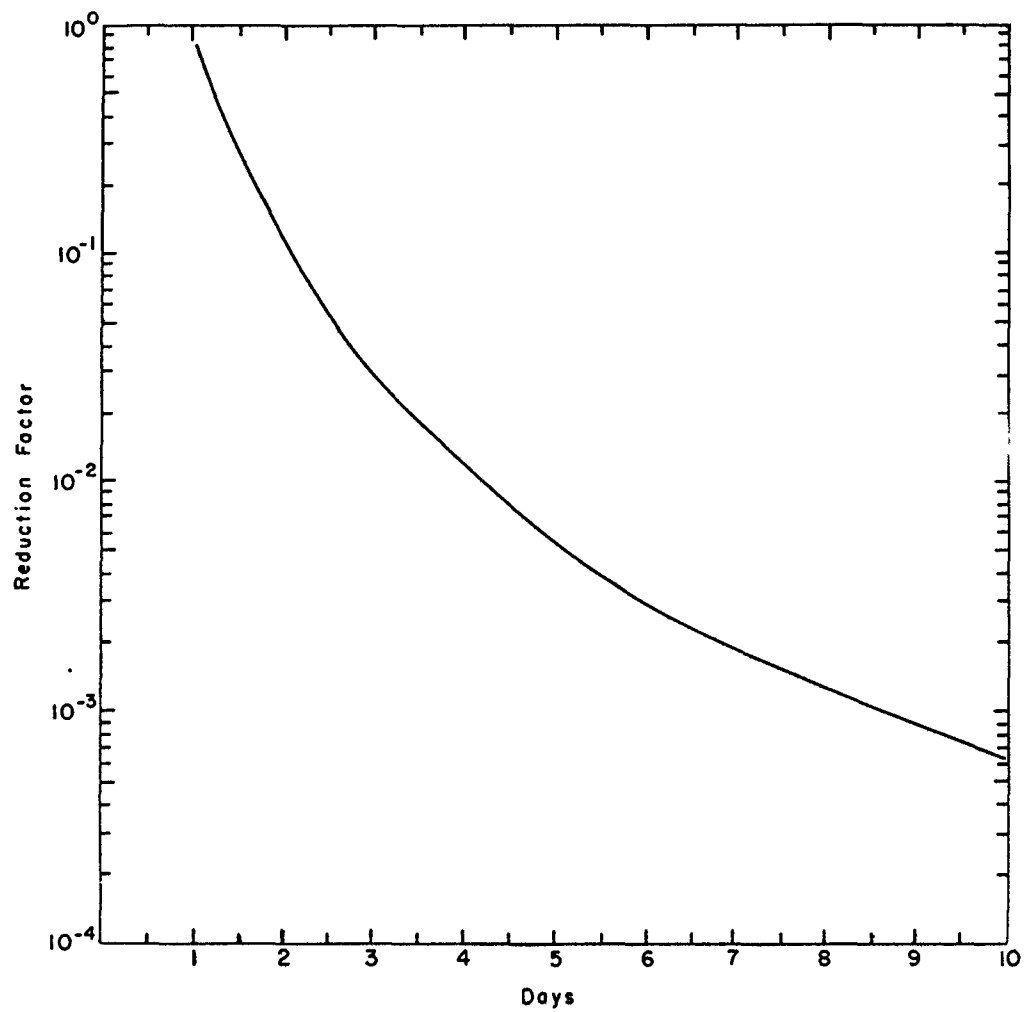


Figure 5. Reduction factor, combining horizontal dispersion and decay, adjusted to D+1.

## Appendix A

Estimate of Initial Vertical Activity Distribution for the High Yield Chinese Tests of June 1967; December 1968; September 1969; October 1970; and June 1973.

### 1-A. Introduction

The initial vertical distribution of radioactivity within a nuclear cloud as a function of yield has been estimated by Ferber (1965). Although the total amount of radioactivity produced by high yield tests may be known, its distribution in the vertical is based on very limited data observed in the equatorial Pacific. The estimated initial vertical distribution for a 3 MT air burst in temperate latitudes is given in Table A-1 (last column). This is based on Ferber's (1965) and Peterson's (1970) estimates. The bulk of activity is between 13 and 21 km (43 to 68 Kft) with the maximum at about 17 km (55 Kft).

An estimate of the initial vertical distribution of debris from the five Chinese 3 MT tests can be made from the measured distribution of radioactivity obtained within several weeks or months after each detonation. Latitudinal cross-sections of the distribution of Zr-95 (65 day half-life) at early times (from 2 to 16 weeks) following the Chinese high yield nuclear tests were used by Telegadas (1974, 1976) to estimate the initial vertical radioactivity distribution.

Certain cautions must be noted. First, it is assumed that the distribution observed along the 90°W to 150°W meridians (western portion of North America) from 2 weeks to 16 weeks after injection are representative of the average concentration in the nuclear cloud; that is, they are zonally well mixed. This assumption is somewhat doubtful, especially at early times. Second, it is assumed that within these time periods vertical

transport of debris is insignificant and the particle size of the debris is small enough for gravitational settling in the lower stratosphere (up to about 25 km) to be negligible.

(The reader is referred to the original papers by Telegadas (1974, 1976) for the detailed study. This appendix is a condensation of those papers.)

## 2-A. Estimate of Initial Vertical Distributions

### a) June 17, 1967 test.

The first series of aircraft and balloon sampling data were collected about 1 1/2 months after the June 17, 1967 test. As mentioned by Telegadas (1974), only about 15% of the estimated total Zr-95 produced by this test was intercepted along the sampling corridor at this time. It was not until 4 months after input that samples were again collected (early November 1967) and the Chinese debris encompassed virtually the whole northern hemisphere lower stratosphere. Even though four months had elapsed, significant fallout of this debris did not occur until January 1968 (Hardy, 1970) and therefore an integration of the concentration distribution for early November 1967 should represent the bulk of the Chinese 1967 input. The distribution pattern reveals that about 39,000 kilocuries (kCi) of Zr-95, equivalent to a fission yield of 1.6 MT, was contained in the stratosphere. The vertical distribution of this pattern shown in Figure A-1 with the breakdown of the distribution in 5,000 foot (1.5 km) increments given in Table A-1. The bulk of the activity for the June 17, 1967 test is seen to be between 14 and 22 km with the maximum at about 19 km.

b) December 27, 1968 test.

An estimate of the initial distribution of the Chinese December 27, 1968 event can be made from the radioactivity distribution about 1 1/2 months after input as given by Telegadas (1974). An integration of the early February 1969 pattern reveals that about 39,000 kCi of Zr-95 is contained in the stratosphere at this time. Significant fallout started by January 1969, that is, about a month before samples were collected along the sampling corridor. Between the time of the start of significant fallout and aircraft sampling the stratosphere lost about 15% of its Zr-95 burden. It is estimated that the December 27, 1968 event injected about 46,000 kCi of Zr-95 into the stratosphere, equivalent to a fission yield of about 1.9 MT.

The vertical distribution estimated for this event, is shown in Figure A-1 and also in Table A-1. The bulk of the activity is between 14 and 22 km with the maximum at about 17 km, about 2 km lower than the estimate for the June 17, 1967 test. The difference in the level of maximum debris is probably due to the initial meteorological conditions. The June 17, 1967 and December 27, 1968 events both had about the same total yield and injected their debris into a very stable stratosphere. The December 1968 test was conducted in a more stable upper troposphere with a lower reported tropopause height than the June 1967 event. Both of these factors would inhibit cloud growth.

c) September 29, 1969 test.

Stratospheric samples were collected along the aircraft sampling corridor in mid-October 1969, about 2 weeks after the September 29, 1969



test. An integration of the latitudinal distribution of radioactivity during this period indicated the stratospheric burden to be about 45,000 kCi of Zr-95, equivalent to a fission yield of 1.8 MT. Integration of the radioactivity pattern for the January 1970 sampling indicated the burden remains about 45,000 kCi, suggesting little or no debris had left the stratosphere, although there has been a significant redistribution of the radioactivity pattern.

The bulk of the activity for the September 29, 1969 event is seen to be between 16 and 22 km with the maximum at about 18 km (Table A-1 and Figure A-1).

d) October 14, 1970 test.

The first North American samples collected along the sampling corridor after the October 14, 1970 event occurred in nearly November 1970. The general circulation of the northern hemisphere lower stratosphere was such that virtually no debris from this event was encountered at this time except along the northerly legs of the sampling corridor. Stratospheric sampling was again conducted in late February 1971, at about four months after the October 1970 test. Significant stratospheric fallout began in February 1971, about the time that stratospheric sampling was being undertaken. An integration of the February 1971 radioactivity pattern indicated the stratospheric burden to be about 38,000 kCi of Zr-95, equivalent to a fission yield of about 1.5 MT. The vertical distribution at this time is shown in Figure A-1 and given in Table A-1. The bulk of the activity was between 11 and 21 km with the maximum at about 14 km. This vertical distribution is quite dissimilar to the estimates for the first three high yield events. It would appear that

between the time of injection of this fall test and the sampling period four months later the atmosphere had redistributed the debris sufficiently that the debris maximum moved from about 18 km to 14 km.

Radioactivity samples were collected in the stratosphere in the vicinity of Japan at very early times after this test (Leies, 1973). Based on very limited gross beta data, 2 to 9 days after the event, between 15 and 19 km, an estimate of the vertical distribution was made (Figure A-1) and is given in Table A-1. This vertical distribution is very similar to the estimate of the September 29, 1969 test. It must be emphasized that this estimate for the vertical distribution of the October 1970 event is based on the assumption that the main nuclear cloud was encountered at a very early time and the limited samples collected were indicative of the average concentrations in that cloud.

e) June 27, 1973 test.

The September 1973 stratospheric radioactivity distribution, about three months after input, was used to estimate the initial vertical distribution of radioactivity of the June 27, 1973 high yield event. An integration of the September 1973 pattern (Telegadas, 1976) indicated the stratospheric burden to be about 35,000 kCi of Zr-95, equivalent to a fission yield of 1.4 MT. The bulk of the activity for this event is seen to be between 15 and 22 km with the maximum at about 19 km (Figure A-1 and Table A-1). As can be seen in Figure A-1, this detonation had a vertical radioactivity distribution similar to that for the June 17, 1967 test.

In this paper, the assumed initial distribution as shown in the insert to Figure 3 was based on a composite of the first four high yield Chinese

nuclear tests. Including the distribution for the fifth test (June 27, 1973) in this average distribution would not alter it significantly.

Table A-1. Observed Vertical Activity Distribution for the 3 MT Total Yield Chinese Nuclear Tests.

Alt. (Kft)	Test Weeks <sup>(3)</sup>	6/17/67 (16)	12/27/67 (6)	9/29/69/ (2)	10/14/70 (16)	10/14/70 <sup>(1)</sup> (1)	6/27/73 (11)	ASSUMED <sup>(2)</sup>
<u>Percent of Total Activity</u>								
85.1	85	0.1						
82.5	80	0.6	0.1	0.1				
77.5	75	3.5	1.3	3.0			0.8	
72.5	70	14.5	6.0	7.4	1.2	0.3	7.0	2
67.5	65	28.1	16.6	17.0	6.5	18.9	33.7	10
62.5	60	27.4	28.6	39.3	11.4	55.1	39.2	22
57.5	55	15.1	32.6	31.2	18.0	23.4	14.9	32
52.5	50	6.9	11.4	1.8	22.5	2.2	3.1	22
47.5	45	3.0	2.8	0.2	24.7	0.1	1.0	10
42.5	40	0.6	0.6		13.7		0.3	2
37.5	35	0.2			2.0		0.1	
35.0								
TOTAL:		100	100	100	100	100	100	100

ESTIMATED STRATOSPHERIC BURDEN OF ZR-95 ON TEST DATE

Test Date	Kilocuries	Estimated Fission Yield (Megatons) <sup>(4)</sup>
June 16, 1967	39,000	1.6
Dec. 27, 1967	46,000	1.9
Sept. 29, 1969	45,000	1.8
Oct. 14, 1970	38,000	1.5
June 27, 1973	35,000	1.4

(1) Estimated from limited data collected several days after the event.

(2) Assumed distribution after Ferber, 1965 and Peterson, 1970.

(3) Weeks after test.

(4) Based on 24,600 kilocuries of Zr-95 per megaton of fission.

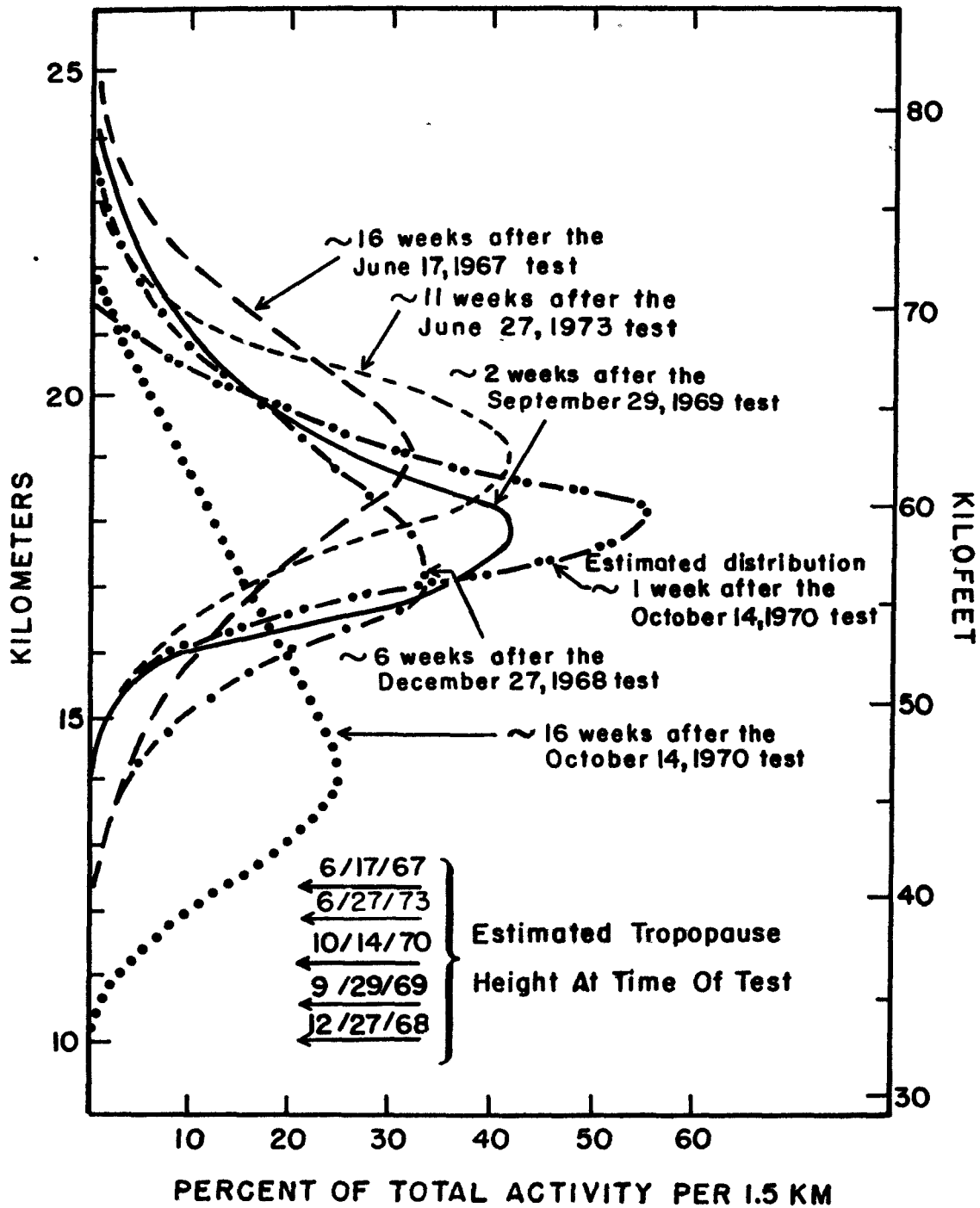


Figure A-1. Observed vertical Zr-95 activity distribution for five high yield Chinese nuclear tests.

STRONTIUM-90 IN HUMAN BONE  
- 1976 Results for New York City and San Francisco -

by B. G. Bennett (HASL)

ABSTRACT

Results are presented of determinations of Sr-90 content of 95 specimens of vertebrae obtained during 1976 in New York City and San Francisco. The average Sr-90 to Ca ratios for adult vertebrae are 1.06 pCi/g Ca in New York and .73 pCi/g Ca in San Francisco, little changed from the average values of the previous year. Average Sr-90 concentrations in children's bone are now little different from the adult values. A two compartment bone model, which accounts for both short and long term retention of Sr-90 in bone, is used to describe the variations of Sr-90 content of bone and provide correlation with dietary Sr-90 intake. Regression analysis of the 16 years of survey data provides values of the relative retention of dietary Sr-90 and the effective bone turnover rates. The bone model gives satisfactory description of observed Sr-90 levels and allows reliable assessment of the long-term behavior of Sr-90 in man.

Human vertebrae specimens obtained in New York City and San Francisco have been analyzed by HASL for Sr-90 content since 1961.<sup>1-12</sup> The sampling during this 16 year period of varying Sr-90 fallout deposition at these locations provides valuable records of variations in Sr-90 content of bones of children as well as adults. A food sampling program to determine the intake of Sr-90 in the diet of residents of the two cities has also been conducted by HASL during this same time period, so that correlations between Sr-90 intake and bone concentrations can be determined. More accurate description of the long-term behavior of Sr-90 in bone is achieved as the sampling programs continue.

## Results for 1976

During 1976, 95 specimens of human vertebrae were analyzed, including 3 from children and 9 from adults obtained in New York City and 50 from children and 33 from adults obtained in San Francisco. Fewer samples than usual were available during the year in New York City. A summary of the results of Sr-90 determinations is given in Table 1. The individual analyses are listed in Table 2 and are plotted in Figures 1 and 2.

The Sr-90 to Ca ratios for adults are relatively constant, as has previously been observed. The variations about the mean are typical of such survey measurements. The average values and standard deviations for adult vertebrae in 1976 were  $1.06 \pm .26$  pCi/g Ca in New York and  $.73 \pm .26$  pCi/g Ca in San Francisco.

There has been some indication of slightly higher values for young adults in New York during the last several years. These individuals were children during the period of greatest Sr-90 deposition. Adult metabolism for the entire fallout period is represented by those individuals  $\geq$ age 20 in 1954, corresponding to  $\geq$ age 42 in 1976.

The Sr-90 to Ca ratios for children's bone show some age dependence with slightly higher values for children than adults in New York. In San Francisco the Sr-90 to Ca ratios for children are little different from the adult values. The approximate average values of Sr-90 concentration have been indicated in Figures 1 and 2.

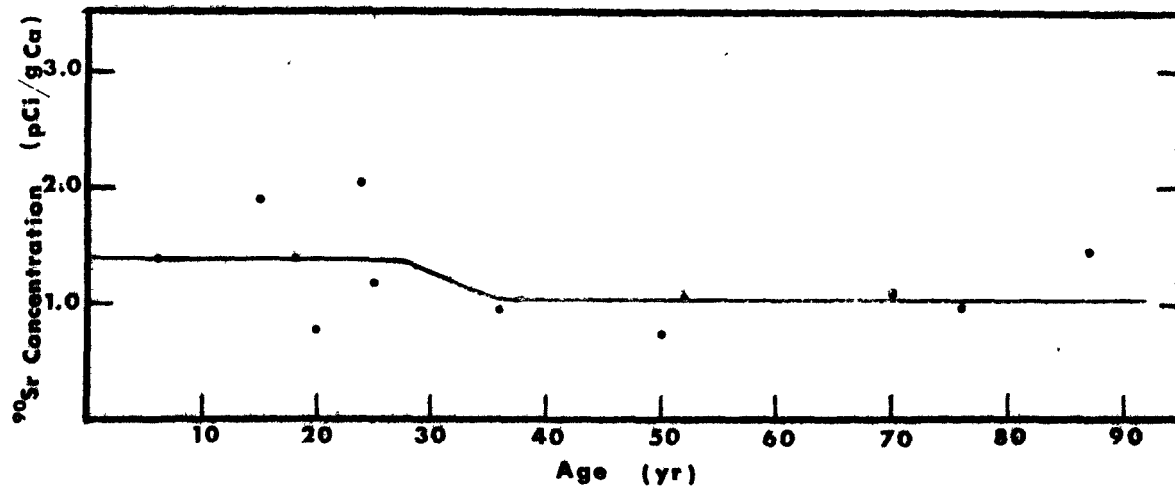


Figure 1. Strontium-90 in New York Vertebrae - 1976.

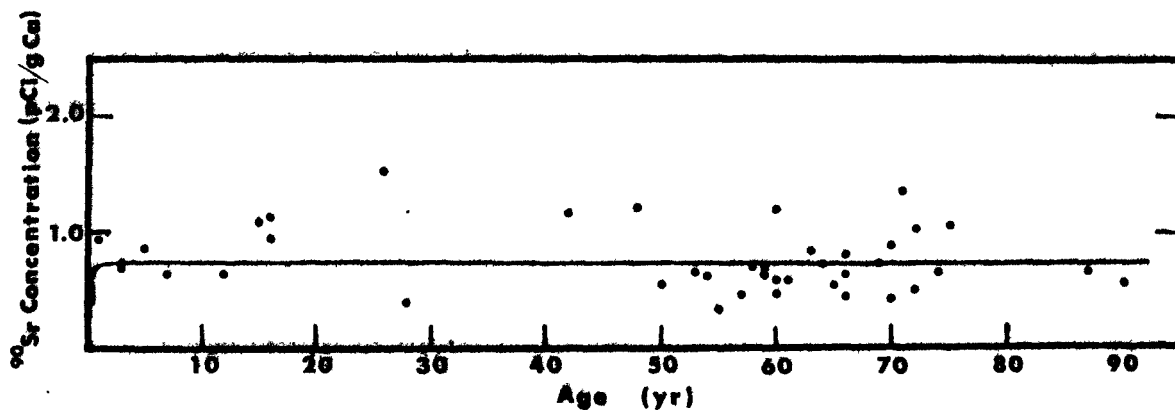


Figure 2. Strontium-90 in San Francisco Vertebrae - 1976.

The higher Sr-90 concentration values in New York bone compared to San Francisco bone correspond to greater dietary Sr-90 intake. The Sr-90 to Ca ratios in total diet in New York and San Francisco during 1976 were 8.3 pCi/g Ca and 3.1 pCi/g Ca, respectively<sup>(13)</sup>. These ratios in diet have decreased an average of 7 to 9% per year since 1966.



The usual practice in correlating the Sr-90 levels in bone and diet is to refer the skeletal levels during the year n to the dietary Sr-90 intake from midyear of the year n-1 to midyear of the year n. Deaths in the early part of the year are then not compared with diet levels not yet experienced. Table 3 lists the diet results based on consumption estimates specific to the particular urban regions.

### Bone Model

An improved bone model was formulated to correlate the Sr-90 concentrations in diet and bone<sup>(8)</sup>. The model as it applies to adult vertebrae is described by the equation

$$B_n = cD_n + g \sum_{m=0}^{\infty} D_{n-m} e^{-m\lambda}$$

where

$B_n$  = Sr-90 concentration in vertebrae in the year n (pCi)

$D_n$  = Sr-90 concentration in diet from midyear in the year n-1 to midyear in the year n (pCi)

$c$  = short-term retention of Sr-90 in bone

$g$  = long-term retention of Sr-90 in bone

$1-e^{-\lambda}$  = effective removal rate for Sr-90 in bone including radioactive decay ( $\text{yr}^{-1}$ )

The formula describes a two compartment model, one compartment associated with short-term retention of Sr on bone surfaces and another compartment in which the Sr is more tightly retained in bone. The parameters  $c$  and  $g$  are independent and not related to the previously used concept of Observed Ratio, since retention can also be associated with processes other than new bone formation. The effective removal rate

or turnover rate for Sr-90 in bone is related to the rate of bone remodeling, but since Sr-Ca discrimination and re-utilization are not explicit in the model, the removal rate can be considered to provide only an upper limit estimate of the actual bone renewal rate. The factors  $c$ ,  $g$ , and  $\lambda$  are constant for adults ( $\geq$  age 20 years) but are age dependent for children.

Variations of the above bone model were investigated, such as the inclusion of an exponential in the short-term retention term. The best fit in this form, however, was with a very high order exponential, indicating essentially complete turnover of the short-term component during the course of a year.

The above formula for the bone model maintains a desired simplicity and yet gives adequate description of the year-to-year changes in Sr-90 content of bone. The values obtained for the model parameters allow reasonable interpretation.

#### Strontium-90 in Adult Vertebrae

The observed Sr-90 concentrations in adult vertebrae are shown in Figure 3. Additional approximate values for the earliest years of contamination (1954-59) in New York from the data of Kulp and Schulert<sup>(14)</sup> have also been included. The adult data include only samples from individuals age 20 years or older in 1954, thus representing adult metabolism for the entire contamination period.

A definite decreasing trend in the observed values is apparent since 1965, corresponding to decreases in dietary Sr-90 intake. The relatively large standard deviations about the average values preclude extremely accurate determination of the actual decreases and thus the effective removal rates, however these are becoming more

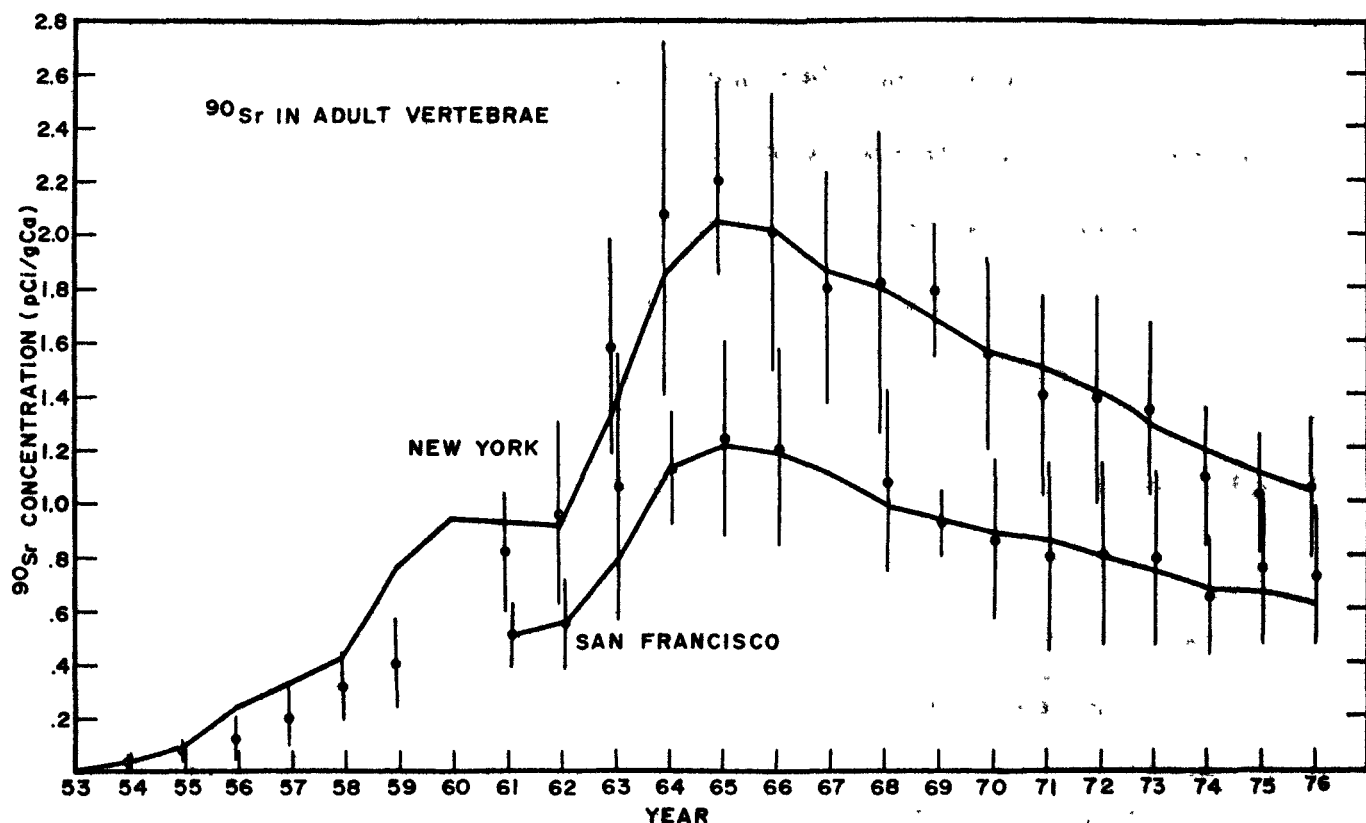


Figure 3. Strontium-90 in adult vertebrae - observations (points with standard deviations) and bone model predictions (solid lines).

firmly established as additional years of data accumulate during periods of decreasing Sr-90 levels in diet.

From regression analysis of the measured New York adult diet and vertebrae values through 1976 in the above bone model, one infers a removal rate of Sr-90 from adult vertebrae of 23% per year. A removal rate of 15% per year gives the best fit to the San Francisco adult data through 1976. The best fits to the observed data using the bone model are shown in Figure 3. The values of the bone model parameters are included in Table 4.

The model shows peak Sr-90 concentrations in adult vertebrae in 1965, in agreement with observations. Subsequent decreases are achieved with a removal rate which is reasonable, in view of the remodeling and diffusion processes occurring in bone. The bone model appears to be quite responsive to Sr-90 levels in diet and gives very satisfactory fits to the observed Sr-90 levels in vertebrae.

### Strontium-90 in Children's Bone

The Sr-90 concentration in children's bone can be determined by the formula

$$B_{i,n} = (c_i + g_i)D_{i,n} + [B_{i-1,n-1} - c_{i-1} D_{i-1,n-1}]e^{-\lambda t}$$

The bone model parameters are defined as before. The subscript  $i$  indicates the age dependence.

The initial Sr-90 concentration in the newborn must be determined separately. For this an empirical relationship with the mother's diet can be used. The Sr-90 to Ca ratio in bone of newborn varies from .1 to .2 times the Sr-90 to Ca ratio in diet of the mother during the year prior to the birth. An average of about .15 is obtained from the survey data. The relationship between newborn and adult bone Sr-90 concentrations has been somewhat more variable. In San Francisco, where a large number of newborn samples are obtained, the ratio of Sr-90 concentration in newborn vertebrae to adult vertebrae has ranged from .5 to .9 with an average of .7 since 1967.

From the regression results for children, one infers that a one compartment, single exponential model ( $c_1 = 0$ ) applies to children under age 8 years. The one compartment formulation of the above model closely corresponds to the previously used Rivera bone model<sup>(1)</sup>. The undifferentiated nature of bone of young children and the relatively high bone renewal rates justify the one compartment treatment.

The Sr-90 turnover rates and relative retention as a function of age are shown in Figures 4 and 5 and are listed in Table 4. The turnover rates reflect bone growth activity and are highest for the youngest children. Nearly 100% per year turnover rate is indicated for the 0-1 year age range. A relatively high turnover rate, about 45%, is maintained through the pre-teenage years. The values then decrease to the adult value.

The relative retention of Sr-90 in bone, shown in Figure 5, is the fractional retention of the dietary Sr-90 intake ( $c_1 + g_1$ ) per gram of skeletal calcium. The highest efficiency for Sr-90 retention is obtained for the youngest children. Increased efficiency associated with increased growth is also indicated for children in the early teenage years.

The magnitude of the relative retention was determined by assuming that vertebrae is representative of entire skeletal behavior. This assumption becomes less satisfactory for older children and adults. Initial estimates of body burden will be high and estimates at later times will be low, assuming less initial retention and slower turnover rates for compact bone. Integral results should be more representative for the entire skeleton, such as the cumulative dose results following a period of Sr-90 intake of one year or more.

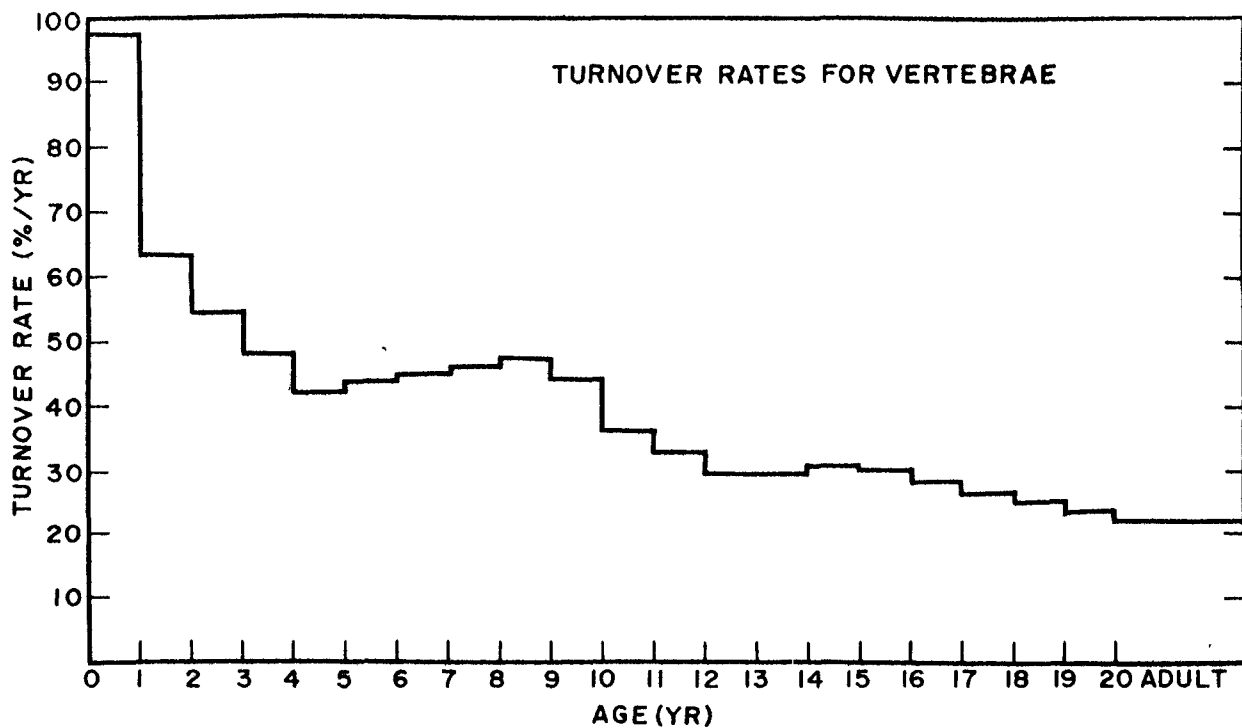


Figure 4. Turnover or effective removal rates for Sr-90 in vertebrae inferred from the two compartment bone model.

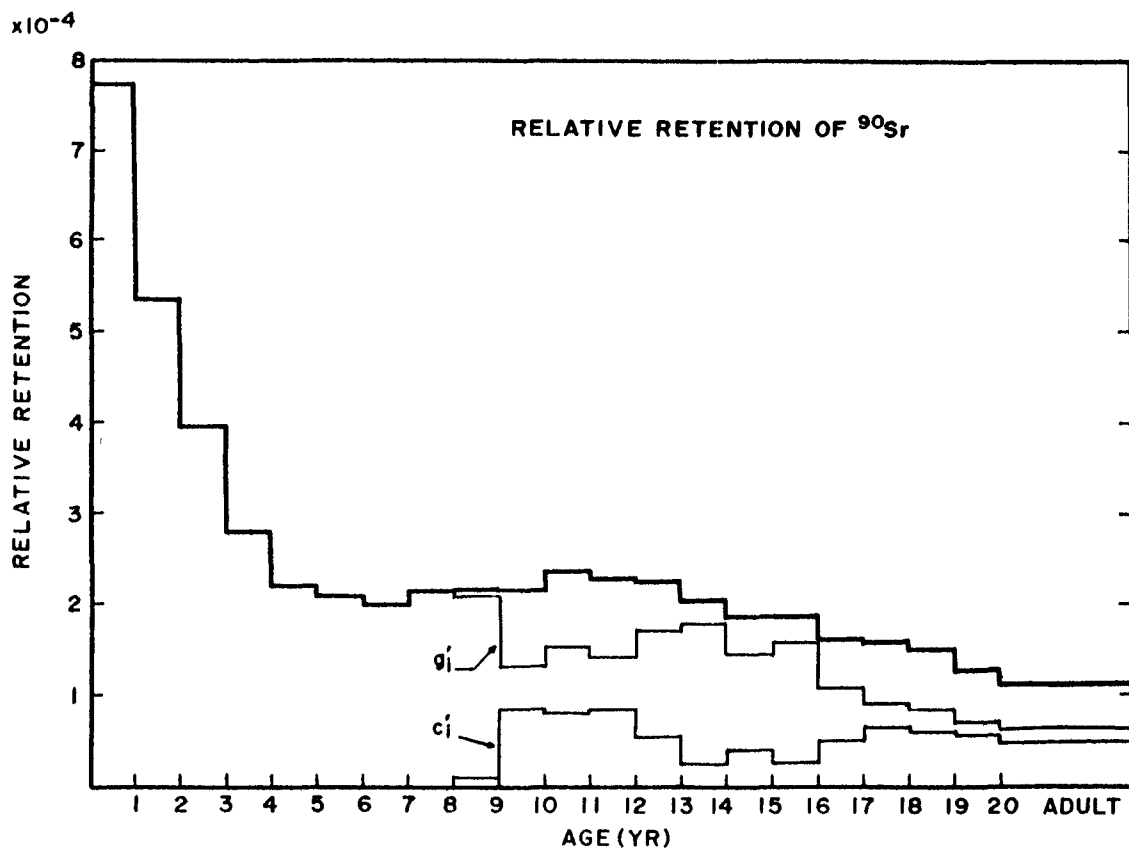


Figure 5. Retention of Sr-90 per gram skeletal Ca. The upper histogram is the total relative retention;  $c_i$  is the short-term component and  $g_i$  is the long-term component.

### Anticipated 1977 Results

According to the bone model, the concentration level of Sr-90 in adult vertebrae in 1976 was due 70% to the residual body burden and 30% to dietary intake during the most recent year. Half this latter amount contributes to the long-term burden, so that 85% of the current total concentration can be expected to be affected by the 23% per year removal rate from bone and 2% per year radioactive decay. For New York this amounts to  $1.06 \text{ pCi/g Ca} \times .85 \times .75 = .68 \text{ pCi/g Ca}$  remaining during 1977. The dietary intake of Sr-90 for the 1976-77 year, with the results for the final quarter yet to be obtained, is estimated to be 3000 pCi. This times c+g (.12) divided by the skeletal Ca content (1078g) gives a contribution to the Sr-90/Ca ratio in bone of .33 pCi/g Ca. The total concentration in adult vertebrae indicated by the bone model for 1977 is thus 1.01 pCi/g Ca, a 5% decrease from 1976. A similar decrease can be expected in San Francisco.

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Table 1  
<sup>90</sup>Sr to Calcium Ratios in Human Vertebrae  
- Summary of 1976 Data -

Age Range at death	New York		San Francisco	
	No. of Specimens	pCi <sup>90</sup> Sr g Ca	No. of Specimens	pCi <sup>90</sup> Sr g Ca
0 - 1 yr			40	.51
1 - 2			1	.94
2 - 3				
3 - 4			2	.71
4 - 5				
5 - 6			1	.85
6 - 7	1	1.39		
7 - 8			1	.63
8 - 9				
9 - 10				
10 - 11				
11 - 12				
12 - 13			2	.64
13 - 14				
14 - 15				
15 - 16	1	1.90	1	1.09
16 - 17			2	1.03
17 - 18				
18 - 19	1	1.40		
19 - 20				
20 - 29	3	1.33	2	.97
30 - 39	1	.95		
40 - 49			2	1.20
50 - 59	2	.90	8	.58
60 - 69			12	.70
70 - 79	2	1.03	7	.85
80 - 90	1	1.45	2	.62
<u>Adult</u>				
≥ Age 20 in 1976	9	1.14	33	.74
≥ Age 20 in 1954	5	1.06	31	.73

Table 2  
<sup>90</sup>Sr to Calcium Ratios in Human Vertebrae

Age at death	HASL No.	Month of death	pCi <sup>90</sup> Sr gCa	Age at death	HASL No.	Month of death	pCi <sup>90</sup> Sr gCa
<u>New York - 1976</u>				<u>San Francisco - 1976</u>			
6y	HB5400	1	1.39	12y	HB5421	7,9	.64
15y	HB5401	3	1.90	15y	HB5426	9	1.09
18y	HB5402	3	1.40	16y	HB5422	1	.96
					HB5427	12	1.13
20y	HB5403	1	.77	26y	HB5428	11	1.53
24y	HB5404	3	2.04	28y	HB5429	10	.40
25y	HB5405	2	1.19				
36y	HB5406	2	.95	42y	HB5424	4	1.18
50y	HB5407	3	.73	48y	HB5430	10	1.22
52y	HB5408	1	1.07	50y	HB5431	1	.56
70y	HB5409	3	1.09	53y	HB5432	11	.68
76y	HB5410	1	.97	54y	HB5433	7	.63
87y	HB5411	1	1.29, 1.61*	55y	HB5435	6	.32
				57y	HB5436	9	.46
				58y	HB5437	12	.69
				59y	HB5438	2	.63
					HB5439	4	.68
<u>San Francisco - 1976</u>				60y	HB5440	5	1.20
0-1 d.	HB5412	2,2,2,3	.41		HB5441	5	.60
		4,5,5,5			HB5442	12	.47
	HB5413	6,6,7,7	.41	61y	HB5443	3	.58
		8,8,8,8		63y	HB5444	10	.84
	HB5414	9,9,9,	.45	64y	HB5445	2	.72
		11, 11		65y	HB5446	11	.54, .57*
1-2 mo.	HB5415	2,5,5,6,6,6	.51	66y	HB5447	9	.81
		6,8,9,11,12			HB5448	11	.64
3-5 mo.	HB5416	1,2,6,6	.71		HB5449	11	.44
		9,11,11		69y	HB5450	4	.74
9-18 mo.	HB5417	4,8	.94		HB5451	10	.74
3y	HB5418	8	.74	70y	HB5434	11	.89, .91*
	HB5419	4	.68		HB5453	12	.43
5y	HB5425	11	.85	71y	HB5454	9	1.36
7y	HB5420	5	.63	72y	HB5455	10	1.02
					HB5456	12	.50
				74y	HB5457	9	.66
				76y	HB5458	10	1.06
				87y	HB5459	9	.66
				90y	HB5460	5	.57

\*blind duplicate analysis

Table 3

Dietary Sr-90 Intake<sup>1</sup>

Year	New York <sup>2</sup>		San Francisco <sup>3</sup>	
	Per Capita	Adult	Per Capita	Adult
1954	1.0	1.0		
1955	1.9	2.0		
1956	5.0	5.2		
1957	5.5	5.7		
1958	6.4	6.6		
1959	13.4	13.8		
1960	13.7	14.1		
1961	10.4	10.7	3.6	3.7
1962	9.8	9.9	4.1	4.2
1963	18.7	18.9	9.4	9.6
1964	29.5	29.8	13.8	14.2
1965	26.0	26.9	11.6	11.9
1966	20.3	21.1	8.3	8.7
1967	15.7	16.4	6.3	6.6
1968	14.8	15.5	4.0	4.2
1969	13.0	13.6	4.2	4.5
1970	12.0	12.4	4.0	4.2
1971	12.1	12.6	4.4	4.6
1972	11.3	11.8	3.6	3.9
1973	9.6	10.1	3.2	3.4
1974	8.9	9.4	2.9	3.0
1975	8.5	8.8	3.3	3.5
1976	7.9	8.3	2.8	3.0

<sup>1</sup>Estimates (pCi/gCa) are for mid-year of the previous year to mid-year of the year listed. 1954-59 values are estimated from milk data<sup>(15)</sup>. 1960-75 values are results of the HASL Quarterly Diet Sampling Program.

<sup>2</sup>Urban N. E. consumption statistics<sup>(16)</sup>  
 per capita milk consumption = 136 kg/yr  
 adult milk consumption = 93 kg/yr

<sup>3</sup>Urban West consumption statistics<sup>(17)</sup>  
 per capita milk consumption = 108 kg/yr  
 adult milk consumption = 74 kg/yr

Table 4

## Parameters of the Bone Model

Age (yr.)	$\lambda_i$	$c_i$	$g_i$	Skeletal Calcium $Ca_i$ (gCa)	Dietary Calcium D $Ca_i$ (gCa/yr)	Relative Retention $(c_i + g_i)/Ca_i$ ( $\times 10^{-3}$ )	Turnover Rate $(1 - e^{-\lambda_i}) - .024$ (%/yr)
0-1	6.72	0	.077	100	320	.77	97
1-2	1.08	0	.079	147	328	.53	64
2-3	.84	0	.071	179	336	.40	54
3-4	.71	0	.056	201	344	.28	48
4-5	.60	0	.048	219	352	.22	43
5-6	.62	0	.050	239	360	.21	44
6-7	.64	0	.053	264	368	.20	45
7-8	.66	0	.063	297	376	.21	46
8-9	.69	.004	.071	341	384	.22	47
9-10	.63	.034	.052	396	387	.22	44
10-11	.49	.037	.072	463	387	.24	36
11-12	.44	.045	.077	539	387	.23	33
12-13	.39	.033	.107	624	387	.22	30
13-14	.39	.017	.128	715	387	.20	30
14-15	.41	.032	.120	806	387	.19	31
15-16	.40	.024	.143	894	387	.19	31
16-17	.37	.051	.106	973	387	.16	29
17-18	.35	.069	.095	1039	376	.16	27
18-19	.32	.066	.093	1073	365	.15	25
19-20	.30	.060	.076	1078	354	.13	24
Adult	.29	.051	.068	1078	342	.11	23

# WORLDWIDE DEPOSITION OF $^{90}\text{Sr}$ THROUGH 1976

by H. W. Feely (HASL)

## ABSTRACT

The total deposition of  $^{90}\text{Sr}$  on the surface of the earth during 1976 was about 46 kilocuries. The worldwide cumulative deposit remained at about 11.5 megacuries as the amount of previously deposited  $^{90}\text{Sr}$  which decayed radioactively only slightly exceeded the amount of the new deposit. The late 1976 Chinese atmospheric nuclear tests may have contributed about 12 kilocuries to the  $^{90}\text{Sr}$  deposition during 1976.

## INTRODUCTION

The Health and Safety Laboratory (HASL), has maintained a worldwide network of samplers since 1957 to collect fallout to be measured for  $^{90}\text{Sr}$ . Based upon data from this network, an estimate has been made each year since 1958 of the annual worldwide deposition and of the cumulative deposit of  $^{90}\text{Sr}$  (e.g., reference 1). In general both the annual and cumulative values have been reasonably well verified when comparison with results from other programs has been possible (e.g., references 2,3). For example, in Table 1 estimates are given of the worldwide annual deposition and cumulative deposition of  $^{90}\text{Sr}$  as made by HASL and by the United Kingdom Atomic Energy Authority (UKAEA) for the years 1970 to 1975. Agreement between the two laboratories is excellent in some years and poor in others,

indicating the degree of uncertainty in making such estimates. The differences average out over intervals of a few years, however, so that the estimates of cumulative deposition are in good agreement.

#### BASIS OF CALCULATION

The HASL network of samplers of  $^{90}\text{Sr}$  fallout included 72 sites during 1976: 47 in the Northern and 25 in the Southern Hemisphere. The sampling sites are indicated on the map on page A-2 of the Appendix to this report. The samples are collected at these sites by exposing to the environment either a steep-walled, stainless steel pot or a funnel which drains through a column containing Dowex-50 ion exchange resin. At the end of each month the total material collected in each pot and each exposed column of ion exchange resin is sent to HASL.

Radiochemical analyses of the samples in this program are performed under contract with commercial laboratories. For the fiscal year ending September 30, 1977 this work was carried out by U. S. Testing Co., Richland, Washington. The quality of these data, which is monitored by the submission of "blind" blanks and standard samples along with ordinary samples, is reported on annually<sup>(4)</sup>. All of the primary monthly precipitation and

radiochemical data which have been accumulated since the inception of the pot and column network are listed and updated in the appendix to each HASL Environmental Quarterly. A summary of existing results, averaged over 10 degree latitude bands was reported in 1971<sup>(5)</sup>.

Various methods for calculating the total fallout on the surface of the earth from limited data were discussed by Volchok<sup>(6)</sup>. The differences between the results given by the different methods were found generally to be small. The approach used at HASL assumes that our sampling sites are representative, on the average, of the fallout in the ten degree latitude band within which they are located. The average monthly  $^{90}\text{Sr}$  deposition (in  $\text{mCi}/\text{km}^2$ ) for each such latitude band is multiplied by the area of the band (in  $\text{km}^2$ ) to give the total monthly deposition in that band. Values are obtained for the unsampled polar areas north of  $80^\circ\text{N}$  and south of  $70^\circ\text{S}$  by extrapolating a smoothly decreasing  $^{90}\text{Sr}$  deposition to zero at the poles. Summing the calculated totals for the 10 degree latitude bands gives the worldwide deposit for each month. No corrections are made for any possible precipitation biases of the sampling sites or for any postulated oceanic effects on local rates of deposition. The  $^{90}\text{Sr}$  global inventories calculated



by this method are in reasonably good agreement with the inventories calculated based on the estimated total production of  $^{90}\text{Sr}$  by all atmospheric tests of nuclear weapons and the estimated burdens of  $^{90}\text{Sr}$  in the atmosphere (Table 2).

It has been mentioned in earlier reports that the ion exchange columns have seldom received optimum treatment in the field, and consequently most of the data for samples from these collectors are systematically low. To compensate for this, the computed average depositions for 10 degree latitude bands are corrected according to the relative numbers of pots and columns used as a basis for calculating them.

## RESULTS

The average monthly deposition of  $^{90}\text{Sr}$  during 1976, in units of millicuries per square kilometer ( $\text{mCi}/\text{km}^2$ ), is listed for each 10 degree latitude band in Table 3. The number of sampling stations represented is shown in parentheses, and values derived by extrapolation, as discussed above, are indicated by (E). The hemispheric average monthly deposition is listed in the last column.

The total monthly deposition of  $^{90}\text{Sr}$  within each 10 degree latitude band, in units of kilocuries (kCi), is listed in Table 4. Again the number of sampling stations represented, or an (E) indicating values obtained by extrapolation, is given in parentheses. The next to last column presents the total monthly fallout in the hemisphere, and the last column gives the cumulative total  $^{90}\text{Sr}$  fallout on the surface of the earth within the hemisphere, corrected for radioactive decay since time of deposition. In Table 5 these cumulative total  $^{90}\text{Sr}$  fallout estimates are subdivided according to 10 degree latitude band.

Table 6 contains the calculated annual  $^{90}\text{Sr}$  deposition in units of megacuries (MCi) within each hemisphere and worldwide, and the cumulative deposit of  $^{90}\text{Sr}$  on the surface of the earth at the end of each year, both within each hemisphere and worldwide. The data presented cover the period from the beginning of the HASL pot and column program in 1958 through 1976.

#### DISCUSSION

The worldwide fallout of  $^{90}\text{Sr}$  during 1976 was 46 kCi, with 28 kCi falling in the Northern, and 18 kCi falling in the Southern Hemisphere. There were three announced atmospheric nuclear detonations during 1976, all carried out by the People's Republic of

China: a <20 kiloton (kT) test on January 23, a 20-200 kT test on September 26, and a high yield test of 4 megatons on November 17. If we assume that each of these tests had a fission yield of 50%, that the  $^{90}\text{Sr}$  yield was 0.11 Curie per ton of fission yield<sup>(7)</sup>, and that none of the debris from any of these tests fell out locally, the worldwide fallout of  $^{90}\text{Sr}$  from the three tests would be <1.1 kCi, 1.1 to 11 kCi, and 220 kCi. If the fission yields were less than 50% or local fallout occurred, the contribution of these tests would be reduced. The January 23rd test should have had a negligible effect on the total deposition of  $^{90}\text{Sr}$  during 1976 because its  $^{90}\text{Sr}$  yield was quite small compared to the 46 kCi total deposit for the year. The effect of the November 17 test was probably also small, both because it came late in the year, and because almost all of the debris from it evidently was injected into the stratosphere. No clear indication of any debris from this test was found in filter samples collected during 1976 as part of the HASL Surface Air Sampling Program. On the other hand, all or most of the debris from the September 26 test should have been injected into the troposphere, and much of it could have been deposited as worldwide fallout during the last three months of 1976. This debris was sampled by the HASL Surface Air Sampling Program during October to December 1976.

The data in Table 3 indicate that the deposition rate of  $^{90}\text{Sr}$  during 1976 was highest during the last quarter. For the most part, this may reasonably be attributed to deposition of fallout from the September 26 test and perhaps the November 17 test. During the first nine months of 1976, 17.6 kCi of  $^{90}\text{Sr}$  were deposited in the Northern Hemisphere. This is 33% of the 54.2 kCi deposited there<sup>(8)</sup> during the equivalent period of 1975. Since 4.1 kCi were deposited during the last quarter of 1975, we might have expected that about 33% of this quantity, or about 1.3 kCi, to be deposited in the Northern Hemisphere during the last quarter of 1976. Instead 10.5 kCi were actually deposited, suggesting that about 9 kCi originating from the late 1976 tests were deposited in the Northern Hemisphere. An equivalent calculation using data for the Southern Hemisphere indicates that a deposition of about 3.1 kCi would have been expected but 6.0 kCi were deposited. Thus an excess of about 3 kCi for the Southern Hemisphere and 12 kCi worldwide of  $^{90}\text{Sr}$ , compared to the amount expected based on data for 1975 and for the first three quarters of 1976, was deposited during the last quarter of 1976. Presumably this represents a contribution by the November 17 and especially the September 26, 1977 Chinese tests.

Using the approach described above, we would have expected a total  $^{90}\text{Sr}$  deposition of 4.4 kCi worldwide during the last quarter

of 1976 had the Chinese weapons tests of late 1976 not occurred. This would have given a total deposit worldwide for 1976 of 34 kCi. Even with the late 1976 tests, only 46 kCi were deposited worldwide. As indicated by the data in Table 6, this made 1976 the year with the lowest deposition rate during the years that worldwide deposition rates have been measured, beginning in 1955<sup>(3)</sup>.

Table 2 lists calculated burdens of  $^{90}\text{Sr}$  in the stratosphere of the Northern Hemisphere during mid-October 1974, mid-April, and July-August 1975 as reported by Leifer et al <sup>(9)</sup> and for May-June 1976 and August-September 1976 (R. Leifer, personal communication). (It is noteworthy that the stratospheric burden of  $^{90}\text{Sr}$  immediately before the November 17, 1976 test was at its lowest point since at least 1954.) The apparent loss of  $^{90}\text{Sr}$  from the stratosphere during each interval between the times appropriate to these calculated burdens is listed in Table 2. For comparison, the calculated deposition of  $^{90}\text{Sr}$  on the surface of the earth in the Northern Hemisphere during each of these intervals is also listed. The total estimated deposition in the Northern Hemisphere between mid-October 1974 and August-September 1976 is 86 kCi, 18% higher than the calculated loss of  $^{90}\text{Sr}$  from the stratosphere of the Northern Hemisphere during this period. This discrepancy is more likely attributable to uncertainties

in estimating both stratospheric burdens and rates of deposition on the surface than it is to actual interhemispheric transport of  $^{90}\text{Sr}$ .

We will not attempt to predict the rate of deposition of  $^{90}\text{Sr}$  during 1977 because we have no estimate as yet of the amount of  $^{90}\text{Sr}$  injected into the stratosphere by the November 17, 1976 Chinese test, or of the distribution of that  $^{90}\text{Sr}$  within the stratosphere. Our prediction<sup>(8)</sup> that total  $^{90}\text{Sr}$  deposition worldwide during 1976 would probably be between 12 and 24 kCi was made with seemingly adequate stratospheric data to rely upon, and it was rather badly in error. Nevertheless we will point out that the concentrations of  $^{137}\text{Cs}$  in surface air in the Northern Hemisphere during the first five months of 1977 were roughly comparable to those measured in the same region during the first five months of 1973, a year in which the worldwide deposition of  $^{90}\text{Sr}$  amounted to only 63 kilocuries.

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Table 1

Comparison of Estimates of Annual Deposition of  $^{90}\text{Sr}$   
Made by the Environmental Measurements Laboratory and  
The United Kingdom Atomic Energy Authority<sup>(4)</sup>.

<u>Year</u>	<u>Annual Deposition</u> <u>(Megacuries <math>^{90}\text{Sr}</math>)</u>		<u>Cumulative Deposition</u> <u>(Megacuries <math>^{90}\text{Sr}</math>)</u>	
	<u>HASL</u>	<u>UKAEA</u>	<u>HASL</u>	<u>UKAEA</u>
1970	0.34	0.21	12.19	12.37
1971	0.39	0.26	12.23	12.33
1972	0.18	0.17	12.10	12.18
1973	0.06	0.12	11.87	12.00
1974	0.16	0.16	11.74	11.87
1975	0.09	0.14	11.54	11.72
1976	0.05	-	11.49	-



Table 2

A Comparison of Estimated Changes in Stratospheric Burdens of  $^{90}\text{Sr}$  With  
Estimated Deposition on the Surface of the Earth in the Northern Hemisphere

<u>Time</u>	<u>Estimated <math>^{90}\text{Sr}</math> Burden of the Stratosphere of the Northern Hemisphere (kilocuries)</u>	<u>Apparent Loss of <math>^{90}\text{Sr}</math> from the Stratosphere (kilocuries)</u>	<u>Estimated Deposition of <math>^{90}\text{Sr}</math> on the Surface (kilocuries)</u>
Mid-October 1974	87	41	37
Mid-April 1975	46	18	25
July-August 1975	28	10	19
May-June 1976	18	4	5
August-September 1976	14	<hr/>	<hr/>
		73	86

Table 3

AVERAGE SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE NORTHERN HEMISPHERE DURING 1976 (mCi/km<sup>2</sup>)

QUARTER	0-10 (NO)	10-20 (NO)	20-30 (NO)	30-40 (NO)	40-50 (NO)	50-60 (NO)	60-70 (NO)	70-80 (NO)	80-90 (NO)	QTR. AVG.
1 ST	0.01( 8)	0.01( 7)	0.03( 6)	0.04( 8)	0.03( 8)	0.09( 1)	0.02( 3)	0.01( E)	0.00( E)	0.03
2 ND	0.02( 8)	0.01( 6)	0.01( 6)	0.04( 7)	0.05( 8)	0.01( 2)	0.02( 4)	0.01( E)	0.00( E)	0.02
3 RD	0.01( 6)	0.01( 6)	0.02( 5)	0.02( 7)	0.03( 8)	0.04( 2)	0.01( 2)	0.01( E)	0.00( E)	0.02
4 TH	0.03( 7)	0.02( 6)	0.09( 1)	0.04( 3)	0.04(10)	0.06( 2)	0.02( 3)	0.01( E)	0.00( E)	0.03

Table 3 (cont'd)

AVERAGE SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE SOUTHERN HEMISPHERE DURING 1976 (mCi/km<sup>2</sup>)

QUARTER	0-10 (NO)	10-20 (NO)	20-30 (NO)	30-40 (NO)	40-50 (NO)	50-60 (NO)	60-70 (NO)	70-80 (NO)	80-90 (NO)	QTR. AVG.
1 ST	0.01( 4)	0.02( 7)	0.03( 4)	0.04( 6)	0.04( 1)	0.0 ( 1)	0.0 ( E)	0.0 ( E)	0.0 ( E)	0.01
2 ND	0.01( 4)	0.01( 7)	0.00( 4)	0.03( 6)	0.02( 1)	0.0 ( 1)	0.0 ( E)	0.0 ( E)	0.0 ( E)	0.01
3 RD	0.02( 1)	0.00( 6)	0.02( 5)	0.03( 6)	0.02( 1)	0.01( 1)	0.01( E)	0.01( E)	0.00( E)	0.01
4 TH	--- ( 0)	0.02( 6)	0.05( 1)	0.03( 5)	0.04( 1)	0.03( E)	0.02( E)	0.01( E)	0.00( E)	0.02

Table 4

TOTAL SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE NORTHERN HEMISPHERE DURING 1976 (KCI)

QUARTER	0-10 (NO)	10-20 (NO)	20-30 (NO)	30-40 (NO)	40-50 (NO)	50-60 (NO)	60-70 (NO)	70-80 (NO)	80-90 (NO)	QTR. TOT.	CUM. TOT.
1 ST	0.45( 8)	0.29( 7)	1.04( 6)	1.51( 8)	0.91( 8)	2.40( 1)	0.37( 3)	0.14( E)	0.02( E)	7.11	8718.76
2 ND	1.10( 8)	0.42( 6)	0.57( 6)	1.60( 7)	1.70( 8)	0.30( 2)	0.33( 4)	0.12( E)	0.01( E)	6.15	8706.94
3 RD	0.60( 6)	0.40( 6)	0.83( 5)	0.55( 7)	0.79( 8)	0.90( 2)	0.22( 2)	0.08( E)	0.01( E)	4.39	8693.38
4 TH	1.18( 7)	0.82( 6)	3.76( 1)	1.54( 3)	1.17(10)	1.50( 2)	0.37( 3)	0.14( E)	0.02( E)	10.49	8685.95
YEAR TOTAL	3.33	1.93	6.20	5.20	4.57	5.09	1.29	0.47	0.05	28.14	

Table 4 (cont'd)

TOTAL SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE SOUTHERN HEMISPHERE DURING 1976 (KCI)

QUARTER	0-10 (NO)	10-20 (NO)	20-30 (NO)	30-40 (NO)	40-50 (NO)	50-60 (NO)	60-70 (NO)	70-80 (NO)	80-90 (NO)	QTR. TOT.	CUM. TOT.
1 ST	0.37( 4)	0.68( 7)	1.06( 4)	1.63( 6)	1.11( 1)	0.0 ( 1)	0.0 ( E)	0.0 ( E)	0.0 ( E)	4.85	2811.90
2 ND	0.52( 4)	0.43( 7)	0.12( 4)	0.99( 6)	0.74( 1)	0.0 ( 1)	0.0 ( E)	0.0 ( E)	0.0 ( E)	2.79	2808.90
3 RD	1.03( 1)	0.15( 6)	0.68( 5)	1.14( 6)	0.74( 1)	0.30( 1)	0.16( E)	0.06( E)	0.01( E)	4.27	2807.38
4 TH	--- ( 0)	0.82( 6)	1.88( 1)	0.94( 5)	1.11( 1)	0.70( E)	0.37( E)	0.14( E)	0.02( E)	5.96	2807.56
YEAR TOTAL	1.92	2.09	3.74	4.70	3.69	1.00	0.53	0.19	0.02	17.87	

1976

Table 5

CUMULATIVE SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE NORTHERN HEMISPHERE DURING 1976 (KCI)

QUARTER	0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
1 ST	657.12	922.05	1320.22	1633.35	1925.54	1457.90	618.72	159.08	25.07
2 ND	656.86	920.57	1318.07	1631.58	1923.28	1455.20	617.77	158.87	25.03
3 RD	656.11	919.07	1316.18	1628.76	1920.11	1453.10	616.72	158.63	24.99
4 TH	655.94	918.00	1317.23	1626.95	1917.32	1451.60	615.82	158.44	24.96

Table 5 (cont'd)

CUMULATIVE SR90 DEPOSITS IN 10 DEGREE LATITUDE BANDS IN THE SOUTHERN HEMISPHERE DURING 1976 (KCI)

QUARTER	0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
1 ST	354.01	323.65	538.07	558.46	564.45	274.93	137.54	52.24	8.41
2 ND	353.79	323.41	537.08	558.30	564.03	274.37	137.26	52.13	8.39
3 RD	354.09	322.90	536.66	558.29	563.60	274.10	137.13	52.08	8.38
4 TH	353.36	323.05	537.43	558.07	563.54	274.23	137.22	52.11	8.38

TABLE 6

ANNUAL-CUMULATIVE WORLDWIDE  $^{90}\text{Sr}$  DEPOSITION (MEGACURIES)

	Annual Deposition			Cumulative Deposition		
	NORTHERN HEMISPHERE	SOUTHERN HEMISPHERE	TOTAL	NORTHERN HEMISPHERE	SOUTHERN HEMISPHERE	TOTAL
Pre-1958				1.7	0.6	2.3
1958	0.630	0.255	0.885	2.28	0.84	3.12
1959	1.052	0.185	1.237	3.26	1.00	4.26
1960	0.262	0.168	0.430	3.44	1.14	4.58
1961	0.351	0.174	0.525	3.70	1.29	4.99
1962	1.444	0.264	1.708	5.04	1.51	6.55
1963	2.622	0.308	2.930	7.51	1.78	9.29
1964	1.656	0.422	2.078	8.96	2.16	11.12
1965	0.774	0.357	1.131	9.50	2.46	11.96
1966	0.328	0.207	0.535	9.59	2.60	12.19
1967	0.169	0.110	0.279	9.52	2.65	12.17
1968	0.195	0.102	0.297	9.48	2.68	12.16
1969	0.147	0.141	0.288	9.40	2.76	12.16
1970	0.206	0.128	0.344	9.37	2.82	12.19
1971	0.188	0.150	0.388	9.33	2.90	12.23
1972	0.086	0.096	0.182	9.18	2.92	12.10
1973	0.032	0.031	0.063	8.99	2.88	11.87
1974	0.121	0.039	0.160	8.89	2.85	11.74
1975	0.058	0.034	0.092	8.73	2.81	11.54
1976	0.028	0.018	0.046	8.69	2.81	11.49



PART II  
HASL FALLOUT PROGRAM DATA

## 1. Fallout Deposition

### 1.1 Sampling Over Land

#### 1.11 $^{90}\text{Sr}$ in Quarterly Deposition

Monthly samples of total deposition (both wet and dry) are collected at 71 worldwide stations. These samples are composited on a quarterly basis and analyzed for  $^{90}\text{Sr}$ . A description of the sampling network and available data for each site are presented in the Appendix, Section A.

#### 1.12 $^{90}\text{Sr}$ and $^{89}\text{Sr}$ in Monthly Deposition

Precipitation and dry fallout were collected over monthly periods at stations in the United States and overseas. The samples were analyzed for  $^{90}\text{Sr}$  and prior to 1971, for  $^{89}\text{Sr}$  whenever possible. These monthly data are presented in final form in USERDA Report HASL-329, October 1, 1977.

#### 1.13 Other Isotopes at Selected Sites

At a number of stations in the United States, monthly deposition collections were analyzed for radiostrontium and other nuclides of interest. Multinuclide analyses were discontinued as of July 1967 and the complete data reported in HASL-193, pp. II-4 through II-25.

Plutonium analyses of monthly deposition were carried out at New York City, Melbourne, Seattle, Honolulu, Salisbury, Durban, and Rio de Janeiro. The data associated with this program were given in HASL-237, pp. II-4 through II-13.

### 1.2 Sampling Over Ocean

#### 1.21 $^{90}\text{Sr}$ and $^{89}\text{Sr}$ Deposition at Atlantic Ocean Weather Stations

Measurements of radiostrontium in precipitation and dry fallout collections at four U. S. Coast Guard Stations in the North Atlantic Ocean were carried out for comparison with land stations in the same latitude band. This program was terminated in the Spring of 1972. A description of the stations and available data are given on pp. II-4 through II-17 of HASL-274.

## 2. Surface Air Sampling Program

The Health and Safety Laboratory has been collecting surface air particulate samples at stations in the Western Hemisphere since January 1963. The filters are analyzed for a number of fission and activation product radionuclides as well as stable lead. A description of the program and available data are given in the Appendix, Section B.

3. Project Airstream

The Health and Safety Laboratory measures radioactivity in the lower stratosphere employing the WB-57F aircraft as a sampling platform. The aircraft are now flown by the National Aeronautics and Space Administration. The missions are scheduled three times a year and the coverage extends from 75°N to the equator in the Western Hemisphere. Air filter samples are collected from 12 to 20 km altitude and analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-7 to II-63 of HASL-321, July 1, 1977.

4. High Altitude Balloon Sampling Program

Balloon borne filtering devices are used to collect nuclear debris at altitudes from 20 - 27 km. Balloon launchings are conducted annually at Fairbanks, Alaska, 65°N and the Panama Canal Zone (9°N), and three times a year at Alamogordo, N.M. (33°N). Filters are analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-9 to II-26 of HASL-306 (July 1976).

5. Radiostrontium in Milk and Tap Water

Strontium-90 levels in fresh milk distributed in New York City and tap water sampled at the Health and Safety Laboratory, have been measured on a monthly basis since 1954. Cesium-137 has been analyzed in tap water since 1965. These data are summarized in tabular and graphical form in the Appendix, Section C, of this report.

6. Strontium-90 in Diets

Quarterly estimates of the annual dietary intake of Sr-90 for New York City and San Francisco residents have been made based on analyses of foods purchased at these cities every three months since 1960. Sampling in Chicago was discontinued in 1967. The program is described and the most recent data reported on pages II-4 to II-6 of this report. The data through 1976 are evaluated beginning on p. I-39 of HASL-321, July 1, 1977.

7. Strontium-90 in Human Bone

Specimens of human vertebrae from New York City and San Francisco have been made available for Sr-90 analysis since 1961. The results for 1976 are evaluated beginning on page I-69 of this report.

Human vertebral specimens are also received, through the World Health Organization, from countries where western-type diets are not typical. The Sr-90 data for samples received in 1975 are reported on pages II-7 and II-8 of HASL-306 (July 1976.)

## 6. HASL Diet Studies: 1st Quarter 1977

Results of the measurements of the  $^{90}\text{Sr}$  content of foods purchased in New York City and San Francisco during the first quarter of 1977 are given in the following table. Also listed are estimates of the total diet intake of  $^{90}\text{Sr}$  based on these measurements and on recent consumption statistics compiled by the U. S. Department of Agriculture.<sup>1</sup>

The estimates of dietary intake of  $^{90}\text{Sr}$  are a continuation of the HASL Tri-City diet studies which were started in March of 1960. Results of the earlier measurements along with those made during the first quarter of 1977 are shown graphically in the figure on page II-6. More detailed discussion of the results for the entire sampling program through 1976 is presented in HASL-321.<sup>2</sup> A description of the sampling methods and philosophy of the HASL diet studies is given in HASL-147.<sup>3</sup>

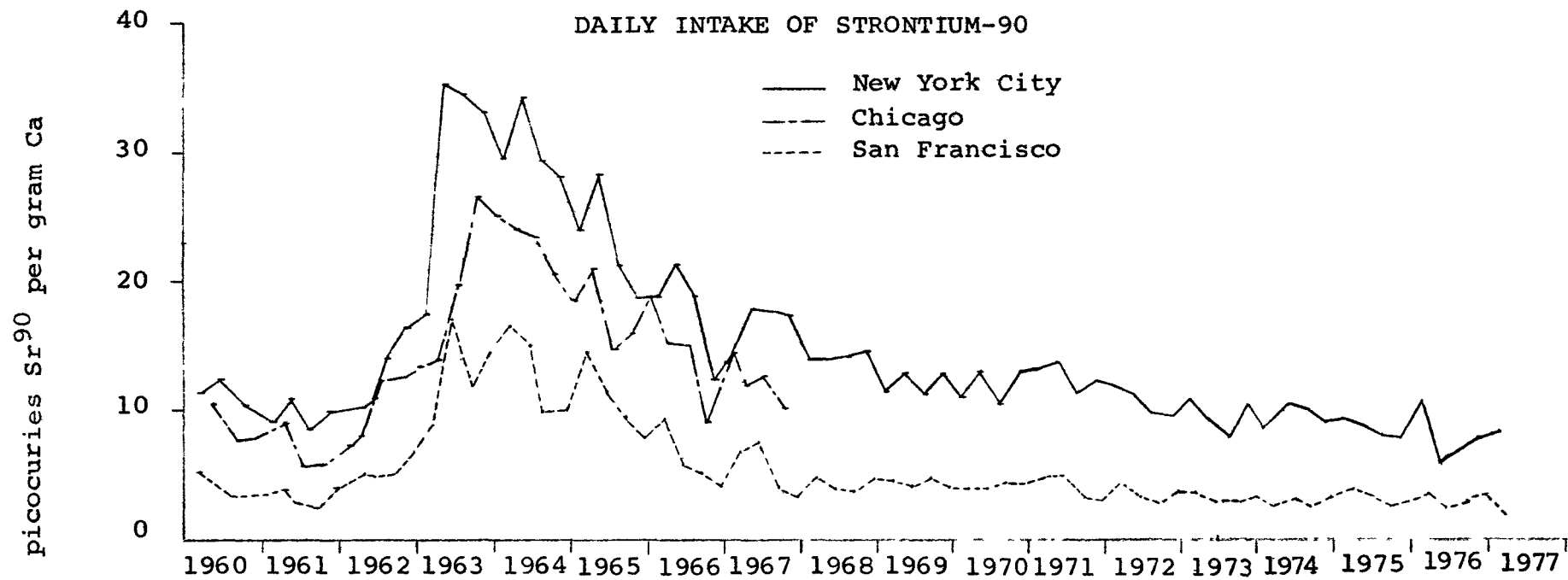
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HASL Contributions to the Study of Fallout in Food Chains  
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STRONTIUM-90 IN NEW YORK CITY AND SAN FRANCISCO DIETS

- First Quarter 1977 -

Diet Category	kg/y	gCa/y	% of yearly intake of Ca	New York City - February			San Francisco - March		
				pCi <sup>90</sup> Sr/kg	pCi <sup>90</sup> Sr/y	% of yearly intake of <sup>90</sup> Sr	pCi <sup>90</sup> Sr/kg	pCi <sup>90</sup> Sr/y	% of yearly intake of <sup>90</sup> Sr
DAIRY PRODUCTS	200	216.0	58	3.6	713.	22	0.9	182	20
FRESH VEGETABLES	48	18.7		7.4	354		2.1	101	
CANNED VEGETABLES	22	4.4		4.1	89		3.9	86	
ROOT VEGETABLES	10	3.8		4.9	49		4.9	49	
POTATOES	38	3.8		5.5	209		1.6	60	
DRY BEANS	3	2.1	9	3.1	9	22	7.6	23	35
FRESH FRUIT	59	9.4		22.2	1311		1.8	103	
CANNED FRUIT	11	.6		1.0	10		0.9	9	
FRUIT JUICES	28	2.5	3	2.4	66	42	1.6	45	17
BAKERY PRODUCTS	44	53.7		4.2	184		2.4	106	
FLOUR	34	6.5		3.7	125		1.8	62	
WHOLE GRAIN PRODUCTS	11	10.3		7.3	80		4.2	46	
MACARONI	3	0.6		1.4	4		2.0	6	
RICE	3	1.1	20	0.8	2	12	1.0	3	24
MEAT	79	12.6		0.4	34		0.2	12	
POULTRY	20	6.0		0.5	9		0.4	9	
EGGS	15	8.7		0.9	13		0.6	9	
FRESH FISH	8	7.6		0.3	3		0.2	2	
SHELL FISH	1	1.6	10	0.8	1	2	0.2	<1	4
Yearly Intake		370g			3265 pCi			913 pCi	
Daily Intake					8.8 pCi/g Ca			2.5 pCi/g Ca	



### PART III

#### DATA FROM SOURCES OTHER THAN HASL

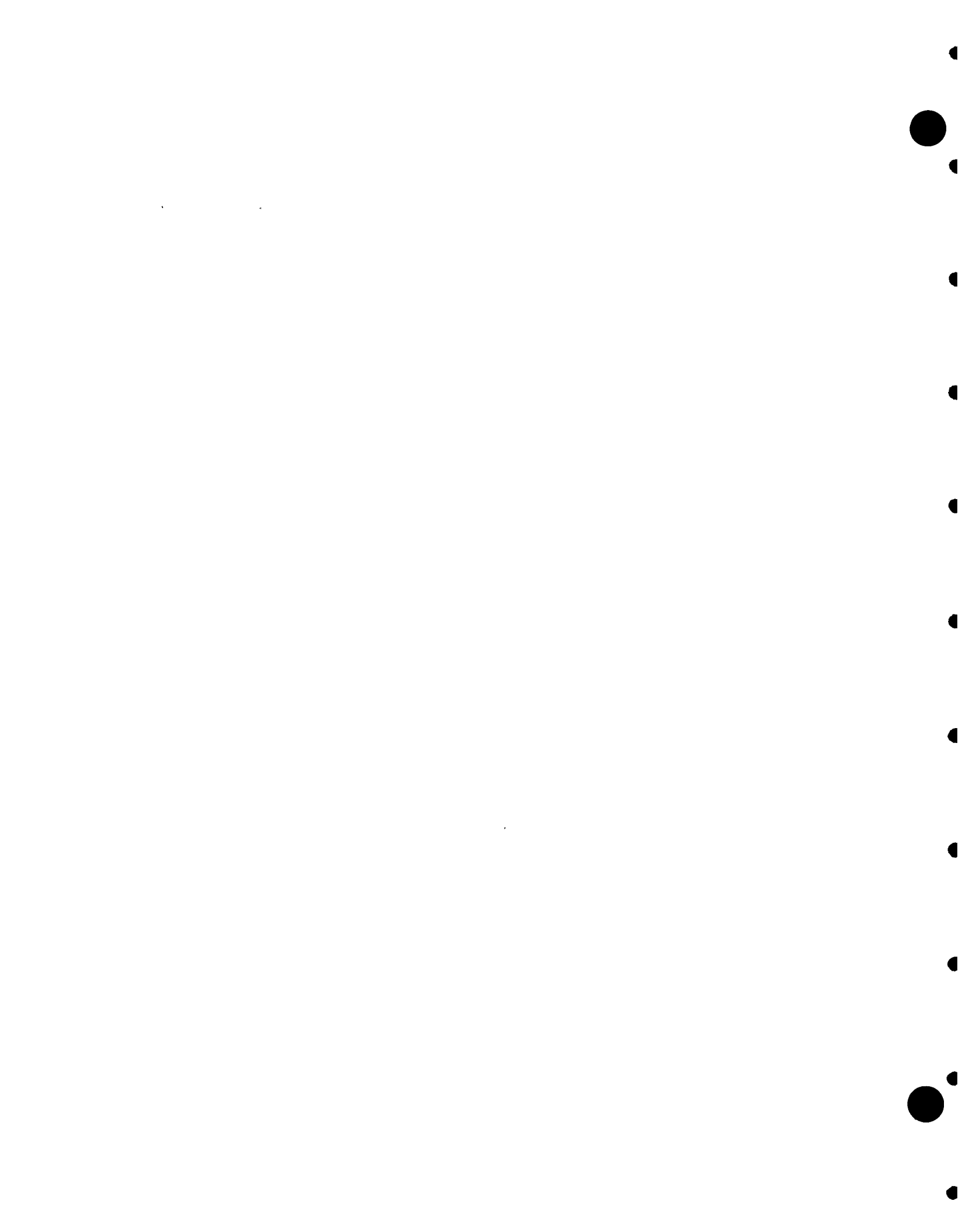
Numerous environmental studies are conducted by other organizations in the United States and abroad. Some of these are sent to the editor for dissemination in these HASL Environmental Quarterlies. Submitted data are reproduced essentially as received and no interpretation by HASL is attempted.

Air Monitoring Section  
Bhabha Atomic Research Centre  
Bombay, India  
Fallout and Atmospheric Radioactivity Measurements  
in India  
by Mrs. S. Gopalakrishnan, C. D. Eapen and  
C. Rangarajan

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National Radiation Laboratory, Department of  
Health  
Christchurch, New Zealand  
Environmental Radioactivity: Annual Report 1976  
NRL- F/56

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# FALLOUT AND ATMOSPHERIC RADIOACTIVITY MEASUREMENTS IN INDIA

BY

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In this article we give a summary of the atmospheric radioactivity measurements carried-out in India for the past several years. The radioactivities measured include those from natural sources (eg: radon-thoron daughter products) and from nuclear tests.

Figure-1 shows the locations of the stations in India where these measurements were carried-out. A few of the stations like Bombay, Bangalore, Delhi, Calcutta, etc., have been in operation since 1956 while the others were added through the course of the years. However, all the stations were not operative for all the time as some of the older stations were closed down when new stations were started, depending on operational and other conveniences. In general, about six stations, covering the whole country and the climatic patterns prevailing, have been in operation at any time. Although rainwater from the above stations were also being processed in the earlier years (1,2,3), the programme is confined to surface level atmospheric samples only, after 1965 (4). This is due to rainfall being confined to a few months of the year at several stations and the difficulty in processing large volumes of water which is necessary when levels of activity are low. Figure-2 summarises the rainfall pattern at some of the sampling stations in India.

## 2. SAMPLING TECHNIQUES

During the early years, air sampling was carried-out using blowers drawing air through Hollingsworth and Vose H-70 type filter discs of 5 cms effective diameter at flowrates of 50 - 100 liters per minute. This system was adequate during the period 1956 - 1963 for the direct counting of the gross beta activity of the filters with end-window counters but the activities collected were inadequate for detailed isotopic analysis. Hence powerful Roots blowers or centrifugal fans drawing air at flow rates of 1200 liters per minute were put into operation, wherever possible. In view of the large flow rates of these blowers, rectangular filters of dimensions 30 cms x 15 cms were used for air dust collection. At Bombay it was possible to operate three such units in parallel to increase the sensitivity of analysis. After collection these filters are either ashed and transferred into planchets of suitable sizes or compressed into circular discs in a heavy duty press prior to counting. Samples are analysed either weekly, two weekly, or monthly, depending on the volume sampled. Finally, the sample discs are subjected to radio-chemical analysis for radio-nuclides of interest.

The original system with the smaller blower and 5 cms. filter discs was however retained at a few selected stations for direct alpha measurement and calculation of atmospheric concentrations of radon-thoron daughters.

## MEASUREMENTS AND ANALYSES

The analysis for fission products is done using NaI(Tl) and recently Ge(Li) detectors and multichannel analysers (1-4). Computer programme are used for the unscrambling of the gamma spectra (5). Radiochemical separation

is carried-out only for the measurement of RaD which is counted using low level beta counters either by the ingrowth of Bi-210 in the separated lead (6,7), or by separation of bismuth itself. In both cases the purity can be checked by following the ingrowth or decay respectively of the sample activity. Radon and thoron daughters' estimation is by direct alpha counting of the disc filters in ZnS Scintillation counter (8). Radioactive equilibrium is assumed in most of the analyses and results are expressed as RaB (Pb-214) and ThB (Pb-212) concentrations respectively. However in certain cases the decay of the sample is followed and a least squares resolution of the individual radio nuclides RaA, RaB and RaC is carried-out.

## RESULTS

Table-1 gives the levels of the measured fission products in the surface air at Bombay, from 1971 to 1975. The results from Bombay and other stations for earlier years have already been reported (1-4). In view of the decreasing concentrations of fission products, the levels of certain isotopes like Sb-125 are not given for the year 1975. Figure-3 gives the levels of Cs<sup>137</sup> at Bombay, while figure 4,5 and 6 show the variations of the fission product ratios through the years. These ratios are useful in estimating the contribution of different test series to air activity. Figure-7 gives the gross beta activity at the different stations during the period 1956-64. These values can be compared with the levels from individual USSR, French and Chinese tests of the period 1957-68 (Figures 8 to 15). The figures also give the time taken by the radioactive debris to reach the sampling stations from the testing sites. This is of interest in a study of the transport and diffusion of radioactive debris on a planetary scale (10,11,12,13). A

complete summary of the travel times from the various test sites to stations in India is given in Table-2.  $Ba^{140}/Zr^{95}$  ratios from some of the French and Chinese tests at Bombay and other stations are shown in Figures 16-18. The increase in these ratios is a sensitive indicator of the arrival of activity from recent tests (14).

The levels and variations of the natural activities due to RaB and ThB at Bombay and Poona are given in figure-19 and Table-3. Table-4 and Figure-20 give the values of Pb-210 in surface air and rainwater at the various stations in India. The meteorological causes for the variation of the natural activity in surface air and their interrelations have been discussed in the publications listed below.

#### ACKNOWLEDGEMENTS

The authors are thankful to Dr. K.G. Vohra, Head, Division of Radiological Protection and Dr. U.C. Mishra, Head, Air Monitoring Section, for their encouragement and interest in the work.

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TABLE - 1A

CONCENTRATIONS OF CE-144,SB-125,RU-106,CS-137,ZR-95 AND BA-140  
IN SURFACE AIR AT BOMBAY

(PICOCURIES PER 1000 CUBIC METERS)

MONTH	CE-144	SB-125	RU-106	CS-137	ZR-95	BA-140
1971						
JANUARY	49	3.4	22	2.5	17	
FEBRUARY	97	12	63	4.3	46	
MARCH	175	32	48		90	
APRIL	142	16	75	6.9	105	
MAY	73	12	48	6.0	194	
JUNE	72	8.3	43	5.3	103	115
JULY	84	3.6	28	3.9	236	143
AUGUST	16	4.5	21	-	116	24
SEPTEMBER	24	5.5	8.2	-	38	
OCTOBER	53	8.8	-	-	16	
NOVEMBER	20	8.7	5.5	1.1	8.6	
DECEMBER	28	5.3	14	3.7	6.0	
AVERAGE	69	10	31	2.8	81	
1972						
JANUARY	38	7.1	10	1.9	8.7	
FEBRUARY	35	2.9	20	3.0	5.1	
MARCH	24	3.0	12	3.4	2.9	
APRIL	39	3.3	18	2.2	97	
MAY	27	2.3	12	2.1	35	
JUNE	21	1.9	9.5	2.5	16	
JULY	17	2.0	15	1.5	2.9	
AUGUST	59	2.3	16	1.8	3.3	
SEPTEMBER	14	1.4	8.5	2.2	3.3	
OCTOBER	58	-	8.0	2.1	1.9	
NOVEMBER	38	2.7	9.1	1.9	1.4	
DECEMBER	49	3.3	21	3.0	2.2	
AVERAGE	35	2.7	13	2.3	15	
1973						
JANUARY	4.8	0.6	2.7	0.9	0.3	
FEBRUARY	2.7	0.7	2.6	0.7	0.3	
MARCH	9.9	1.2	6.7	1.9	0.4	
APRIL	7.8	1.0	5.9	2.5	0.2	
MAY	1.3	1.8	12	1.6	1.0	
JUNE	6.9	0.3	2.0	3.6	0.2	
JULY	-	-				
AUGUST	-	-	*	*		
SEPTEMBER	2.0	0.4	2.1	0.4	2.5	
OCTOBER	5.2	1.3	4.1	1.0	3.3	
NOVEMBER	11	-	6.1	-	7.9	
DECEMBER	22	1.2	8.1	0.8	9.5	
AVERAGE	7.4	0.9	5.2	1.3	2.1	

TABLE - 1A(CONTD.....)

CONCENTRATIONS OF CE-144,SB-125,RU-106,CS-137,ZR-95 AND BA-140  
IN SURFACE AIR AT BOMBAY

(PICOCURIES PER 1000 CUBIC METERS)

MONTH	CE-144	SB-125	RU-106	CS-137	ZR-95	BA-140
1974						
JANUARY	31	2.6	28	2.9	21	
FEBRUARY	58	3.6	58	5.3	37	
MARCH	80	5.7	64	9.6	41	
APRIL	71	6.5	52	6.2	31	
MAY	59	5.9	44	7.3	23	
JUNE	32	3.5	24	4.1	7.3	
JULY	40	3.3	24	3.4	24	175
AUGUST	40	2.2	21	2.8	111	120
SEPTEMBER	23	1.2	10	1.6	61	
OCTOBER	16	1.5	9.4	2.3	8.6	
NOVEMBER	25	2.0	34	3.0	26	
DECFMBER	38	1.5	22	3.2	21	
AVERAGE	43	3.3	33	4.3	32	148
1975						
JANUARY			28	3.9		
FEBRUARY	29		20	3.8		
MARCH	52		37	6.9		
APRIL	50		36	7.6		
MAY	12		19	7.7		
JUNE	9.0		11	5.6		
JULY	2.5		4.6	3.2		
AUGUST	2.7		4.6	2.8		
SEPTEMBER	1.2		3.2	2.6		
OCTOBER	1.3		3.3	2.8		
NOVEMBER	1.6		4.5	1.8		
DECEMBER	2.1		13	4.0		
AVERAGE	14		15	4.4		

TABLE-1B

SHORT-LIVED FISSION PRODUCTS FROM THE CHINESE NUCLEAR TEST OF 1976

(PICOCURIES PER 1000 CUBIC METERS)

BOMBAY			HYDERABAD			PORT BLAIR		
PERIOD	ZR-95	BA-140	PERIOD	ZR-95	BA-140	PERIOD	ZR-95	BA-140
8-15 OCT	94	135	9-16 OCT	266	271			
16-23 OCT	120	286	18-25 OCT	206	290	18-25 OCT	28	50
24-31 OCT	139	239	26-30 OCT	88	101	25-31 OCT	83	74
1-7 NOV	46	73						

TABLE - 2

## TRAVEL TIMES OF DEBRIS FROM TEST SITES TO MONITORING STATIONS IN INDIA

TEST SERIES	TEST SITE	TEST DATES	MONITORING STATIONS	DETECTION DATE	TRAVEL TIME (IN DAYS)*	METHOD OF DETECTION
1	2	3	4	5	6	7
1.USSR	SIBERIA 52°N 78°E	3- 4-57 TO 16-4-57	DELHI	17-4-57	14	GROSS BETA
			CALCUTTA	19-4-57	16	,, ,,
			NAGPUR	20-4-57	17	,, ,,
			BOMBAY	18-4-57	15	,, ,,
2.USSR	ARCTIC 75°N 78°E	30-9-58 TO 3-10-58	DELHI	26-10-58	26	GROSS BETA
			CALCUTTA	26-10-58	26	,, ,,
			BOMBAY	23-10-58	23	,, ,,
3.FRANCE	ALGERIA 27°N, 0°	13-2-60	SRINAGAR	22-2-60	9	GROSS BETA
			DELHI	26-2-60	13	,, ,,
			CALCUTTA	20-2-60	7	,, ,,
			NAGPUR	19-2-60	6	,, ,,
			BOMBAY	18-2-60	5	,, ,,
			BANGALORE	21-2-60	8	,, ,,
			OOTY	24-2-60	11	,, ,,
4.USSR	SIBERIA AND ARCTIC	1-9-61 TO 4-10-61	SRINAGAR	18-9-61	17	GROSS BETA
			DELHI	30-9-61	29	,, ,,
			CALCUTTA	8-10-61	37	,, ,,
			BOMBAY	27-9-61	26	,, ,,
			BANGALORE	26-9-61	25	,, ,,
			OOTY	26-9-61	25	,, ,,
5.CHINA	WESTERN CHINA 40°N 90°E	14-5-65	GULMARG	19-5-65	5	GROSS BETA
			SRINAGAR	31-5-65	17	,, ,,
			NAINITAL	1-6-65	18	,, ,,
			DELHI	4-6-65	21	,, ,,
6.CHINA	WESTERN CHINA	9-5-66	GULMARG	23-5-66	14	GROSS BETA
			SRINAGAR	23-5-66	14	,, ,,
			NAINITAL	23-5-66	14	,, ,,
			GANGTOK	28-5-66	19	,, ,,
			CALCUTTA	6-6-66	28	,, ,,
			BOMBAY	4-6-66	26	ZR-95
7.FRANCE	POLYN. 20°S 138°W	2-7-66 19-7-76	BOMBAY	3-8-66	15-32	ZR-95
8.FRANCE	POLYN.	11-9-66 24-9-66 4-10-66	BOMBAY	4-11-66	31-54	ZR-95



TABLE - 2 (CONTD.....)

1	2	3	4	5	6	7
9. CHINA	WESTERN CHINA	28-12-66	GULMARG NAINITAL BOMBAY	14-1-67 13-1-67 17-1-67	17 16 20	GROSS BETA ,, ,, ZR-95
10. FRANCE	POLYN.	5-6-67 27-6-67 2-7-67	BOMBAY OOTY THUMBA	22-7-67 22-7-67 21-7-67	20-47 20-47 19-46	ZR-95 GROSS BETA ,, ,,
11. CHINA	WESTERN CHINA	24-12-67	NAINITAL DELHI NAGPUR BOMBAY	8-1-68 10-1-68 10-1-68 9-1-68	15 17 17 16	GROSS BETA ,, ,, ,, ,, ZR-95 AND BA-140
12. FRANCE	POLYN.	7-7-68	NAINITAL BOMBAY  THUMBA	28-7-68 25-7-68  25-7-68	21 18  18	GROSS BETA ZR-95 AND BA-140 GROSS BETA
13. FRANCE	POLYN.	15-5-70	BOMBAY THUMBA	3-6-70 5-6-71 TO	19 21-27	BA-140 BA-140
14. FRANCE	POLYN.	12-6-71	BOMBAY  THUMBA	11-6-71 23-6-71 TO 30-6-70 25-6-71 TO 1-7-71	11-18   13-19	BA-140   BA-140
15. FRANCE OR CHINA		17-6-76	BOMBAY	15-7-74 TO 22-7-74	28-35	BA-140
16. CHINA	WESTERN CHINA	26-9-76	BOMBAY  HYDERABAD  PORT BLAIR	8-10-76 TO 15-10-76 9-10-76 TO 16-10-76 18-10-76 TO 25-10-76	12-19  13-20  22-29	BA-140  BA-140  BA-140

\* THE LIMITS OF TRAVEL TIME ARE GIVEN IN CASE ASSIGNMENT OF ACTIVITY TO A TEST IN A SERIES IS NOT POSSIBLE

TABLE - 3

## MONTHLY AVERAGE PB-214 (RA-B) ACTIVITIES IN SURFACE AIR AT BOMBAY

(PICOCURIES PER CUBIC METER)

		1966	1967	1968	1969	1970	1971	1972	1975	1976	1977	AVERAGE
JANUARY	M	78	132	161	139	123	163	92	77	161	95	122
	A	-	-	64	86	74	87	61	51	83	61	71
FEBRUARY	M	50	161	78	105	129	126	75	36	102	88	95
	A	-	-	66	68	66	59	42	22	59	57	55
MARCH	M	26	122	90	60	75	98	83	64	152	54	83
	A	-	-	47	29	51	54	51	40	122	40	54
APRIL	M	13	78	70	63	37	38	39	45	58		49
	A	-	-	27	27	21	14	22	29	42		26
MAY	M	6.5	48	61	18	22	20	24	19	29		28
	A	-	9.4	8.0	21	11	4.1	15	7.5	14		11
JUNE	M	7.5	73	23	20	29	8.7	16	16	24		22
	A	-	17	7.5	13	15	10	8.7	10	16		12
JULY	M	12	119	21	13	14	11	11	11	11		25
	A	-	14	16	11	11	10	9.0	8.6	9.4		11
AUGUST	M	17	63	16	15	12	14	13	14	8.6		19
	A	-	15	14	14	9.4	9.5	10	13	8.1		12
SEPT.	M	39	129	47	26	24	26	44	19	14		41
	A	-	10	20	12	13	18	19	15	8.1		14
OCTOBER	M	55	242	112	93	70	56	89	39	84		93
	A	-	50	73	59	40	26	59	28	56		49
NOVEMBER	M	59	135	150	99	138	109	-	113	93		112
	A	-	52	95	65	64	67	-	66	57		67
DECEMBER	M	149	210	139	151	145	93	-	160	146		140
	A	-	61	91	97	72	71	-	91	99		83
AVERAGE		43	87	62	54	53	50	39	41	60	66	54
	M	43	126	81	67	68	64	49	50	73	79	69
	A	-	29	44	42	37	36	30	32	48	53	39

TABLE - 3 (CONTD.....)

## MONTHLY AVERAGE PB-212 (TH-B) ACTIVITIES IN SURFACE AIR AT BOMBAY

(PICOCURIES PER CUBIC METER)

		1968	1969	1970	1971	1972	1975	1976	1977	AVERAGE
JANUARY	M	-	5.8	5.8	7.2	4.6	2.8	5.0	3.5	5.0
	A	-	5.4	4.5	5.3	3.2	2.9	3.4	2.8	3.9
FEBRUARY	M	-	4.9	5.7	7.0	6.2	1.5	3.7	3.7	4.7
	A	-	4.6	4.5	4.8	4.0	1.5	3.2	3.2	3.7
MARCH	M	-	3.5	4.2	6.6	9.7	3.0	9.7	2.9	5.7
	A	-	2.9	4.1	4.8	5.6	3.0	10	2.5	4.7
APRIL	M	-	4.5	3.5	4.6	4.7	2.8	5.9		4.3
	A	-	2.8	2.1	2.1	2.9	1.9	3.8		2.6
MAY	M	-	3.0	3.2	2.7	3.5	1.6	4.2		3.0
	A	-	1.8	1.8	0.93	1.6	0.74	1.5		1.4
JUNE	M	-	2.1	2.0	1.3	2.4	0.81	1.2		1.6
	A	-	1.6	1.4	0.99	1.1	0.49	0.67		1.0
JULY	M	-	0.76	0.99	1.4	2.0	0.55	0.25		0.99
	A	-	0.71	0.99	1.3	1.7	0.49	0.44		0.94
AUGUST	M	1.5	0.93	1.0	1.2	1.7	0.54	0.50		1.1
	A	1.5	0.93	0.73	1.1	1.3	0.57	0.45		0.94
SEPTEMBER	M	3.5	1.6	2.5	2.1	3.0	0.76	0.62		2.0
	A	2.1	1.1	1.6	1.5	1.5	0.54	0.43		1.3
OCTOBER	M	4.1	4.6	5.1	3.0	5.3	1.3	3.1		3.8
	A	4.8	3.9	3.4	1.8	2.9	0.81	2.3		2.8
NOVEMBER	M	5.8	5.2	5.3	5.2	-	3.1	3.4		4.9
	A	4.4	3.8	5.1	3.1	-	2.4	2.2		3.5
DECEMBER	M	5.3	6.1	6.8	5.0	-	4.9	5.0		5.5
	A	7.5	5.3	5.8	4.5	-	4.0	4.8		5.3
AVERAGE		4.1	3.2	3.4	3.3	3.4	1.8	3.2	3.1	3.1
	M	4.0	3.6	3.8	3.9	4.3	2.0	3.5	3.4	3.5
	A	4.1	2.9	3.0	2.7	2.6	1.6	2.8	2.8	2.7

TABLE - 3 (CONTD.....)

MONTHLY AVERAGE PB-214 (RA-B) ACTIVITIES IN SURFACE AIR AT POONA  
(PICOCURIES PER CUBIC METER)

		1974	1975	1976	*AVERAGE
APRIL	M		26		26
	A		5.9		5.9
MAY	M		22		22
	A		16		16
JUNE	M	10	12	9.7	11
	A	7.1	10	4.7	7.3
JULY	M	9.0	12	7.8	9.6
	A	8.3	7.7	6.3	7.4
AUGUST	M	6.9	9.2	6.3	7.5
	A	9.8	7.8	4.5	7.4
SEPTEMBER	M	17	14	13	15
	A	13	11	6.6	10
OCTOBER	M	34	27	55	39
	A	17	13	24	18
NOVEMBER	M		84		84
	A		21		21
DECEMBER	M		94		94
	A		25		25
AVERAGE *		13	23	13	24
	M	15	33	18	34
	A	11	13	9.0	13

TABLE - 3 (CONTD.....)

MONTHLY AVERAGE PB-212 (TH-B) ACTIVITIES IN SURFACE AIR AT POONA  
(PICOCURIES PER CUBIC METER)

		1974	1975	1976	*AVERAGE
APRIL	M		3.9		3.9
	A		0.4		0.4
MAY	M		3.0		3.0
	A		2.1		2.1
JUNE	M	1.1	1.0	1.1	1.1
	A	0.8	0.8	1.2	0.9
JULY	M	1.3	0.8	0.4	0.8
	A	1.5	0.7	0.6	0.9
AUGUST	M	1.2	0.5	0.4	0.7
	A	1.5	0.7	0.5	0.9
SEPTEMBER	M	1.7	1.0	1.0	1.2
	A	0.7	1.1	0.8	0.9
OCTOBER	M	2.1	2.3	4.3	2.9
	A	0.6	2.2	2.5	1.8
NOVEMBER	M		7.2		7.2
	A		4.5		4.5
DECEMBER	M		6.0		6.0
	A		3.8		3.8
AVERAGE *		1.3	2.3	1.3	2.4
	M	1.5	2.9	1.4	3.0
	A	1.0	1.8	1.1	1.8

\* THE AVERAGES ARE NOT THE ANNUAL MEANS AS THE DATA IS FOR PART OF THE YEAR ONLY

TABLE - 4 A

CONCENTRATIONS OF PB-210 IN SURFACE AIR AT DIFFERENT STATIONS IN INDIA  
(PICOCURIES PER 1000 CUBICMETERS)

STATION YEAR	SRINAGAR	DELHI	GANGTOK	CALCUTTA	NAGPUR	BOMBAY	BANGALORE	OOTACAMUND
1962	20	22	27	24	23	22	17	12
1963	30	25	39	30	30	28	19	15
1964	28	28	45	35	31	21	26	21
1965	24	29	26	22	24	17	16	20
1966	19	23	18	21	13	17	12	7
1967			23			11		
1968			16			13		
1969						25		
AVERAGE	24	25	28	26	24	19	18	15

TABLE - 4 B

YEARLY AVERAGE PB-210 ACTIVITY IN RAINWATER (PICOCURIES PER LITER) AND ANNUAL  
RAINFALL (MILLIMETER)

STATION	SRINAGAR	DELHI	GANGTOK	CALCUTTA	NAGPUR	BOMBAY	BANGALORE	OOTACAMUND
1962	8.0 (623)	4.5 (579)	1.2 (3173)	1.1 (1156)	1.4 (1259)	4.1 (2226)	1.0 (1054)	0.8 (1474)
1963	6.5 (785)	7.5 (827)	3.3 (3845)	1.8 (1678)	4.2 (933)	1.9 (2660)	2.0 (1042)	1.8 (1192)
1964	4.5 (694)	2.1 (1234)	1.2 (3277)	3.2 (1486)	2.6 (1051)	3.0 (2125)	2.2 (1193)	1.7 (1839)
1965	8.0 (678)	3.7 (592)	1.3 (3204)	6.9 (1604)	7.0 (1019)	3.4 (2626)	2.3 (692)	2.4 (871)
1966	7.3 (892)	5.1 (652)	3.1 (3510)	3.5 (1101)	2.2 (1046)	3.0 (1952)	2.6 (1207)	2.2 (1446)
AVERAGE	6.8 (734)	4.4 (777)	2.1 (3422)	3.4 (1405)	3.4 (1062)	3.0 (2318)	2.0 (1038)	1.7 (1364)

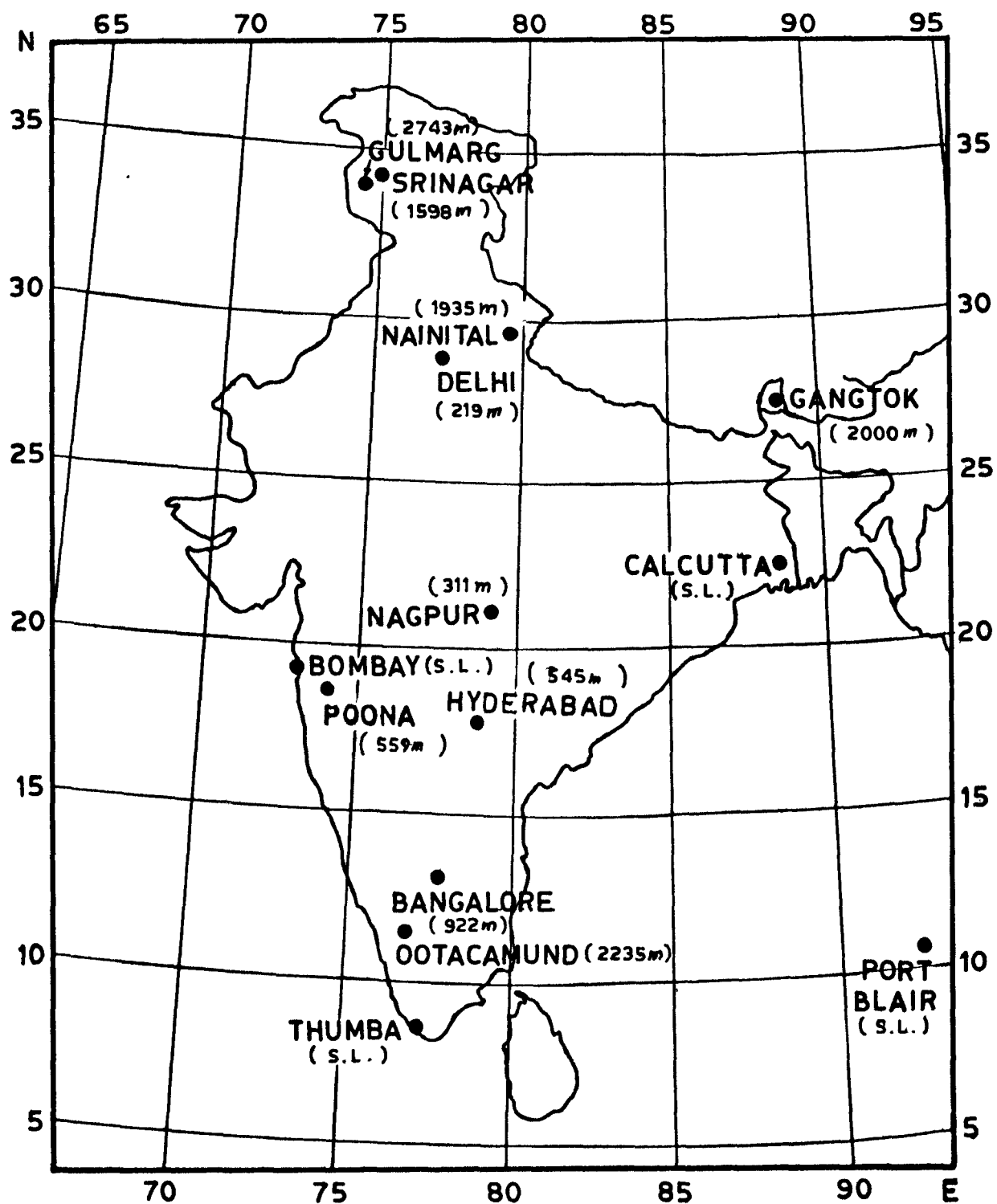


FIGURE:1. LOCATION OF FALLOUT SAMPLING STATIONS  
IN INDIA. (ALTITUDE OF STATIONS ARE GIVEN INSIDE  
BRACKETS) .

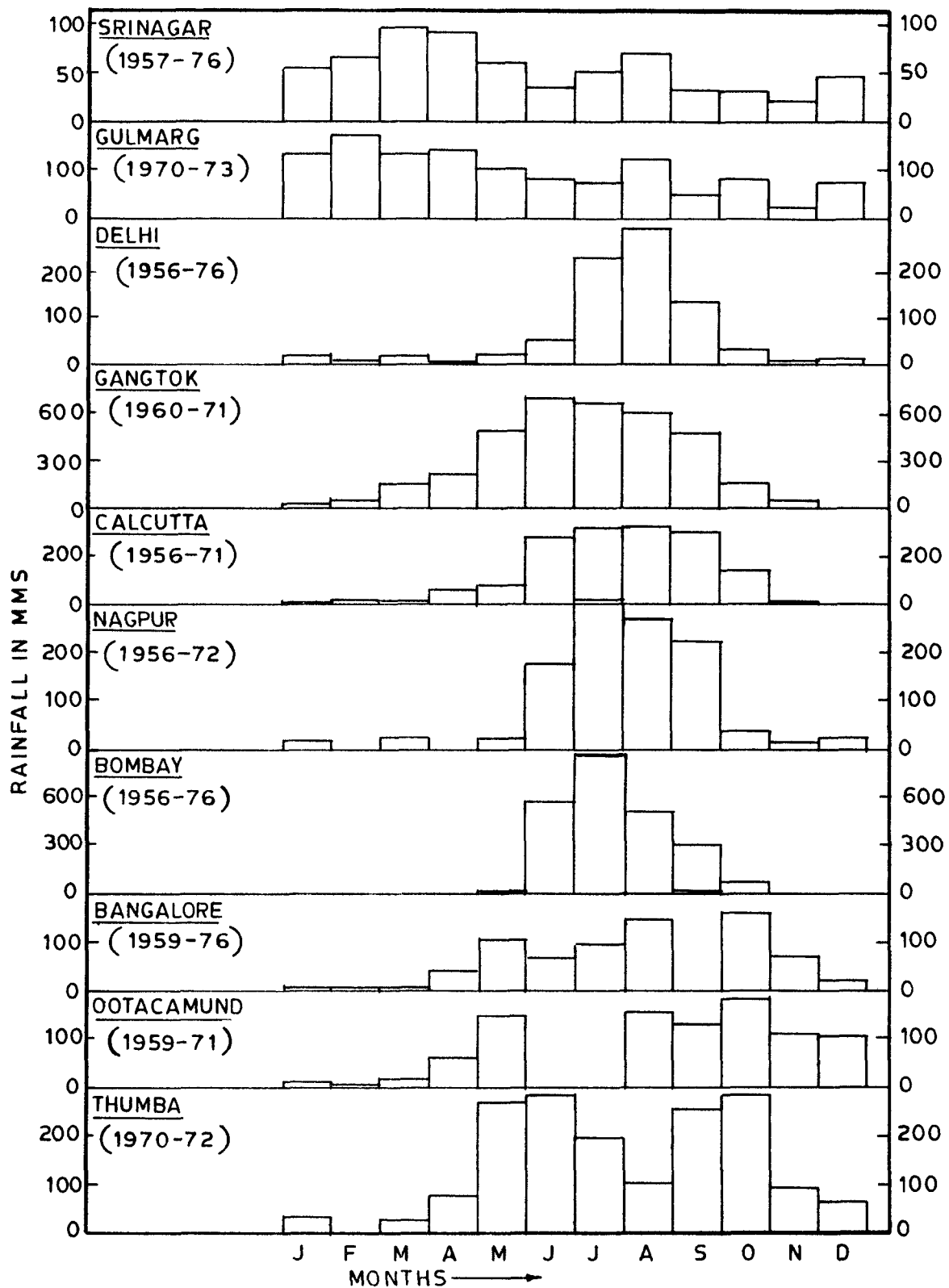


FIGURE: 2. AVERAGE MONTHLY RAINFALL AT THE SAMPLING STATIONS IN INDIA.

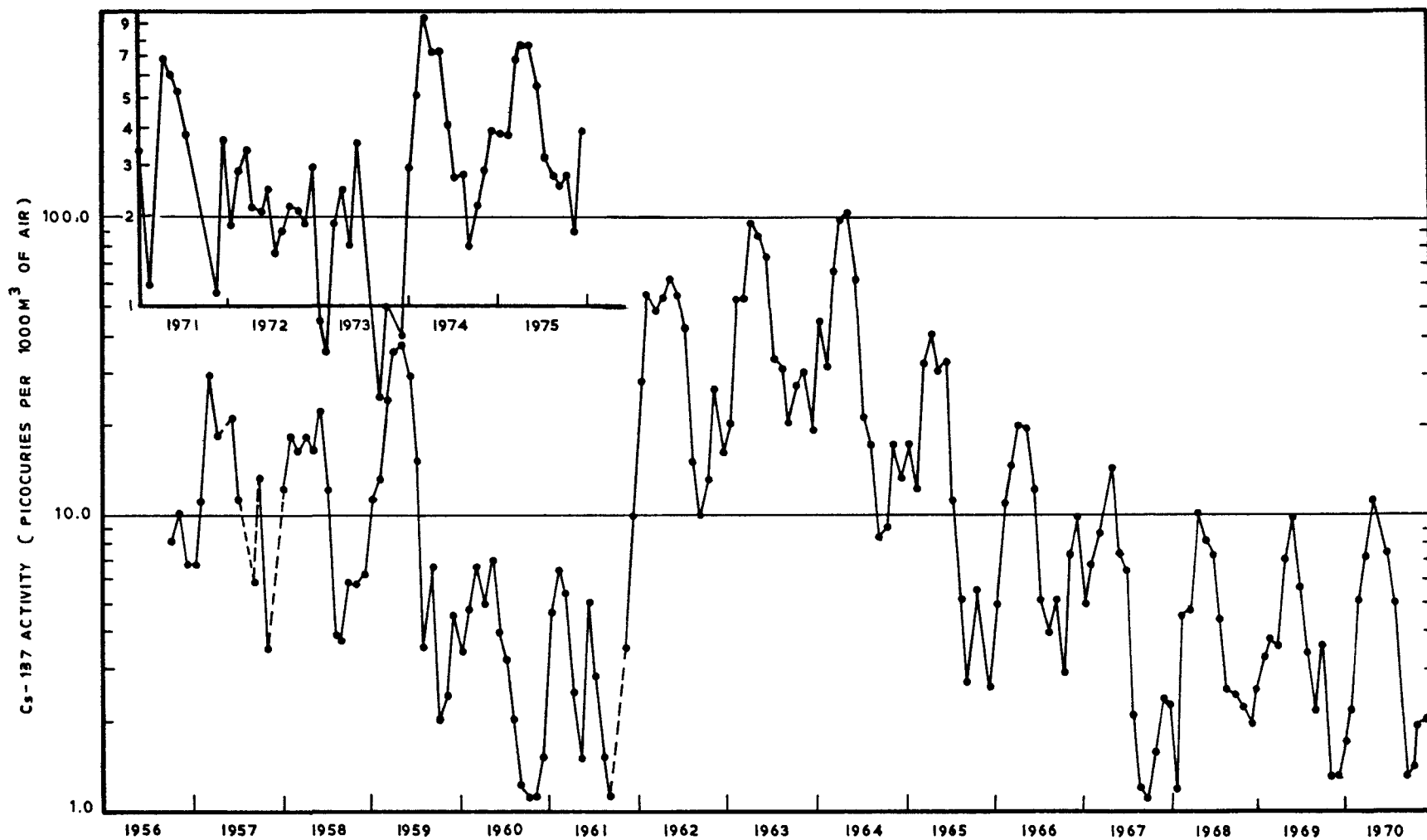


FIGURE 3. CONCENTRATIONS OF  $\text{Cs-137}$  IN GROUND LEVEL AIR AT BOMBAY.





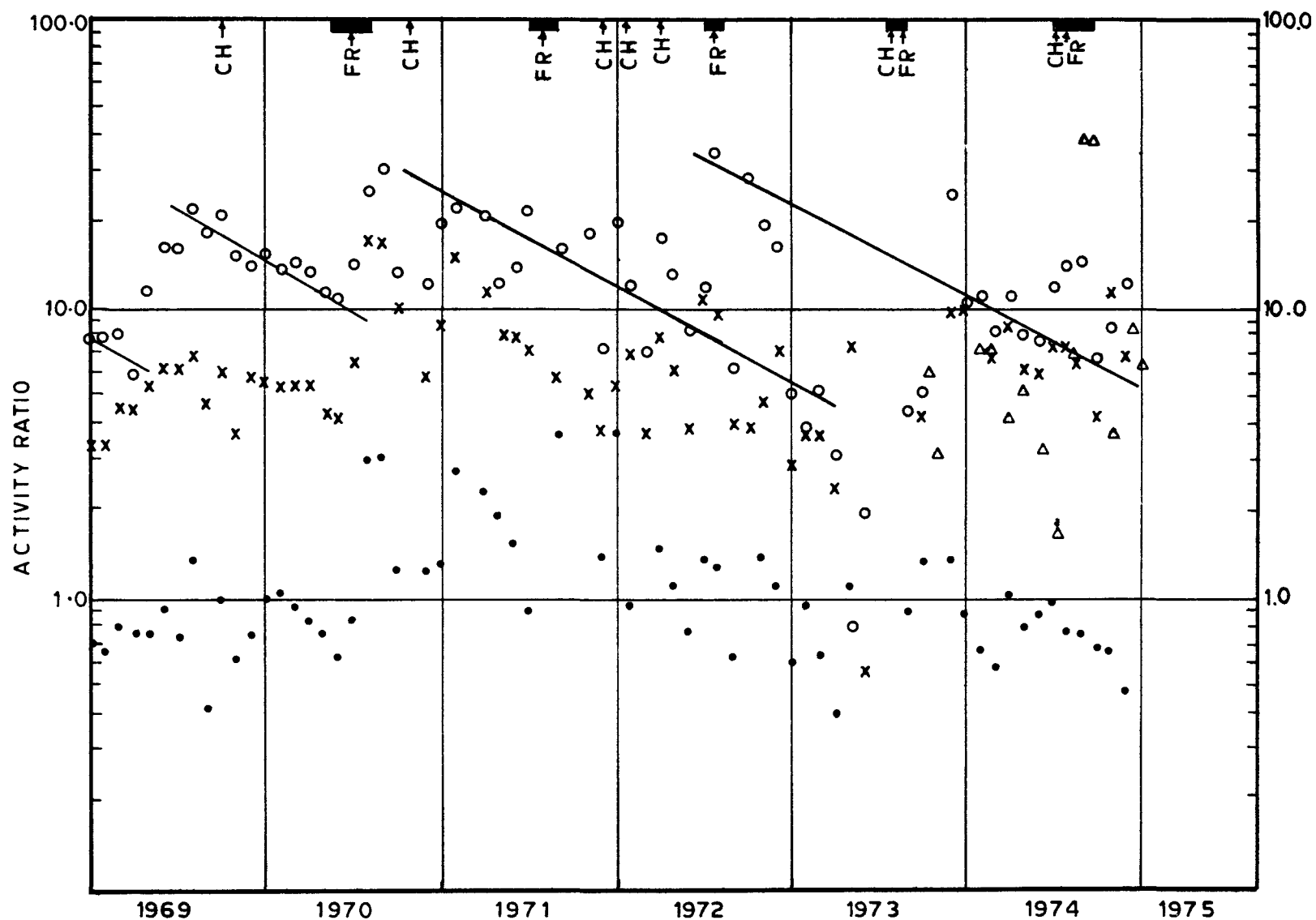


FIGURE: 5 FISSION-PRODUCT ACTIVITY RATIOS IN SURFACE AIR AT BOMBAY.  
(LEGEND AS SHOWN IN FIG. 4.  $\Delta = \text{Zr-95/Cs-137}$ )

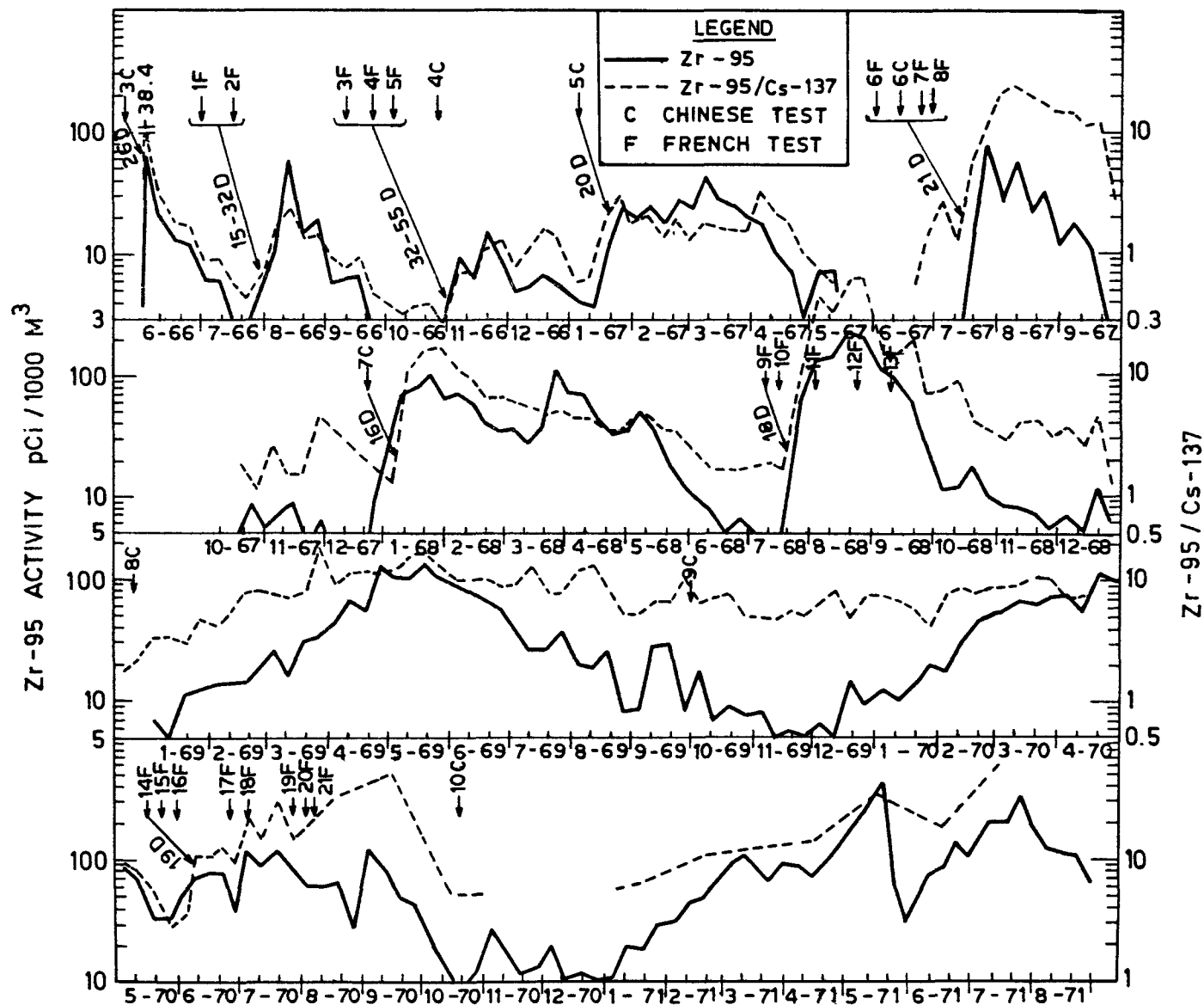


FIGURE 6 . Zr-95 ACTIVITY AND Zr-95/Cs-137 IN SURFACE AIR AT BOMBAY .  
(SEVEN DAY MEAN CONCENTRATIONS. ARROWS INDICATE INCREASE  
DUE TO PARTICULAR TEST. NUMBERS GIVE THE TRAVEL TIME ).

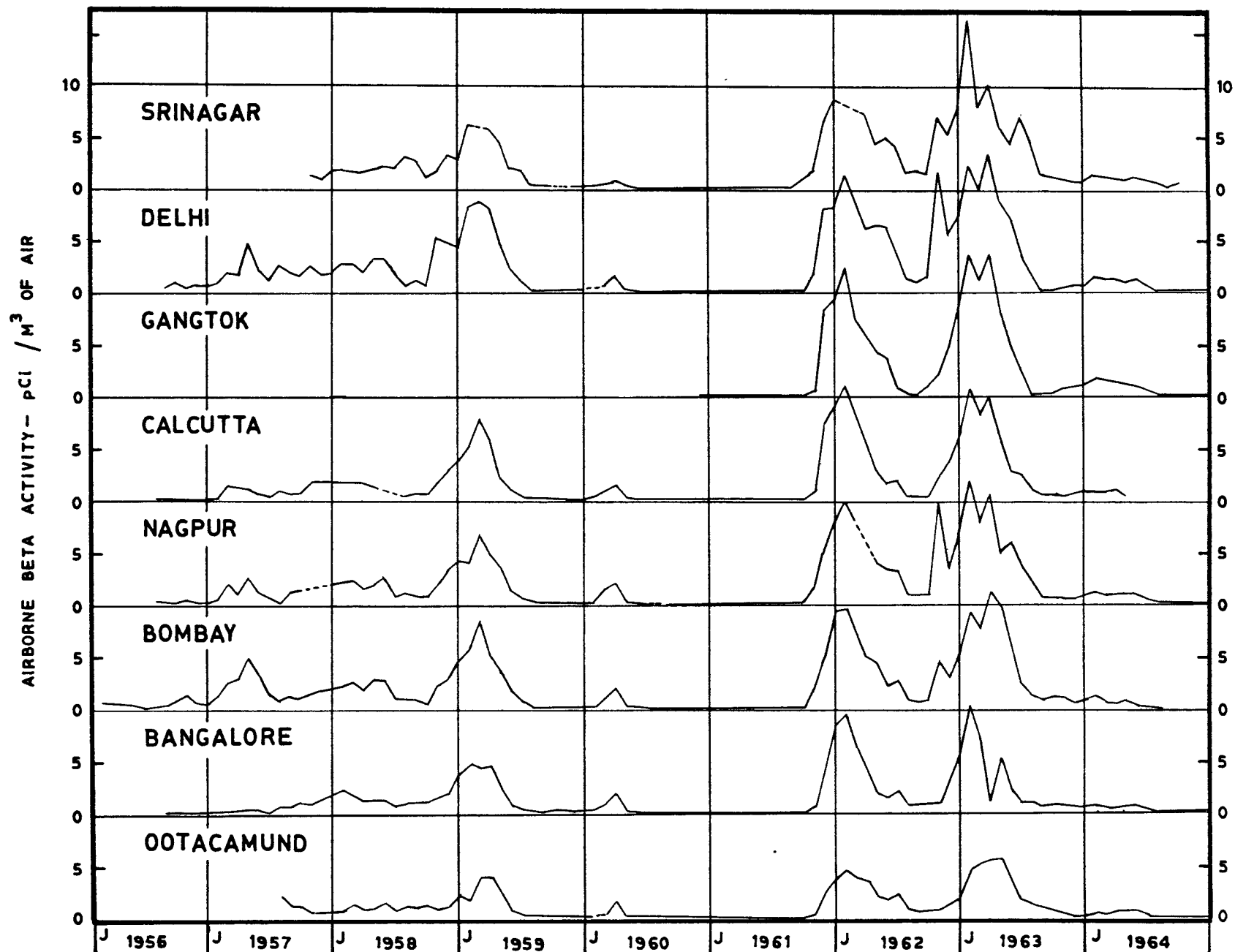


FIGURE:7 . AIRBORNE FALLOUT BETA ACTIVITY AT DIFFERENT STATIONS IN INDIA (1956 -1964

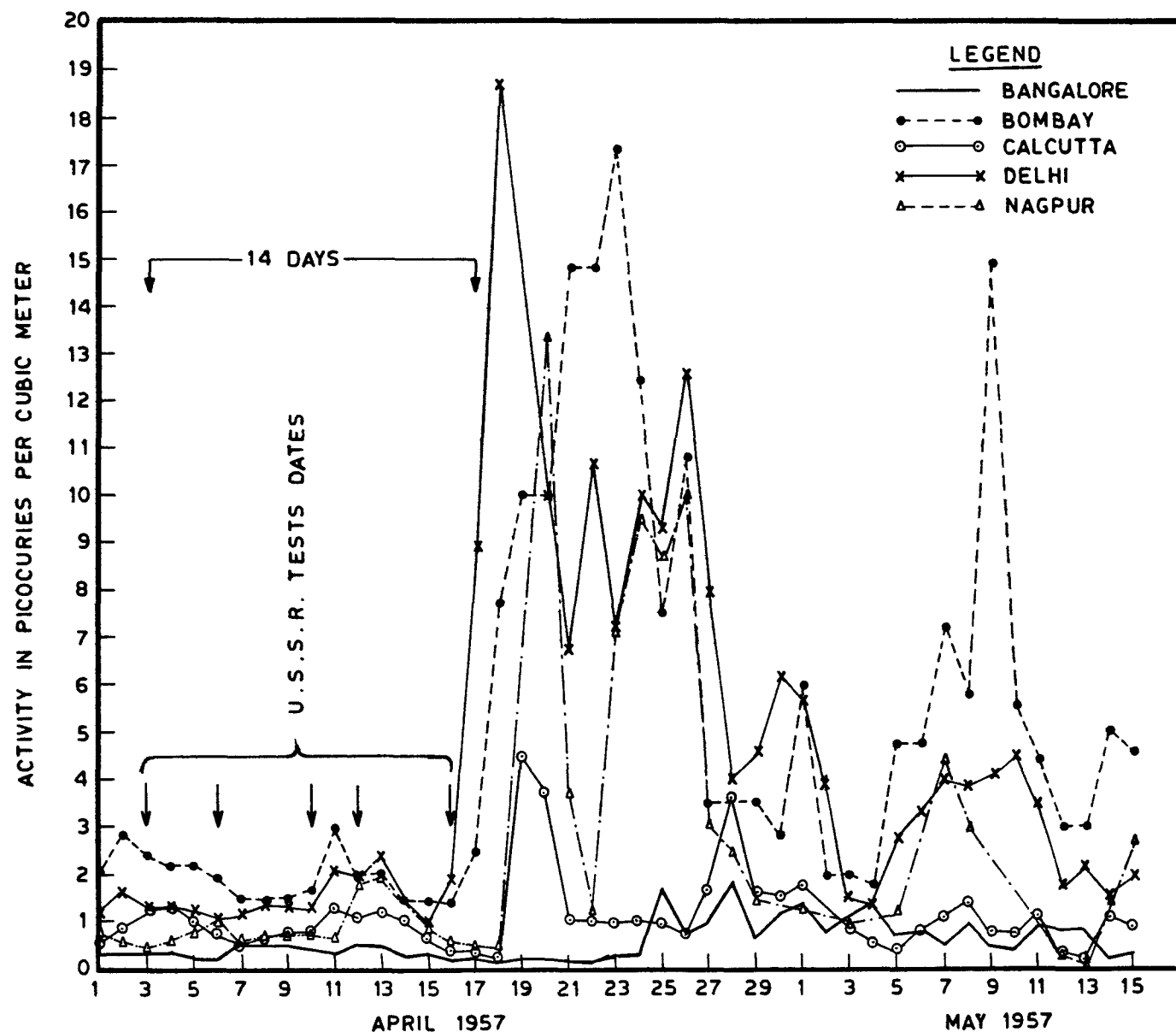


FIGURE : 8 . FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT BANGALORE , BOMBAY , CALCUTTA , DELHI AND NAGPUR FROM U.S.S.R. TESTS OF APRIL 1957.

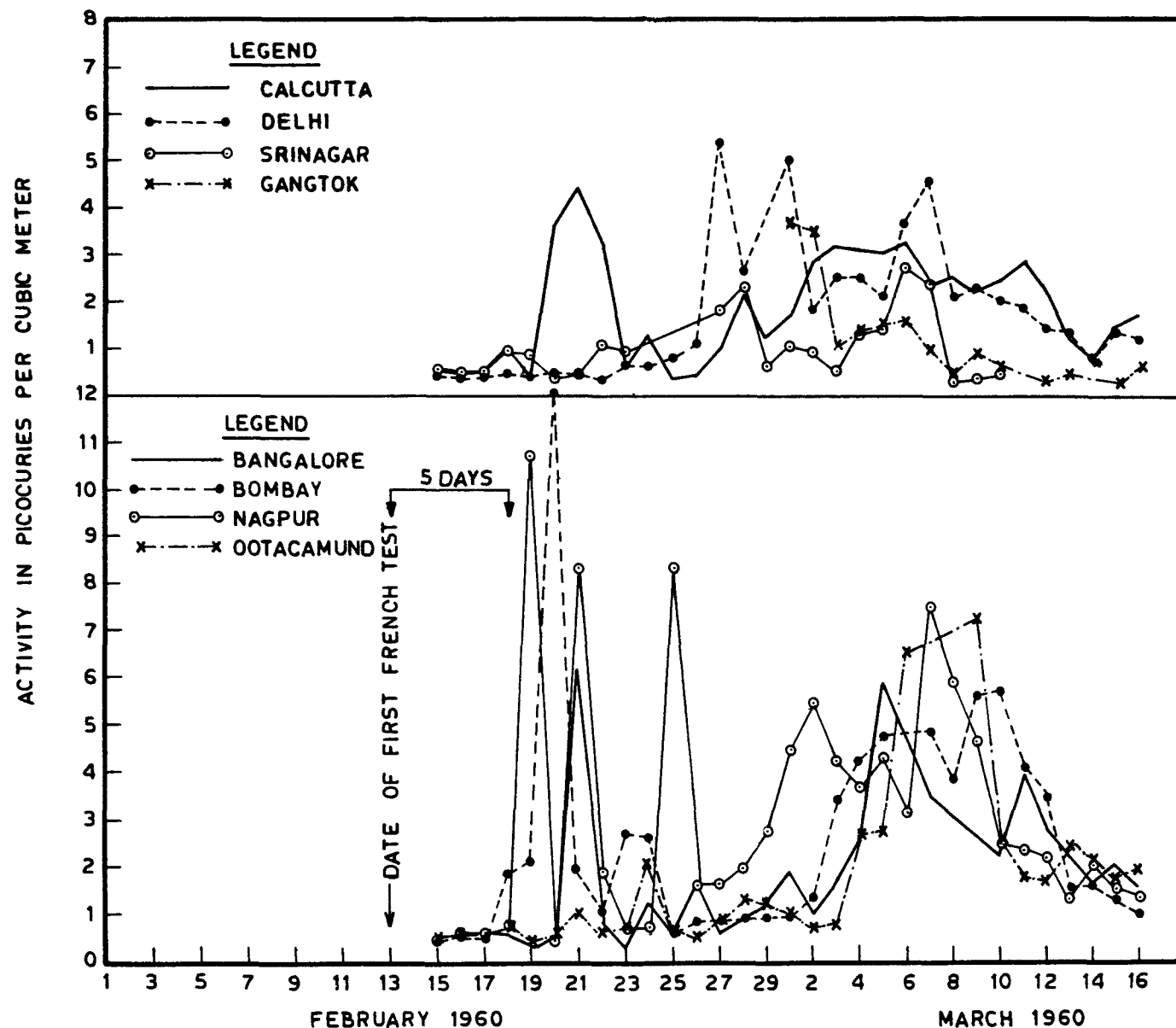


FIGURE:9 .FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT BANGALORE , BOMBAY, CALCUTTA , DELHI, NAGPUR , OOTACAMUND, SRINAGAR AND GANGTOK FROM THE FIRST FRENCH TEST OF 13 FEBRUARY 1960.

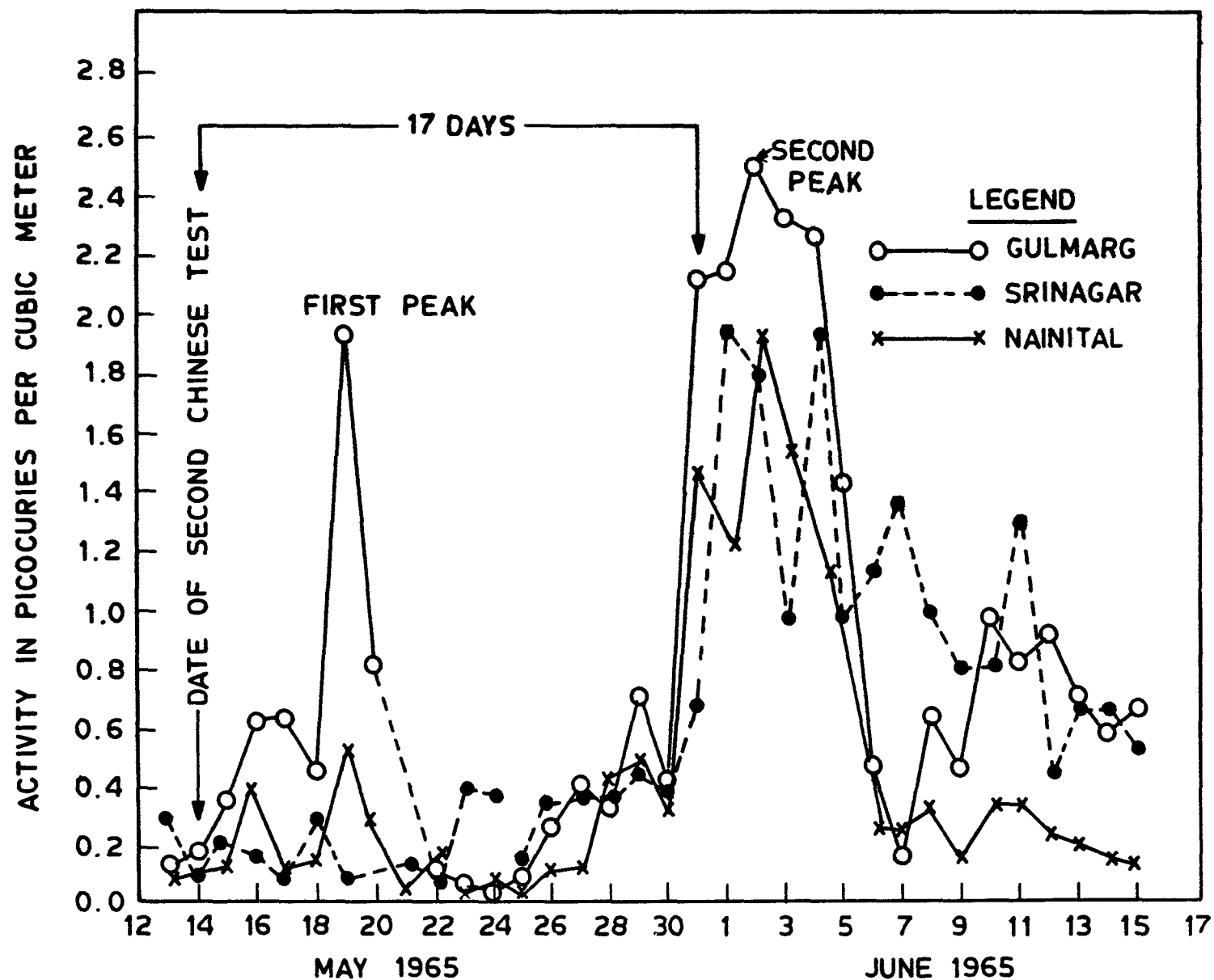


FIGURE: 10 . FISSION PRODUCT RADIOACTIVITY OF SURFACE AIR AT GULMARG, SRINAGAR AND NAINITAL FROM THE SECOND CHINESE TEST OF 14 MAY 1965. ( TOTAL BETA ACTIVITY ).

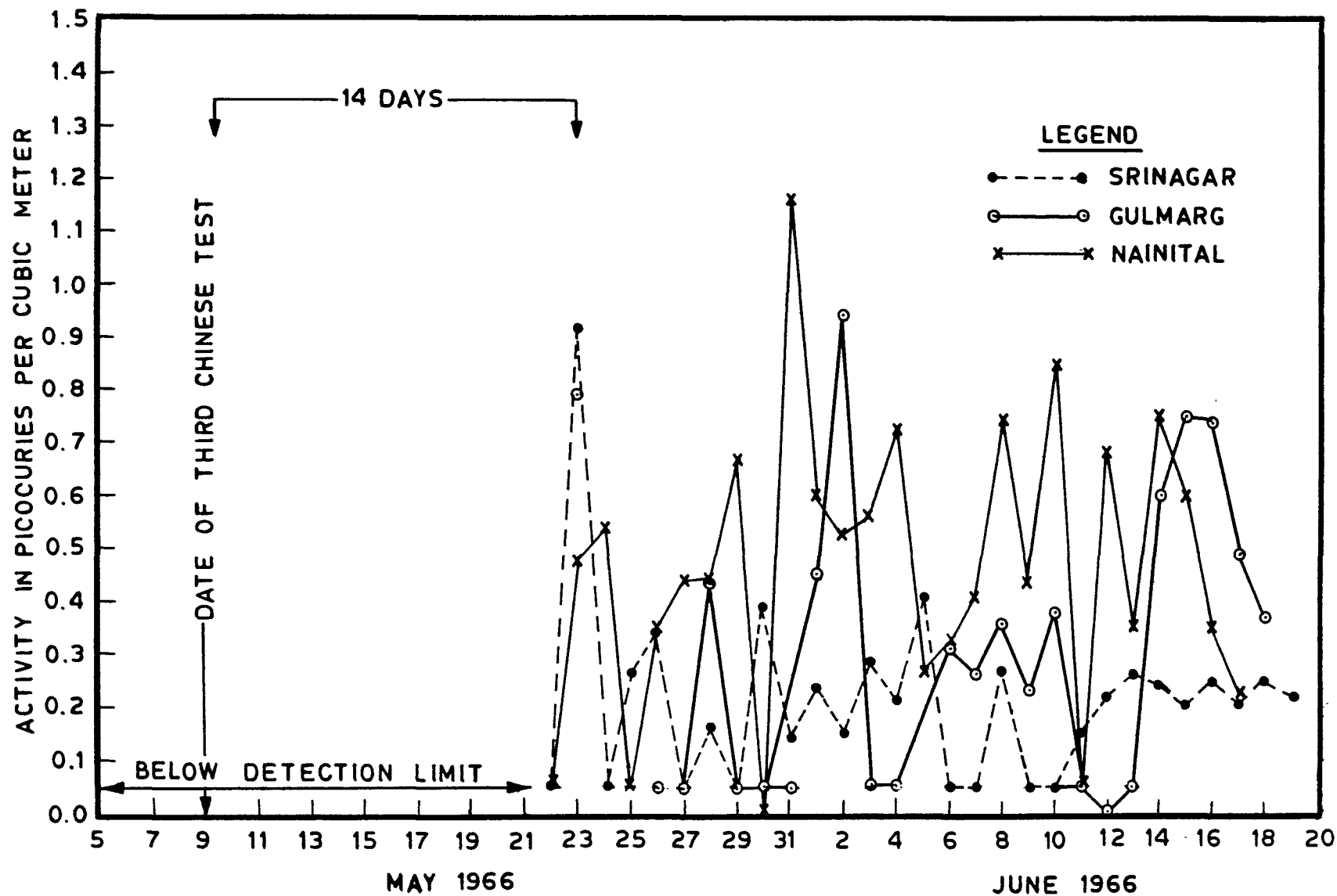


FIGURE:11 .FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT GULMARG , SRINAGAR AND NAINITAL FROM THE THIRD CHINESE TEST OF 9 MAY 1966.



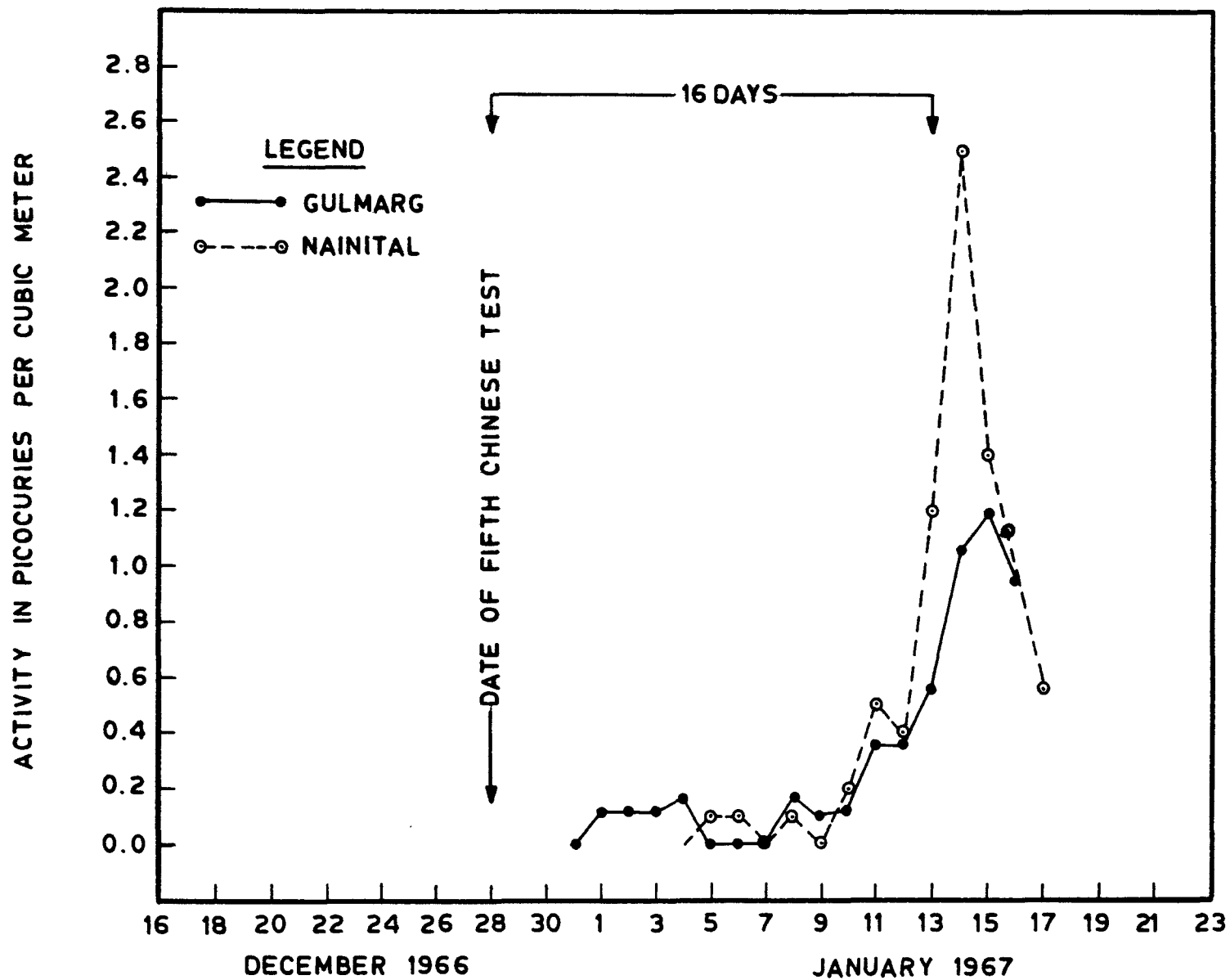


FIGURE:12 . FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT GULMARG AND NAINITAL FROM THE FIFTH CHINESE TEST OF 28 DECEMBER 1966.

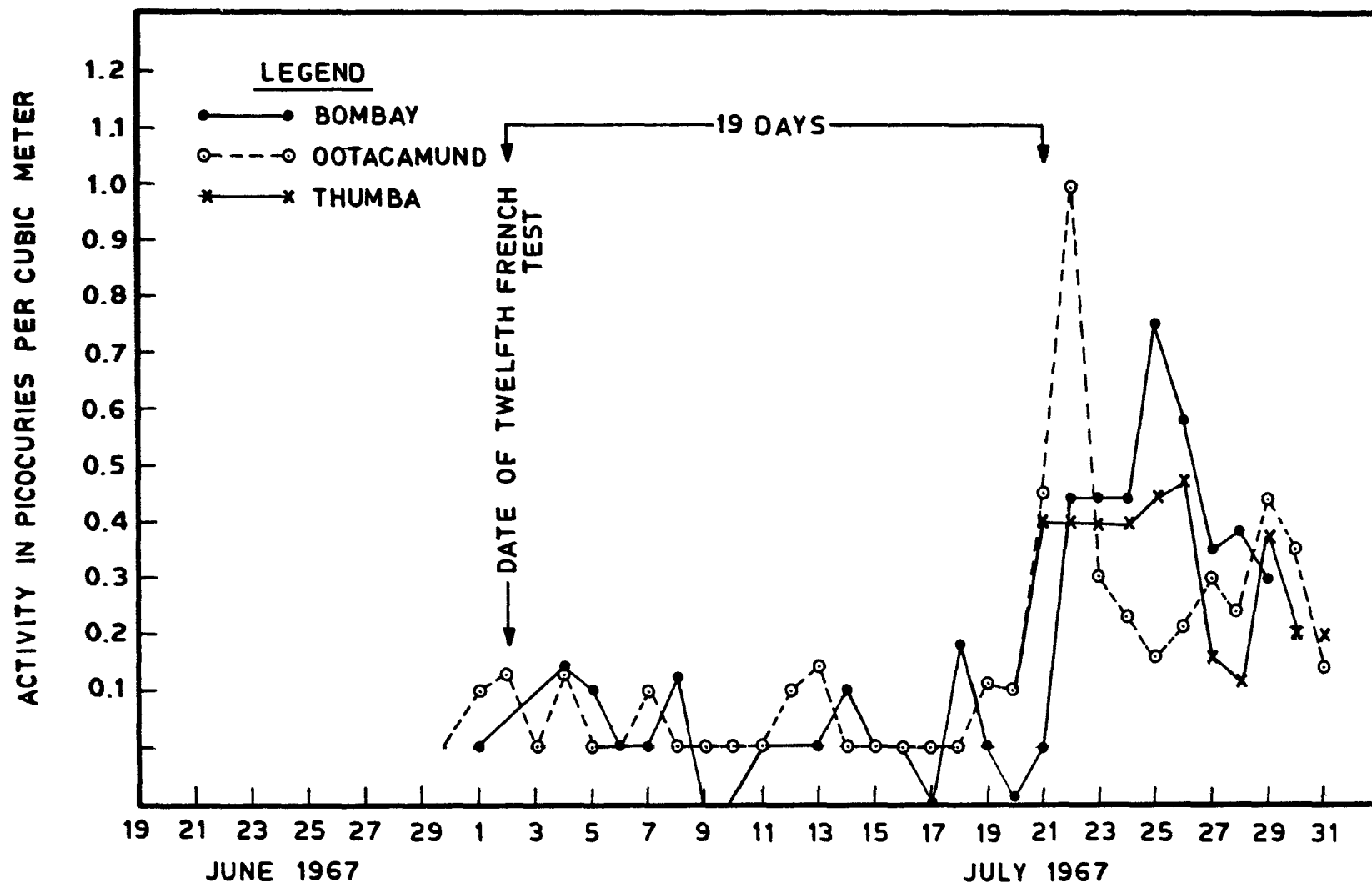


FIGURE:13 .FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT BOMBAY , OOTACAMUND AND THUMBA FROM THE TWELFTH FRENCH TEST OF 2 JULY 1967.

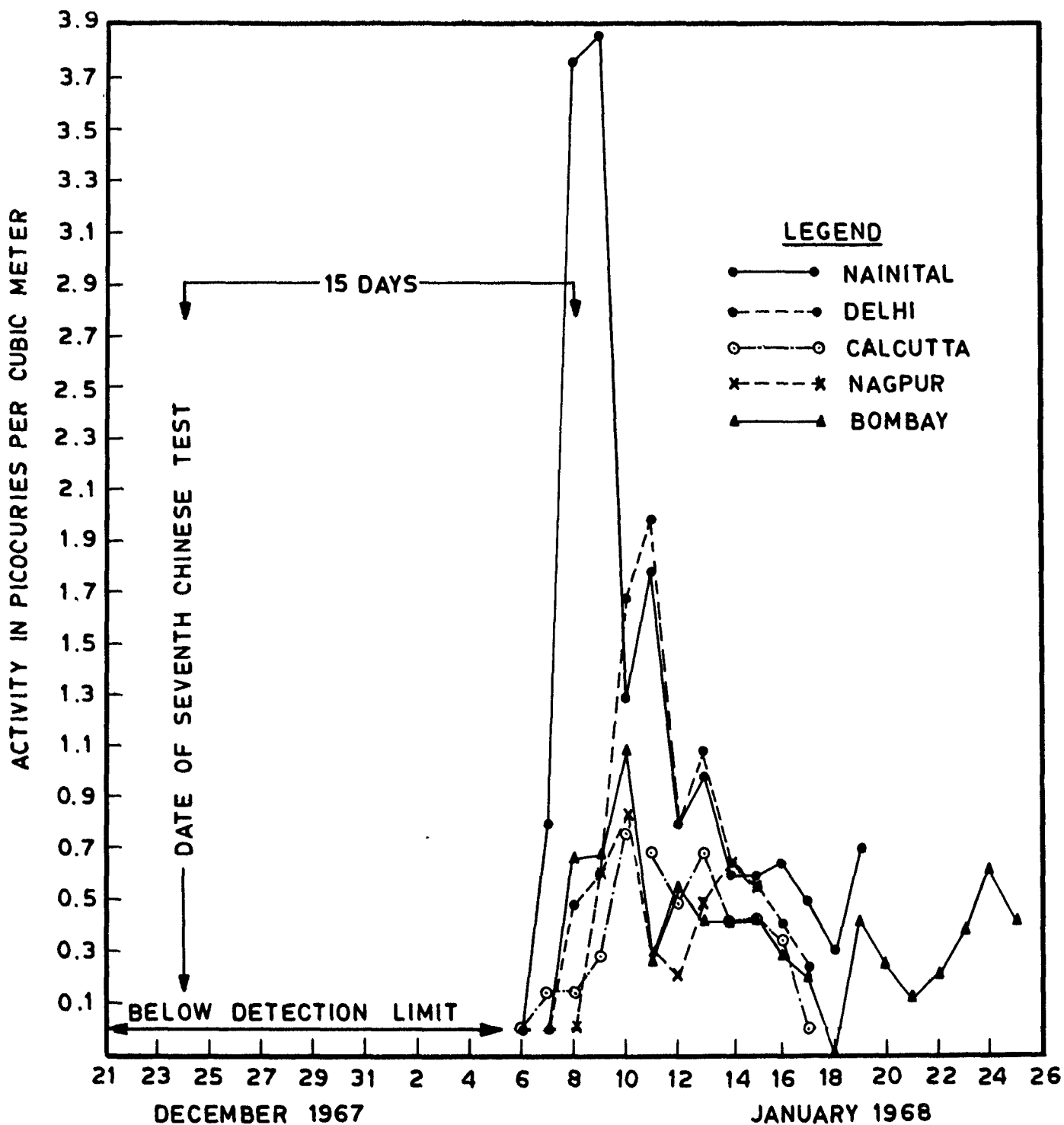


FIGURE:14. FISSION PRODUCT TOTAL BETA RADIOACTIVITY OF SURFACE AIR AT NAINITAL, DELHI, CALCUTTA, NAGPUR AND BOMBAY FROM THE SEVENTH CHINESE TEST OF DECEMBER 1967.

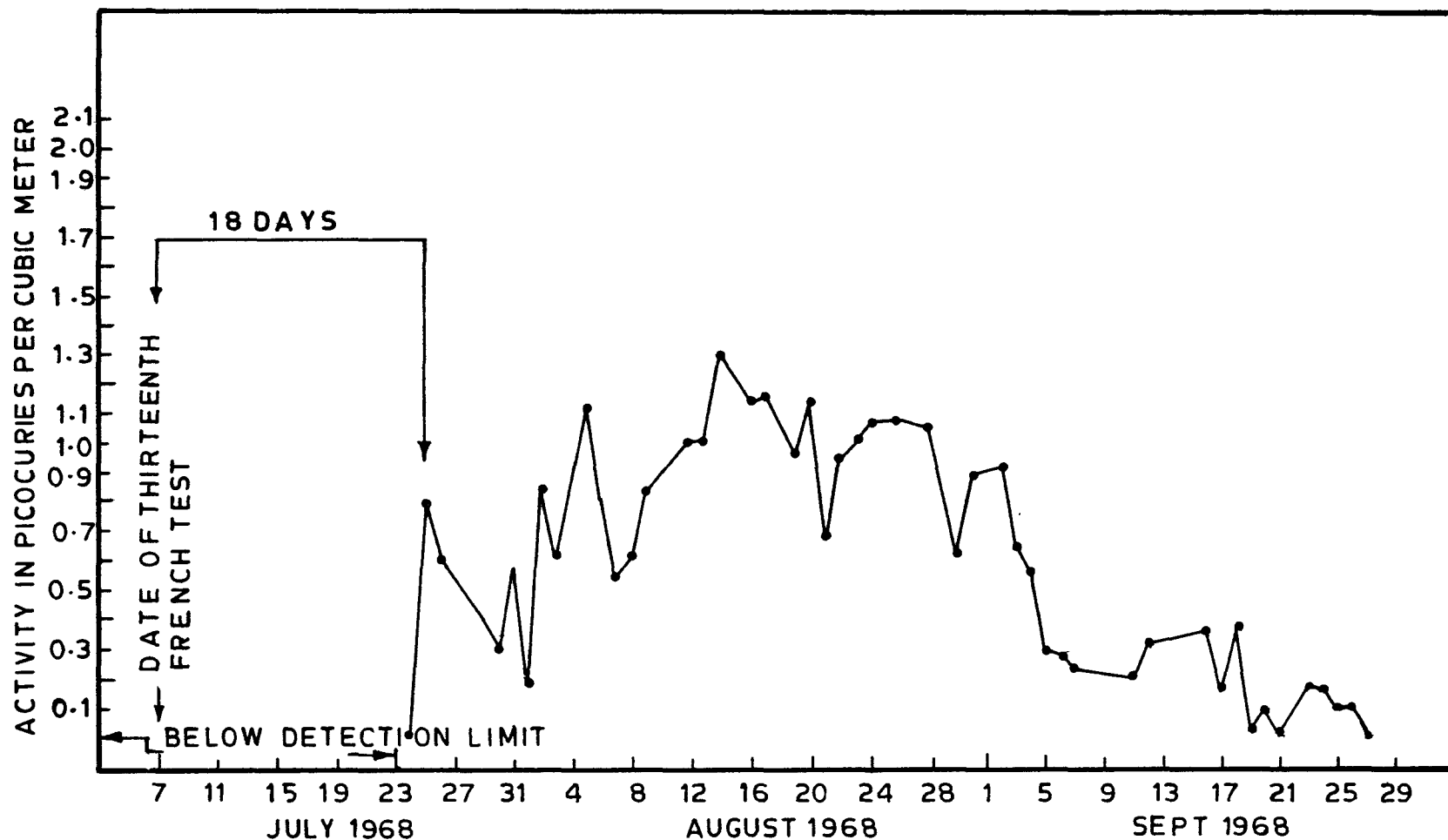


FIGURE: 15 . FISSION PRODUCT TOTAL BETA ACTIVITY OF SURFACE AIR AT BOMBAY FROM THE THIRTEENTH FRENCH TEST OF 7 JULY 1968 .

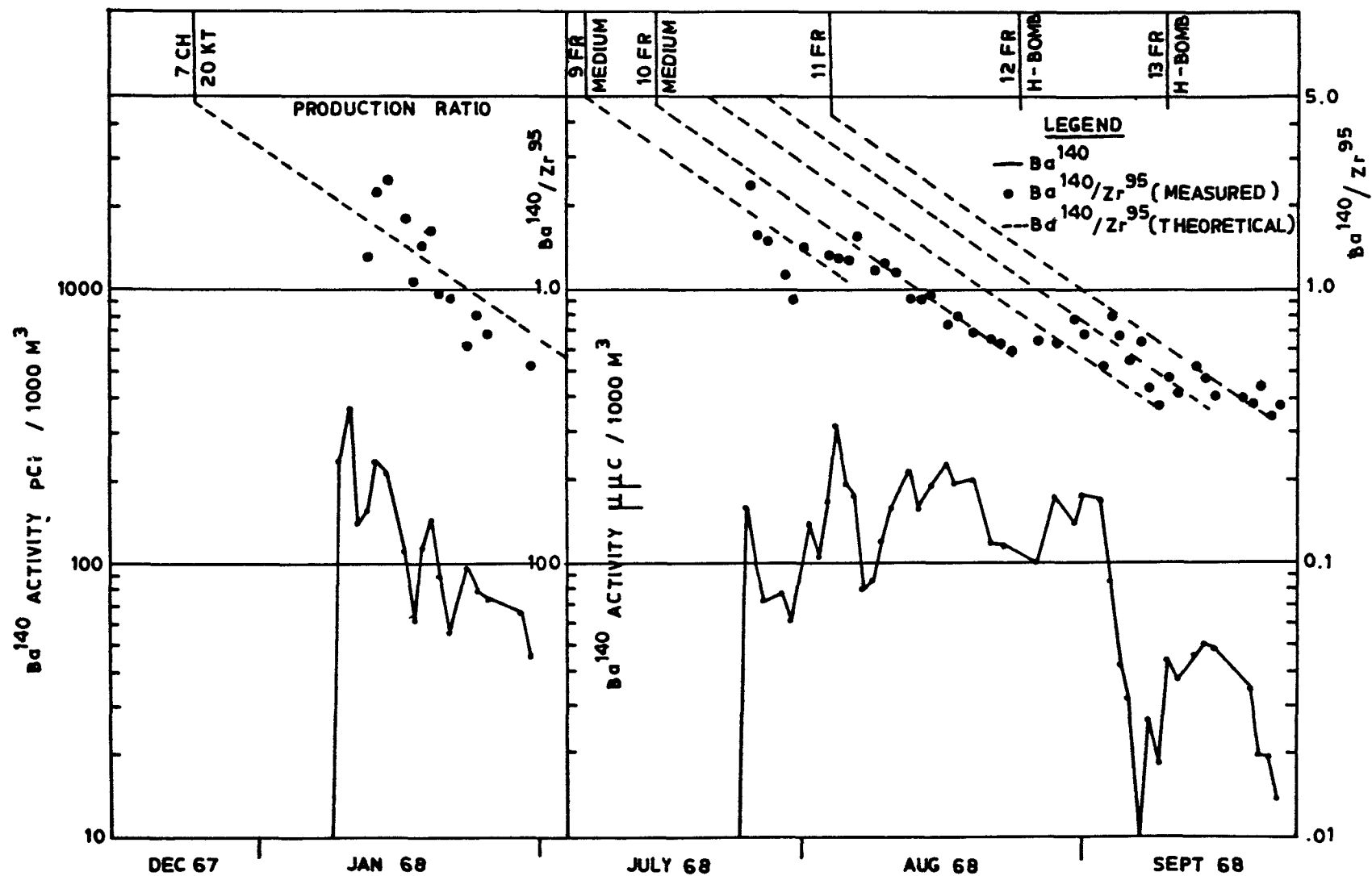


FIGURE 16.  $Ba^{140}$  ACTIVITY AND  $Ba^{140}/Zr^{95}$  IN SURFACE AIR AT BOMBAY. AFTER THE CHINESE TEST OF DEC. 1967. AND THE FRENCH TESTS OF 1968.

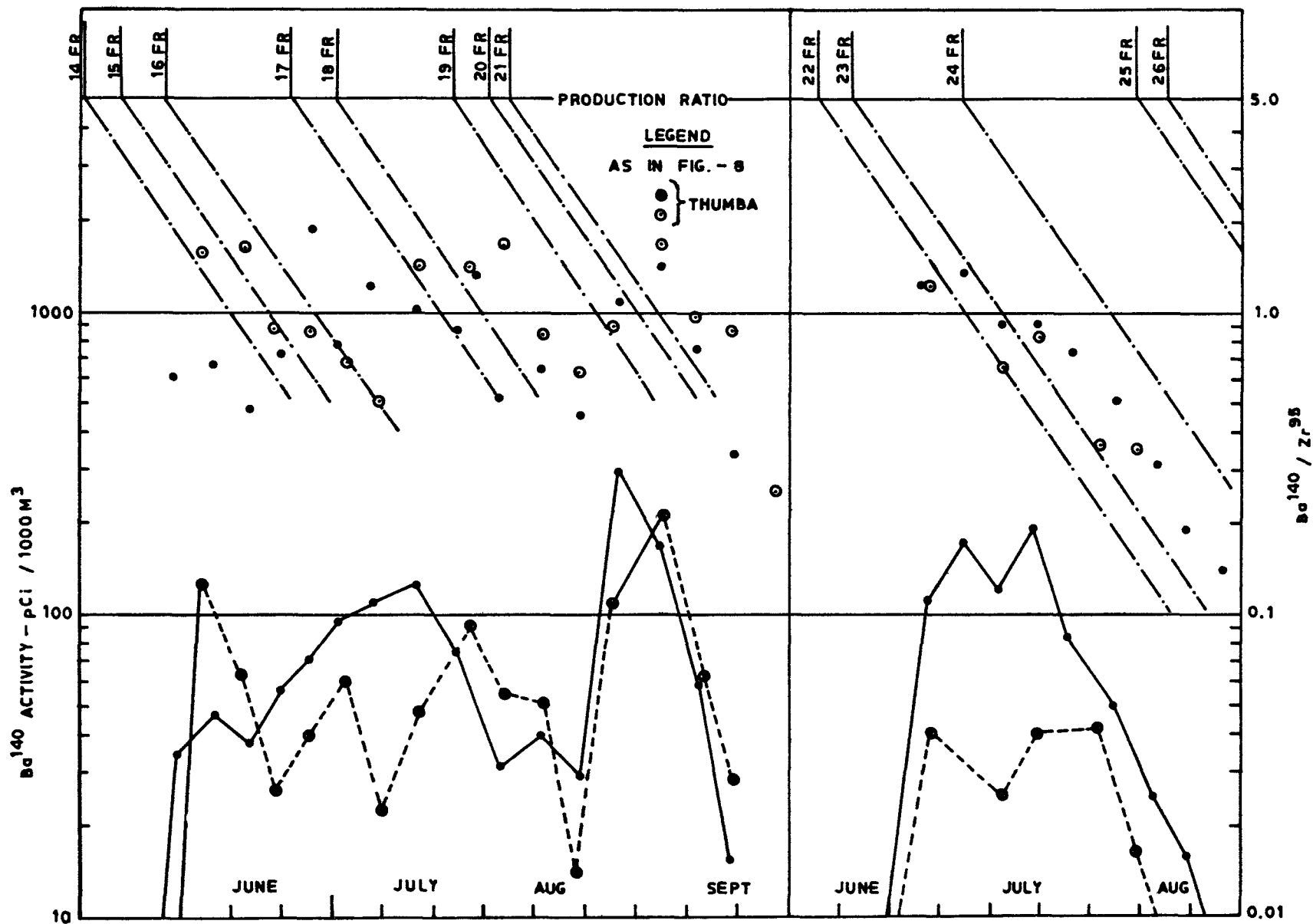


FIGURE 17.  $Ba^{140}$  ACTIVITY AND  $Ba^{140}/Zr^{95}$  IN SURFACE AIR AT BOMBAY AND THUMBA AFTER THE FRENCH TESTS OF 1970 AND 1971. (SEVEN DAY MEAN CONCENTRATIONS)

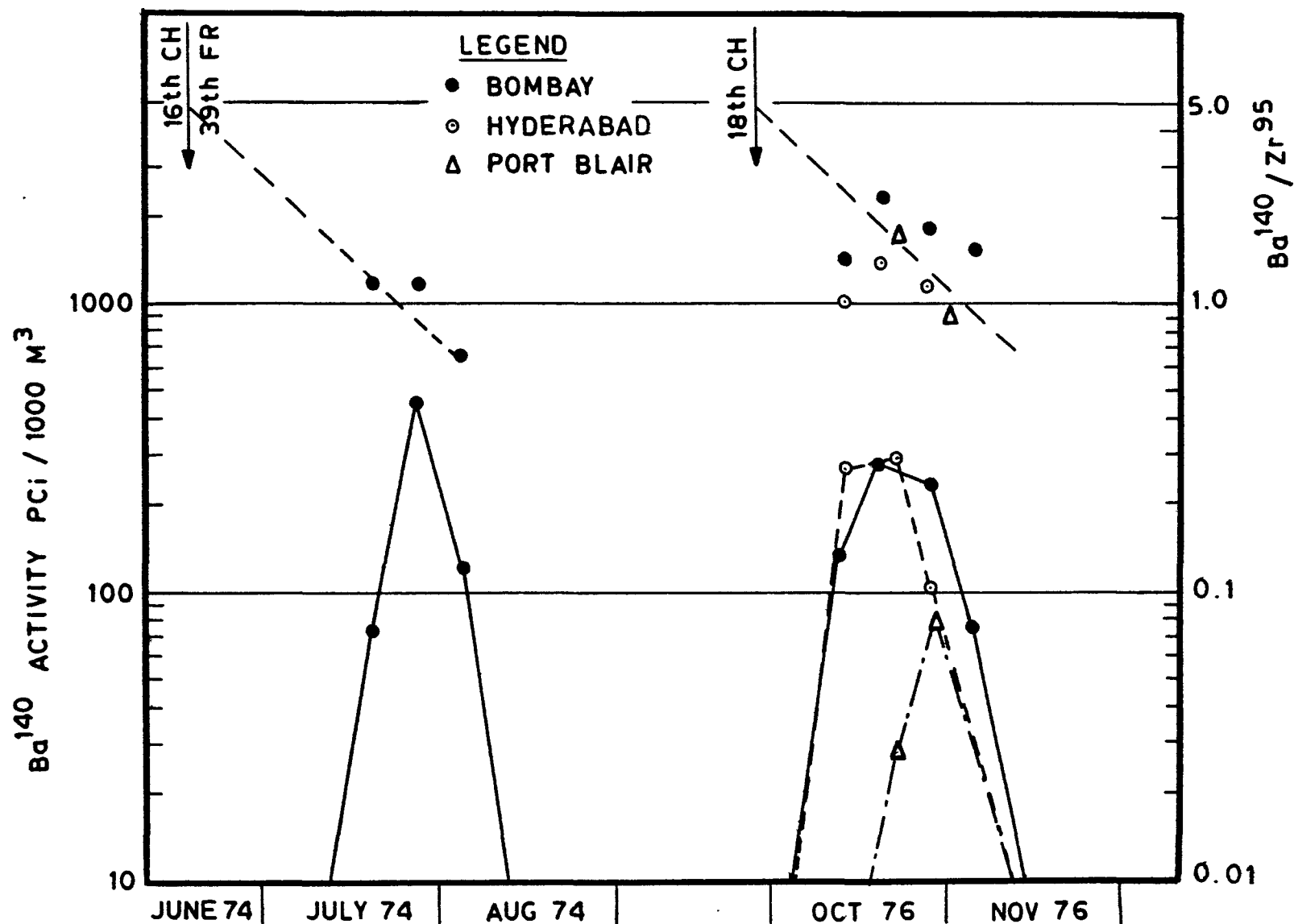


FIGURE:18 . Ba<sup>140</sup> ACTIVITY AND Ba<sup>140</sup>/Zr<sup>95</sup> IN SURFACE AIR AFTER THE FRENCH /CHINESE TESTS OF JUNE 74 AND THE CHINESE TEST OF SEPT. 76. (SEVEN DAY MEAN CONCENTRATIONS )

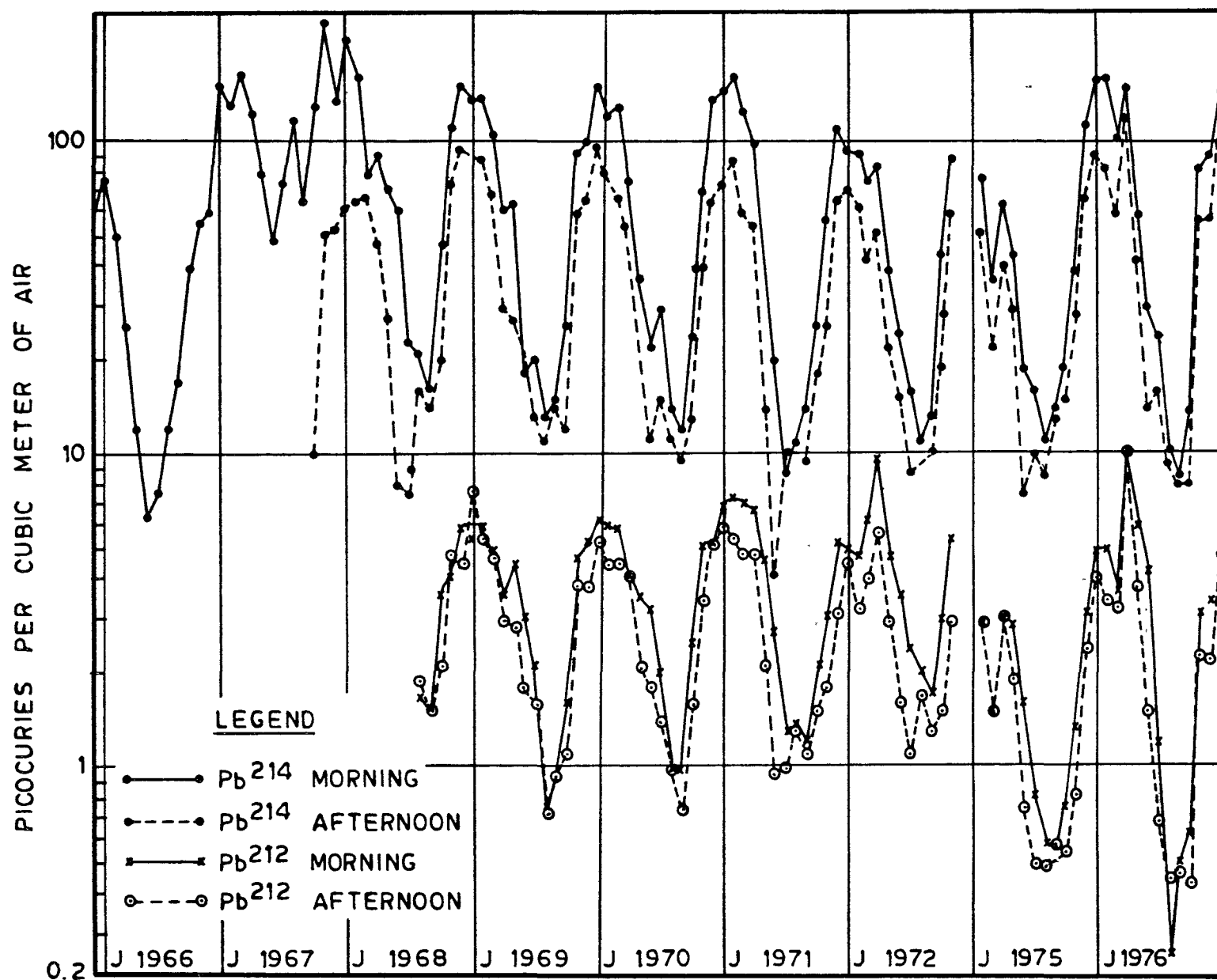


FIGURE 19  $Pb^{212}$  AND  $Pb^{214}$  ACTIVITIES IN SURFACE AIR AT BOMBAY —  
MONTHLY MEAN CONCENTRATIONS ( NO DATA FOR 1973 AND 1974 )



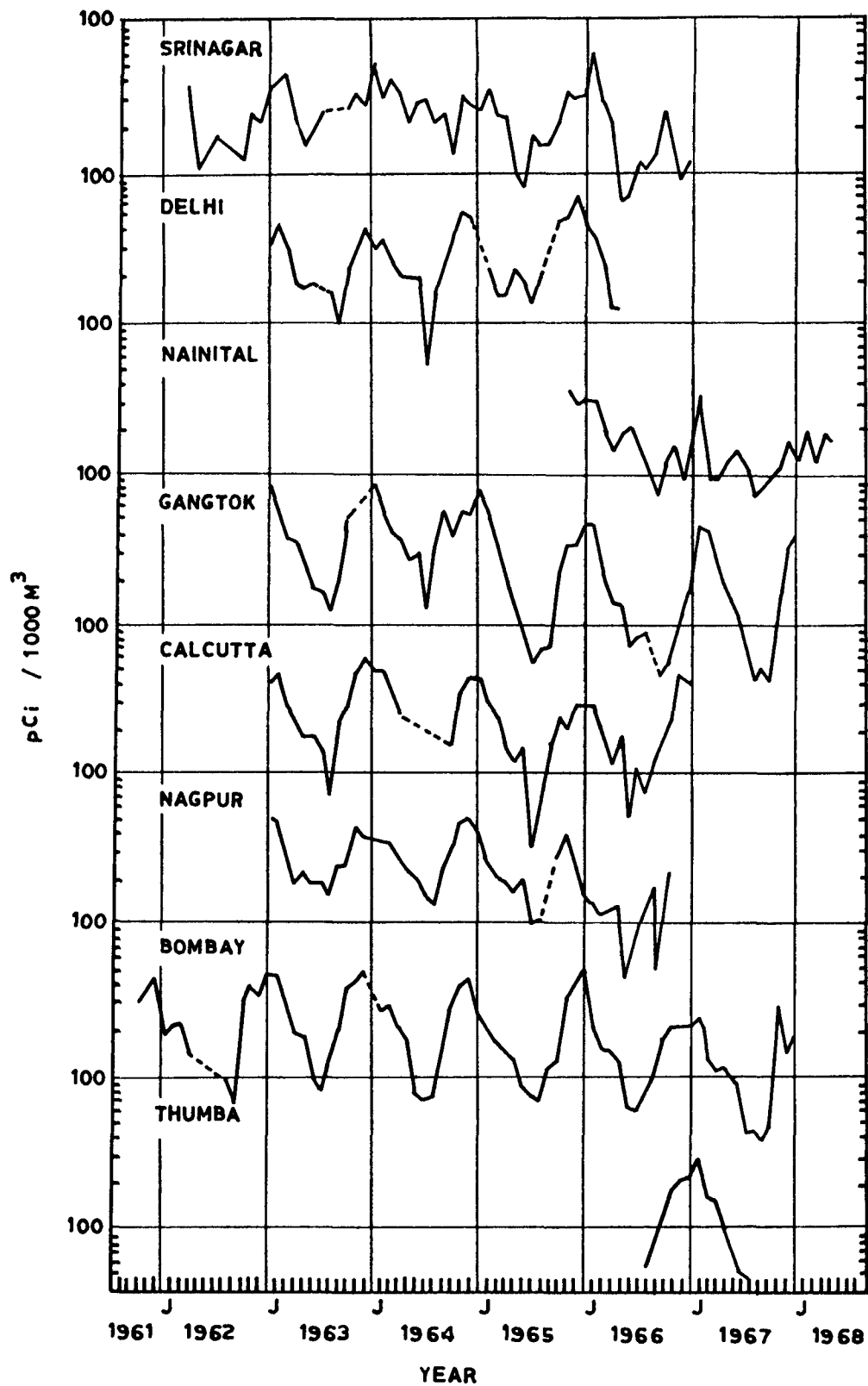


FIGURE 20. Pb-210 (RnD) ACTIVITY IN SURFACE AIR AT DIFFERENT STATIONS IN INDIA.





DEPARTMENT OF HEALTH  
NEW ZEALAND

ENVIRONMENTAL RADIOACTIVITY  
ANNUAL REPORT  
1976

NATIONAL RADIATION LABORATORY  
P. O. BOX 25-099, CHRISTCHURCH  
NEW ZEALAND

APRIL 1977

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The Environmental Radioactivity Section of the Laboratory organised the monitoring operations and analysed the samples. The Officer in Charge of this section, Mr L. P. Gregory, was responsible for reporting and interpreting results. He was assisted professionally by Mr T. Baltakmens and Dr K. M. Matthews, and technically by Mr G. N. Connor and Miss M. Roberts.

for H. R. Atkinson  
(Director)

Published with the authority of the Director-General of Health.

## UNITS AND REFERENCE LEVELS

### Units

The unit of radioactivity, the "Curie" (Ci), equal to  $3.7 \times 10^{10}$  disintegrations per second, is too large for environmental levels, and subdivisions are used in this report: the millicurie (mCi) =  $10^{-3}$  Ci, and the picocurie (pCi) =  $10^{-12}$  Ci or 2.22 disintegrations per minute.

Deposition of radioactivity is given as millicuries per square kilometre (mCi/km<sup>2</sup>)

### Concentration of radioactivity

in air: is given as picocuries per cubic metre (pCi/m<sup>3</sup>)

in rain: is given as picocuries per litre (pCi/l) and is derived from the relationship:

$$\text{pCi/l} = \frac{\text{mCi/km}^2 \times 100}{\text{centimetres of rain}}$$

in milk: strontium-90 (<sup>90</sup>Sr) concentration is given as picocuries per gram of calcium (pCi/gCa)

caesium-137 (<sup>137</sup>Cs) concentration is given as picocuries per gram of potassium (pCi/gK)

(One litre of milk contains about 1.2 g of calcium and about 1.4 g of potassium)

### Reference Levels

The following reference levels, against which measured levels reported herein may be compared, have been adopted for New Zealand:

### Mixed fission products between 10 and 80 days old (Total Beta Activity)

in air: 300 pCi/m<sup>3</sup>

in rain: 6000 pCi/l

strontium-90 in milk: 270 pCi/gCa

caesium-137 in milk: 7000 pCi/gK

## SUMMARY

During 1976 the deposition of strontium-90 at nine New Zealand stations averaged 0.1 millicuries per square kilometre. This was the lowest value recorded since measurements commenced in 1960:

During 1964 a maximum deposition, averaging  $3.6 \text{ mCi/km}^2$ , resulted from the large scale U.S.S.R. and U.S.A. atmospheric nuclear tests of 1961-2. Subsequently levels decreased and during the French atmospheric tests in the South Pacific from 1966 to 1974 the annual average depositions in New Zealand ranged from 0.3 to  $1.4 \text{ mCi/km}^2$ .

The concentrations of strontium-90 and caesium-137 in milk reflect these changes in fallout deposition. The average concentrations during 1976 were the lowest since measurements commenced.

French underground nuclear tests in the South Pacific commenced in mid-1975. Since then continuous monitoring has also been conducted at six Pacific Island stations. No fresh fission products, either from venting of underground tests or from atmospheric tests in the Northern Hemisphere, have been detected since this programme started.

The levels recorded during 1976 were very small fractions of the reference levels and thus do not constitute a public health hazard. Moreover, the radiation dose resulting from the long-term average levels, summarised herein, is small compared not only with natural background but also with common variations in natural background.

## CONTENTS

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Lead-210 Deposition	5
Strontium-90 and Caesium-137 in Milk	6
Miscellaneous	8
Appendix	10

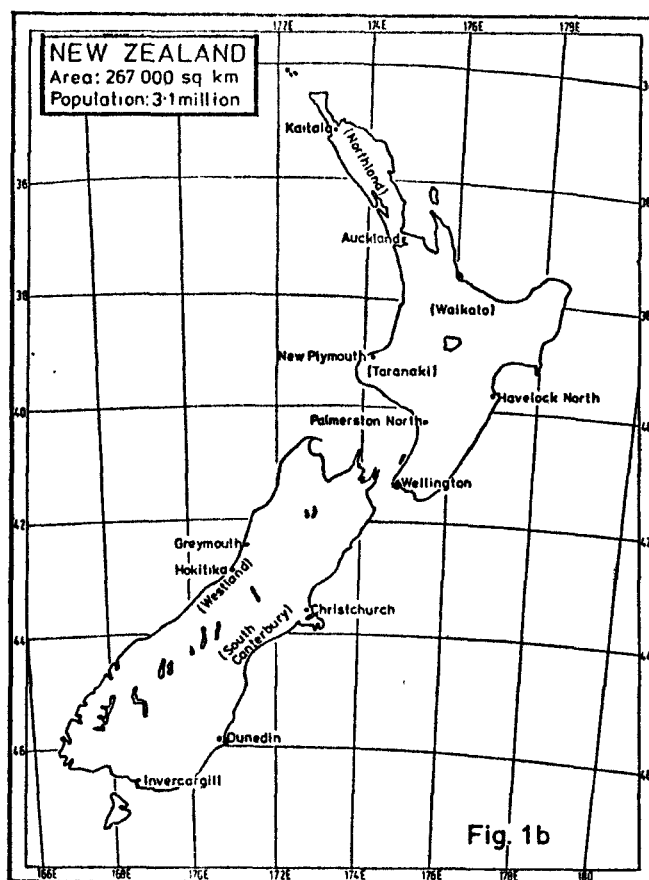
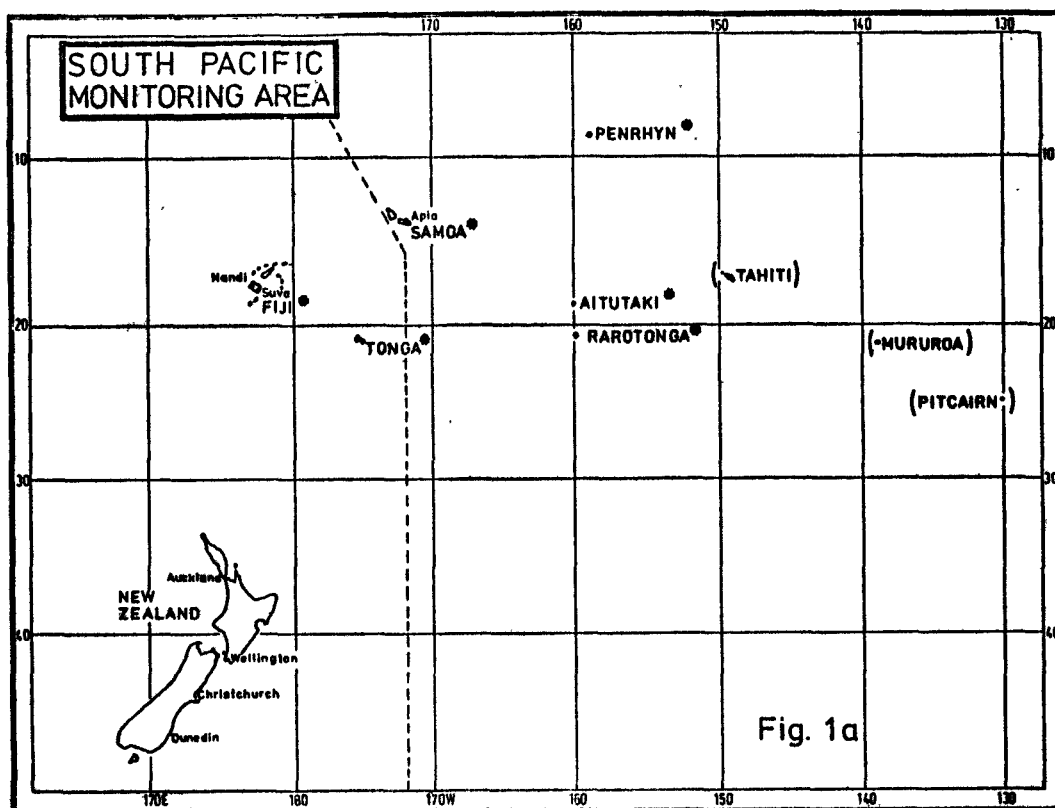


FIG. 1 MONITORING AND COLLECTING STATIONS ON PACIFIC ISLANDS AND IN NEW ZEALAND.

## INTRODUCTION

This report continues the series of annual reports on levels of environmental radioactivity in New Zealand and in the South Pacific area.

In September 1974 France terminated the series of atmospheric nuclear tests which had been conducted in the Tuamotu Archipelago in the South Pacific, and in June 1975 commenced underground testing in the same area. Consequently the Laboratory's monitoring programme was modified in mid-1975 and this programme has been continued during 1976. The modified programme, designed to detect any venting to the atmosphere of fission products from underground tests, consists of daily gamma radiation monitoring at Penrhyn Island and continuous collection of air filter and rainwater samples from five Pacific Island and four New Zealand stations. The samples are sent to the Laboratory for measurement of total beta activity.

The routine programme for measurement of long-term radioactive fallout from earlier atmospheric tests was also continued during 1976. In this programme emphasis is given to the measurement of the two most potentially hazardous long-lived radionuclides, strontium-90 and caesium-137. Monthly depositions of strontium-90 in rain are measured at nine New Zealand and two Pacific Island stations. (Naturally-occurring lead-210 is also evaluated concurrently.) Strontium-90 and caesium-137 concentrations are measured in milk from nine New Zealand stations.

Monitoring and sample collecting stations are shown in Figs. 1 a and b.

Fallout levels during recent years and particularly during 1976 have been very low and this report has been considerably shortened. The reader is referred to the previous annual reports (1), and the reports (2) on special monitoring of French atmospheric nuclear tests (1966-1974). These reports give further information on terms of reference, potential health hazard, adoption of reference levels, and technical information on procedures. They also include graphical presentation of results allowing historical and geographical comparison.

### GAMMA RADIATION MONITORING AT PENRHYN ISLAND

No increases above the normal background radiation level have been detected at Penrhyn Island since this monitoring programme started in July 1975.

---

(1) "Environmental Radioactivity":

Annual Report 1971, Report No. NRL-F/48, June 1972  
Annual Report 1972, Report No. NRL-F/50, April 1973  
Annual Report 1973, Report No. NRL-F/52, June 1974  
Annual Report 1974, Report No. NRL-F/54, June 1975  
Annual Report 1975, Report No. NRL-F/55, June 1976

(2) "Environmental Radioactivity. Fallout from Nuclear Weapons Tests Conducted by France in the South Pacific . . . and comparisons with previous test series." Report Nos: NRL-F/47, March 1972 (Summarising all previous monitoring results since 1966); NRL-F/49, October 1972; NRL-F/51, November 1973, and NRL-F/53, November 1974.

## TOTAL BETA ACTIVITY IN AIR AND RAIN

Normally the short-lived decay products of naturally-occurring radon account for most of the beta activity in air. Ground level air over continents has a beta activity commonly ranging between 60 and 600 picocuries per cubic metre, but under certain conditions the beta activity may be up to ten times the upper value of this range. Air filter samples, and also rainwater samples, which are collected for measurement of fission products, are held for four days to allow this natural radioactivity to decay away and are then measured for residual beta activity which is due to radioactive fallout. Hereafter the term "total beta activity" refers only to this residual radioactivity due to fission products.

### 1. Fission Products in Air

During 1976 air was monitored continuously at the New Zealand and Pacific Island stations listed in Table 1 below. The filters were changed three times each week and were measured for total beta activity at the Laboratory. Without exception average levels each month during 1976 were at or below the limit of detection (0.01 pCi/m<sup>3</sup>) at each station. Therefore, for the first time in this series of reports, individual results or monthly averages are not tabulated in the Appendix. However, the 1976 annual averages are included in Table 1 for comparison with those during previous years.

**TABLE 1 - Total Beta Activity in Air - Annual Averages (pCi/m<sup>3</sup>)**

	<u>New Zealand</u>				<u>Pacific Islands</u>				
	AK	WN	HK	CH	FJ	SM	TO	AI	RA
1966	0.14	(0.10)		0.11					
1967	0.08	0.05		0.06					
1968	0.12	0.10		0.07	Average levels ranged from 0.04 - 6.22 pCi/m <sup>3</sup> in the South Pacific area during the special monitoring programmes				
1969	0.12	0.09		0.07					
1970	0.16	0.12	(0.12)	0.10					
1971	0.21	0.12	0.16	0.15					
1972	0.06	0.05	0.05	0.05					
1973	0.02	0.01	0.02	0.02					
1974	0.08	0.05	0.07	0.05					
1975	0.03	0.03	0.03	0.02	<0.01	<0.01	0.01	<0.01	0.01
1976	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

- Notes: 1. The stations are: Auckland, Wellington, Hokitika, Christchurch, Fiji, Samoa, Tonga, Aitutaki, and Rarotonga.  
 2. Values in parenthesis are estimates: At Wellington measurements commenced 12 July 1966; at Hokitika, 1 May 1970.  
 3. At the Pacific Islands the 1975 results are for June-Dec. only (during this period the New Zealand results were similar, averaging 0.01 pCi/m<sup>3</sup>).

It can be seen that levels in New Zealand have been latitude dependent during the period of French atmospheric tests (1966-1974). For example, levels at Auckland are higher in most cases than those at Christchurch. Moreover, levels at the Pacific Islands, during the 3-6 month special monitoring programmes covering these tests, were significantly higher than New Zealand levels, averaging from 0.04 to 6.22 pCi/m<sup>3</sup>, depending on the locality monitored, the extent of nuclear testing, and meteorological conditions.

Since mid-1975, about nine months after the termination of the French atmospheric nuclear tests, levels have been at the limit of detection at all stations. There has been no indication of fresh fission products, either from venting of underground tests or from Northern Hemisphere atmospheric tests.

All levels of fission products in air tabulated here and particularly those during recent years have been very small fractions of the reference levels.



## 2. Fission Products in Rain

During 1976 weekly funnel and bottle collections of rainwater were made at the same stations providing air filter samples. The rainwater samples were despatched to the Laboratory where they were measured for total beta activity. Results for individual samples are given in Table 7 Appendix. The annual totals, since measurements started, are listed for each station in Table 2 below.

**TABLE 2 - Total Beta Activity in Rain - Weekly Depositions Summed Annually**  
(mCi/km<sup>2</sup>)

	New Zealand				Pacific Islands				
	AK	WN	HK	CH	FJ	SM	TO	AI	RA
1963				28					
1964				15					
1965				17					
1966			106	32					
1967			77	14					
1968			205	28					
1969			61	18					
1970	101	75	133	26					
1971	98	80	99	32					
1972	25	22	33	15					
1973	5	7	8	4					
1974	59	60	42	22					
1975	9	13	19	13	3	4	2	3	4
1976	3	3	4	2	3	3	4	4	3

- Notes:
1. See Table 1 (Note 1) for station names.
  2. Measurements commenced in Westland at Greymouth in July 1966 - the first result is for July-Dec. 1966 only. The station was transferred to Hokitika starting Jan. 1976.
  3. Measurement commenced at AK and WN in May 1970. The first results are for May-Dec. only.
  4. At the Pacific Islands the 1975 results are for June-Dec. only (during this period the New Zealand results were similar, averaging about 4 mCi/km<sup>2</sup>).

During 1976 the totalled depositions, ranging from 2 to 4 mCi/km<sup>2</sup>, were the lowest recorded and the average concentrations, between 1 and 3 pCi/l (see Table 7 Appendix), were very small fractions of the reference level.

## STRONTIUM-90 DEPOSITION

### 1. Routine Measurement

The measurement of strontium-90 deposition started in New Zealand in 1960, and in Suva in 1961. Since 1963 measurements have been made continuously at nine New Zealand stations, and since 1967 at two Pacific Islands. Collections are made each month in high walled stainless steel pots. The collected rainwater is passed through a column of cation exchange resin at the collecting site. The resin is then mailed to the Laboratory for measurement of strontium-90.

Annual depositions since 1960 have been averaged for the New Zealand stations and are listed in Table 3 below, together with the annual depositions at each station. (Individual monthly results during 1976 are given in Table 8 Appendix.)

TABLE 3 - Annual Deposition of Strontium-90 ( $\mu\text{Ci}/\text{km}^2$ ), Mean Annual Rainfall (cm)

	New Zealand Stations										Pacific Islands	
	KA	AK	NP	HN	WN	HK	CH	DN	IN	Average	SU	RA
1960		1.2		0.7	0.8	1.5	0.5		0.5	0.9		
1961		1.1		0.8	1.1	2.2	0.7		1.2	1.2	1.0	
1962		1.8		1.0	1.8	2.8	0.7		1.2	1.6	1.6	
1963	1.8	2.0	2.0	1.0	2.0	3.7	1.2	1.0	1.7	1.8	2.4	
1964	4.1	4.0	5.3	1.6	3.4	7.8	1.3	1.8	3.0	3.6	2.5	
1965	3.1	2.9	4.2	1.7	3.9	5.9	1.7	2.0	2.8	3.1	2.0	
1966	1.6	1.3	1.9	0.8	1.6	2.2	0.7	0.7	1.1	1.3	1.2	
1967	1.0	0.9	1.3	0.5	1.0	1.7	0.4	0.6	0.9	0.9	0.8	(0.9)
1968	0.9	0.7	1.0	0.6	0.9	1.4	0.4	0.4	0.5	0.8	1.0	0.7
1969	1.5	1.3	1.5	0.7	1.1	2.2	0.7	0.7	1.2	1.2	1.3	0.7
1970	1.0	0.9	1.2	0.6	1.2	2.1	0.5	0.5	0.7	1.0	0.9	1.0
1971	2.0	1.3	1.9	1.0	1.2	2.5	0.7	0.8	1.1	1.4	(1.5)	(0.9)
1972	0.9	0.7	0.9	0.5	0.8	1.8	0.4	0.6	0.9	0.8	0.9	0.8
1973	0.4	0.3	0.3	0.2	0.4	0.6	0.2	0.2	0.3	0.3	0.4	0.6
1974	0.3	0.2	0.3	0.2	0.3	0.5	0.2	0.2	0.2	0.3	0.3	0.3
1975	0.3	0.2	0.3	0.2	0.3	0.6	0.2	0.2	0.3	0.3	0.2	0.1
1976	0.1	0.1	0.1	<0.1	0.2	0.2	<0.1	<0.1	<0.1	0.1	0.1	0.1

Rainfall	138	117	149	78	134	241	60	63	104		310	209
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- Notes: 1. The New Zealand stations are: Kaitiia, Auckland, New Plymouth, Havelock North, Wellington, Hokitika, Christchurch, Dunedin, and Invercargill.  
The Pacific Island stations are: Suva (Fiji), and Rarotonga.
2. The station in Westland (HK) was at Greymouth from 1960-1975 inclusive and was resited at Hokitika starting Jan. 1976.
3. Values in parenthesis are estimates.
4. The mean annual rainfall is for 1963 to 1976 inclusive (at Rarotonga for 1967-1976). At Westland annual rainfalls at the actual collecting sites were used to obtain the mean.

The large scale Northern Hemisphere (U.S.S.R.) and Pacific area (U.S.A.) nuclear tests, which were conducted in 1961 and 1962 before the signing of the Partial Test Ban Treaty, resulted in a delayed stratospheric fallout over New Zealand. The maximum deposition from these tests was recorded in late 1964 and early 1965. Thereafter the annual deposition steadily decreased until 1968.

During the years 1966 to 1974 a series of smaller scale nuclear tests were conducted by France in the South Pacific each year except 1969. Each series, lasting from one to three months and comprising from three to eight nuclear explosions, has taken place during the Southern Hemisphere winter. During these tests a total of 41 nuclear devices were reported to have been exploded in the atmosphere, most of them being in the low to medium power (kiloton) range. However, megaton explosions were reported to have occurred twice in 1968, twice in 1970, and once in 1971 (2). As a result the annual deposition of strontium-90 in New Zealand increased during the period 1969 to 1971 reaching a maximum in 1971 which, however, was less than one-half of the 1964 maximum. Since then the annual depositions have again decreased to reach, during 1976, the lowest values recorded since these measurements were started.

Estimates of the French nuclear tests' contribution to the total strontium-90 deposition in New Zealand, and also comparisons of New Zealand and Northern Hemisphere depositions were made in earlier reports (1).

Unlike the deposition of fresh fission products from the troposphere after the French atmospheric tests, the long term strontium-90 deposition, which includes a

significant stratospheric component, has shown no latitude dependence within New Zealand. The deposition, however, is rainfall dependent and high rainfall areas such as Greymouth or Hokitika in Westland have shown elevated values compared to low rainfall areas such as Christchurch on the east coast. When normalised for rainfall the mean annual deposition at the New Zealand stations, during 1963-1975 inclusive, was 1.08 mCi/km<sup>2</sup> per 100 cm of rain (standard deviation 0.06 mCi/km<sup>2</sup>). However, at Suva during 1974, the year of maximum deposition of stratospheric fallout in New Zealand, the deposition was lower than the New Zealand average, despite the higher rainfall at Suva. This demonstrates a characteristic feature of stratospheric fallout, namely that the tropics receive less stratospheric fallout than mid-latitudes. Since then, during the past ten years depositions at the two Pacific Islands have been similar to the average depositions in New Zealand. During the period of French Pacific nuclear testing it seems that the smaller stratospheric component and larger tropospheric component of the fallout at the Pacific Islands resulted in depositions similar to those in New Zealand where the relative contributions of these components were reversed.

## 2. Cumulative Deposition

Direct measurement of strontium-90 in soil to determine the cumulative deposition has been made at selected sites since 1953. The most recent routine measurements were made on soils sampled at the end of 1972. Results for this and all previous measurements were tabulated in the 1973 Annual Report (1). Sampling depth was initially 15 cm in 1953. The depth was increased to 20 cm in 1960 and 30 cm in 1970. Such direct measurements frequently give lower values than those obtained by summing annual fallout increments. It was considered that surface run-off of rainwater and leaching of part of the strontium-90 below the sampling depth could account for the lower values.

During 1976 a special survey to investigate the penetration of strontium-90 (and caesium-137) in soil at selected sites was completed (3). The object was to relate the findings to the ion exchange properties of the soils and to the known levels of milk contamination in the same districts. The soils used in the survey had been sampled at the end of 1974 to a depth of 75 cm at Northland, Taranaki, Wellington, Greymouth, and South Canterbury. The total strontium-90 depositions measured at these stations were 19, 25, 16, 31, and 9 mCi/km<sup>2</sup> respectively. It was found that strontium-90 had penetrated below 60 cm at three of these stations. However, at least 60% was still in the top 15 cm, and at least 75% was in the top 30 cm at each station.

## LEAD-210 DEPOSITION

Lead-210 is a naturally-occurring radionuclide produced in the atmosphere by decay of gaseous radon which is exhaled from land surfaces. Like strontium-90 the subsequent deposition of lead-210 is rainfall dependent and high rainfall areas such as Hokitika show elevated values compared to low rainfall areas such as Christchurch.

Measurement of lead-210 deposition was continued during 1976. Evaluation was made in the same monthly rainwater samples in which strontium-90 was determined. The individual results for 1976 are listed in Table 9 Appendix. Earlier results, including higher levels during 1965 at four stations, and also levels in milk,

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- (3) "Profiles of <sup>90</sup>Sr and <sup>137</sup>Cs Concentrations in Selected New Zealand Soils and Their Bearing on Milk Contamination Levels." T. Baltakmens and L. P. Gregory. N.Z. Journal of Science (in press September 1977).

were discussed in the 1971 annual report (1). Annual depositions since 1967 have been averaged for the New Zealand stations and are listed in Table 4 below, together with the annual depositions at each station.

TABLE 4 - Annual Deposition of Lead-210 (mCi/km<sup>2</sup>)

	<u>New Zealand Stations</u>										<u>Pacific Islands</u>	
	KA	AK	NP	HN	WN	HK	CH	DN	IN	<u>Average</u>	SU	RA
1967	0.62	1.15	1.72	0.73	1.02	2.38	0.36	0.56	0.99	1.06	1.25	0.60
1968	1.75	1.64	2.08	0.84	1.86	3.20	0.64	0.76	1.06	1.54	2.46	0.61
1969	1.83	1.33	1.54	0.88	1.20	3.94	0.56	0.92	1.29	1.50	1.91	0.98
1970	1.43	1.00	1.82	0.63	1.52	3.09	0.65	0.74	0.92	1.31	1.85	0.85
1971	2.07	1.04	0.96	0.65	1.26	2.34	0.52	0.74	1.09	1.19	1.83	-
1972	2.28	1.62	1.99	0.88	1.70	3.41	0.70	1.22	1.31	1.68	2.65	-
1973	1.92	1.42	2.29	0.80	1.80	3.31	0.48	0.67	0.81	1.50	2.11	-
1974	1.24	1.08	1.76	0.76	1.61	2.85	0.71	0.66	0.58	1.25	1.86	4.00
1975	1.61	1.51	1.81	1.12	1.97	3.94	0.92	0.91	1.11	1.66	2.91	1.40
1976	1.41	1.33	1.43	0.93	1.46	2.73	0.70	0.71	0.76	1.27	2.03	-

Notes: 1. See Table 3 (Notes 1 and 2) for station names and the resiting of the Westland station at Hokitika.  
 2. The 1967 results are for May-Dec. only.  
 3. Estimates have been made for some missing monthly deposition results in order to reduce the resulting bias in the annual total. Where this has not been practicable annual totals are omitted.

During the last nine years the annual deposition in New Zealand has averaged about 1.4 mCi/km<sup>2</sup>. During the same period the annual deposition of weapons test strontium-90 at the same stations has averaged about 0.7 mCi/km<sup>2</sup>. At the Pacific Island stations lead-210 depositions at Suva are generally somewhat higher, and at Rarotonga generally somewhat lower, than those in New Zealand.

Since 1967, there does not appear to have been any marked seasonal variation nor, unlike strontium-90, any significant change in the average annual deposition.

#### STRONTIUM-90 AND CAESIUM-137 IN MILK

Strontium-90 measurement in New Zealand milk started in 1961 and caesium-137 measurement in 1964. Since 1965 continuous measurements have been made in samples from nine collecting stations. Caesium-137 and potassium are determined directly in monthly samples by gamma spectroscopy. Samples are then aggregated quarterly for radiochemical analysis for strontium-90 and also for the determination of calcium.

##### 1. Strontium-90

The all-station average concentrations each year since 1961 are listed in Table 5 below, together with average levels for each station. (Individual quarterly results during 1976 are listed in Table 10 Appendix). The average concentration during 1976, 2.6 pCi/gCa, was the lowest recorded since measurements commenced.

TABLE 5 - Strontium-90 in Milk - Annual Averages (pCi/gCa)

	ND	AK	WK	TA	PN	WN	WD	CH	DN	Average
1961	4.5		4.1	7.1			12.7	1.6		6.0
1962	6.3	5.5	4.9	9.4	4.3		13.5	2.1	3.0	6.1
1963	7.5	5.3	5.6	9.9	4.9		17.2	2.7	3.7	7.1
1964	11.2	9.1	9.5	17.1	7.1		26.0	2.6	4.1	10.8
1965	10.6	9.4	9.8	16.7	8.4	8.8	28.8	4.3	7.4	11.6
1966	6.5	6.1	6.3	12.5	4.8	6.1	22.7	2.4	4.0	7.9
1967	5.1	5.2	5.0	10.4	3.9	5.4	17.8	1.9	3.1	6.4
1968	4.1	3.8	4.1	8.0	3.6	4.8	14.0	1.6	2.4	5.2
1969	6.3	6.0	5.4	9.4	5.8	5.1	17.9	1.7	3.0	6.7
1970	5.2	5.1	5.2	9.7	3.6	4.7	21.0	2.2	2.5	6.6
1971	7.3	5.8	6.0	10.2	5.0	4.8	18.3	2.0	3.0	6.9
1972	4.8	4.6	4.4	8.2	5.0	4.1	14.7	1.9	3.1	5.6
1973	3.8	3.4	3.5	5.7	2.7	3.5	10.8	1.2	1.9	4.1
1974	3.3	3.0	2.7	5.4	2.5	3.0	8.8	1.3	1.9	3.5
1975	3.1	2.7	3.0	5.1	2.4	3.4	8.7	1.2	1.6	3.5
1976	2.6	2.4	2.5	3.5	1.6	2.4	6.1	1.1	1.1	2.6
Average	5.8	5.2	5.1	9.3	4.4	4.7	16.2	2.0	3.1	6.3

Note: The stations are: Northland, Auckland, Waikato, Taranaki, Palmerston North, Wellington, Westland, Christchurch, and Dunedin.

Average levels in New Zealand milk reached maximum values of 10.8 and 11.6 pCi/gCa during 1964 and 1965 when the rate of strontium-90 deposition was also a maximum. Milk levels then fell steadily reaching a minimum of 5.2 pCi/gCa in 1968, about half the 1964-65 maximum. This indicated that the level in milk was dependent to a considerable extent on fallout rate. However, during the period 1965-68, milk levels decreased at a slower rate than the strontium-90 deposition, thus indicating in addition, some uptake by grass of the cumulative deposit in the soil. After the start of French Pacific nuclear tests in 1966, milk levels increased slightly during the period 1969-1971. However, following the subsequent decrease in deposition since 1972, milk levels also decreased again, reaching the minimum level recorded in 1976.

Milk samples from the lowest and highest rainfall stations, i.e. Christchurch and Westland, give the range of strontium-90 contamination in New Zealand milk. Generally the extent of this range is from about one-third to nearly three times the country-wide average.

## 2. Caesium-137

The all-station average concentrations each year since 1964 are listed in Table 6 below, together with average levels for each station. (Individual monthly results during 1976 are listed in Table 11 Appendix.) The average concentration during 1976, 6 pCi/gK, was the lowest recorded since measurements commenced. Again the highest levels were recorded in 1964 and 1965 and they have steadily decreased since then except for a slight increase in 1969 and 1970. The higher levels at Taranaki due to the "soil effect" have been discussed in the 1971 annual report (1), and this effect has been the subject of further investigation (3).

TABLE 6 - Caesium-137 in Milk - Annual Averages (pCi/gK)

	ND	AK	WK	TA	PN	WN	WD	CH	DN	<u>Average</u>
1964	49	51	69	168	19		76	7	11	56
1965	54	53	84	185	26	29	77	11	18	60
1966	37	33	60	141	11	18	43	4	9	39
1967	26	26	48	123	7	13	33	3	5	31
1968	15	18	36	102	3	7	21	1	3	23
1969	27	26	41	101	5	9	38	2	4	28
1970	22	18	35	89	6	11	39	4	5	25
1971	23	18	36	80	7	9	30	3	5	23
1972	21	15	28	72	2	7	22	2	4	19
1973	14	9	21	49	3	4	14	1	2	13
1974	7	7	16	41	2	3	8	1	1	10
1975	9	7	14	34	1	3	8	1	1	9
1976	6	5	11	23	2	2	4	1	2	6
Average	24	22	38	93	7	10	32	3	5	26

Note: See Table 5 for station names.

### 3. Comparison of Measured Levels with the Reference Levels

When comparing measured levels in milk with the reference levels, long-term averages are more meaningful. Since measurements commenced, the "country-wide" average levels of strontium-90 (6.3 pCi/gCa) and caesium-137 (26 pCi/gK), have been 2.3% and 0.4% of the reference levels respectively. At the stations with the highest levels of contamination the corresponding percentages are about three times higher.

Thus the long-term average levels, even at the stations with highest concentrations, are very small fractions of the reference levels and do not constitute a public health hazard.

## MISCELLANEOUS

### 1. Monitoring During Visits of Nuclear Powered Ships

The environmental radioactivity section of the Laboratory participated in the special monitoring of harbour environs during the visit of the USS TRUXTON to Wellington, and the USS LONGBEACH to Auckland in 1976. Pre-visit seawater samples were collected and then regular sampling downstream of the ships was undertaken at each high tide. Sampling of bottom sediments within the swinging distance of the ships' anchorage, and filter feeding molluscs from selected sites was undertaken before the ships' arrival and again after departure. All samples were airmailed to the Laboratory where they were evaluated by gamma spectroscopy. Measurements on the pre-visit samples established the background levels of radioactivity. All samples collected during and after each visit showed only those traces of natural radioactivity, and at the same levels, as were measured in the pre-visit samples.

Air sampling was conducted continuously during each visit at selected sites around the harbours. Sampling was by means of pumps each drawing 17 cubic metres of air per day through a special cartridge. The cartridges were made at the Laboratory from 22 mm diameter plastic cylinders containing a glass-fibre pre-filter for trapping particulates followed by a 35 mm bed of activated charcoal for trapping radioiodines. During routine monitoring, cartridges were changed each day and airmailed to the Laboratory for evaluation. Iodine-131 was below the limit of detection in all cartridges. The limit of detection was less than 1% of the dose limit for continuous exposure over one year for critical groups in the population,

as set by the International Commission on Radiological Protection.

A full report on these monitoring operations has been published (4).

## 2. International Intercomparison

During 1976 the Laboratory again participated successfully in the intercomparison of measurements on environmental samples, and also for the first time on the testing of tentative radiochemical procedures:

The International Reference Centre, WHO, provided a milk sample for measurement of strontium-90, caesium-137, calcium, and potassium; and also a mineral water sample for measurement of total beta activity, potassium, radium-226, and natural uranium.

The U.S. Environmental Protection Agency provided on three occasions milk samples for measurement of strontium-89, strontium-90, iodine-131, caesium-137, barium-140, and potassium, and also a polonium-210 standard solution for a round robin study. Three Tentative Reference Methods: "Total Alpha and Total Beta Radioactivity in Waters", "Total Radium and Radium-226 in Waters", and "Strontium-89 and Strontium-90 in Waters", were provided by the Agency together with radioactive standards and water samples for testing these procedures with the objective of raising their status to Standard Reference Methods.

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- (4) "Report on Radioactive Monitoring During the Visits of Nuclear Powered Ships." National Radiation Laboratory, Department of Health, New Zealand, Report NPS-1, 22 February 1977.

TABLE 7 - Total Beta Activity of Weekly Rainwater Samples 1976 : Deposition ( $\text{mCi}/\text{km}^2$ ), Rainfall (mm)

The collection period is from the date shown to the start of the next collection.  
N.S. No sample or result available, ( ) estimated result.

AUCKLAND			WELLINGTON			HOKITIKA			CHRISTCHURCH			FIJI			SAMOA			TONGA			AITUTAKI			RAROTONGA		
Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>
Jan 2	0.6	<0.1	Jan 2	0.4	<0.1	Jan 2	0.4	<0.1	Jan 2	0.4	<0.1	Dec 29	3.2	<0.1	Dec 28	12.7	<0.1	Jan 2	3.3	<0.1	Dec 29	0.3	<0.1	Jan 2	4.8	<0.1
Jan 9	17.2	0.2	Jan 9	3.2	0.1	Jan 9	9.4	0.1	Jan 9	4.0	<0.1	Jan 7	8.8	0.1	Jan 8	21.0	<0.1	Jan 9	3.4	<0.1	Jan 5	2.2	<0.1	Jan 9	8.7	0.1
Jan 16	<0.1	<0.1	Jan 16	0.5	<0.1	Jan 16	5.1	0.1	Jan 16	<0.1	<0.1	Jan 14	5.6	<0.1	Jan 16	27.5	<0.1	Jan 16	13.3	0.2	Jan 12	7.7	<0.1	Jan 16	2.2	<0.1
Jan 23	0.3	<0.1	Jan 23	6.2	0.2	Jan 23	15.1	N.S.	Jan 23	1.4	<0.1	Jan 21	13.1	0.3	Jan 23	3.9	<0.1	Jan 23	3.1	<0.1	Jan 19	0.4	<0.1	Jan 23	8.3	<0.1
																		Jan 26	1.6	<0.1						
Jan	18.1	0.2	Jan	10.3	0.4	Jan	30.0	(0.3)	Jan	5.8	0.1	Jan	30.7	0.5	Jan	65.1	0.1	Jan	23.1	0.2	Jan	12.2	<0.1	Jan	24.0	0.2
Jan 30	0.3	<0.1	Jan 30	0.2	<0.1	Jan 30	1.7	N.S.	Jan 30	0.6	<0.1	Feb 10	5.0	<0.1	Jan 30	9.3	<0.1	Jan 30	2.3	0.1	Feb 2	22.3	0.2	Jan 30	10.9	0.1
Feb 6	4.9	0.2	Feb 6	5.0	0.1	Feb 6	1.1	<0.1	Feb 5	3.5	<0.1	Feb 18	4.6	0.1	Feb 6	7.2	<0.1	Feb 6	5.3	<0.1	Feb 8	4.8	<0.1	Feb 6	23.5	<0.1
Feb 13	0.3	<0.1	Feb 13	1.1	<0.1	Feb 13	5.8	0.3	Feb 13	0.1	<0.1	Feb 25	2.7	<0.1	Feb 13	20.2	0.2	Feb 13	9.8	0.2	Feb 16	4.2	<0.1	Feb 13	1.0	<0.1
Feb 20	<0.1	<0.1	Feb 20	0.5	<0.1	Feb 20	1.2	<0.1	Feb 20	1.6	<0.1				Feb 20	4.1	<0.1	Feb 20	4.7	<0.1	Feb 23	2.3	0.1	Feb 20	2.5	<0.1
															Feb 25	5.9	<0.1	Feb 27	15.2	<0.1						
Feb	5.5	0.2	Feb	6.8	0.2	Feb	9.8	0.4	Feb	5.8	0.2	Feb	12.3	0.1	Feb	46.7	0.3	Feb	37.3	0.4	Feb	33.6	0.4	Feb	37.9	0.3
Feb 27	nil	<0.1	Feb 27	<0.1	<0.1	Feb 27	0.2	<0.1	Feb 27	0.5	<0.1	Mar 3	11.9	0.1	Mar 5	7.1	0.2	Mar 1	8.5	<0.1	Mar 1	0.4	<0.1	Feb 27	0.1	<0.1
Mar 5	1.0	<0.1	Mar 5	0.2	<0.1	Mar 5	7.4	0.1	Mar 5	<0.1	<0.1	Mar 10	2.8	<0.1	Mar 12	2.6	0.1	Mar 5	5.2	<0.1	Mar 8	1.3	<0.1	Mar 5	0.9	0.1
Mar 12	<0.1	<0.1	Mar 12	<0.1	<0.1	Mar 12	nil	<0.1	Mar 12	nil	<0.1	Mar 17	5.2	<0.1	Mar 19	1.0	<0.1	Mar 12	5.6	<0.1	Mar 15	1.3	<0.1	Mar 12	3.5	<0.1
Mar 19	<0.1	<0.1	Mar 19	<0.1	<0.1	Mar 19	4.3	<0.1	Mar 19	nil	<0.1	Mar 23	0.8	<0.1	Mar 26	7.9	0.1	Mar 19	3.2	<0.1	Mar 22	1.9	<0.1	Mar 20	12.4	<0.1
Mar 26	2.5	<0.1	Mar 26	5.0	N.S.	Mar 26	2.5	<0.1	Mar 26	1.1	0.1							Mar 26	0.3	<0.1						
Mar	3.4	0.2	Mar	5.3	(0.1)	Mar	14.4	0.3	Mar	1.6	0.1	Mar	20.7	0.2	Mar	18.6	0.4	Mar	22.8	0.1	Mar	4.9	0.2	Mar	16.9	0.2
Apr 2	0.7	<0.1	Apr 2	1.1	<0.1	Apr 2	3.7	<0.1	Apr 2	1.9	<0.1	Mar 31	0.8	<0.1	Apr 2	1.2	<0.1	Apr 2	5.1	<0.1	Mar 29	1.5	<0.1	Mar 26	14.9	(0.1)
Apr 9	6.8	0.2	Apr 9	4.8	0.1	Apr 9	<0.1	<0.1	Apr 9	0.6	<0.1	Apr 7	7.6	0.1	Apr 9	1.7	<0.1	Apr 9	27.1	0.4	Apr 5	21.3	<0.1	Apr 16	10.9	<0.1
Apr 16	0.4	<0.1	Apr 16	<0.1	<0.1	Apr 16	<0.1	<0.1	Apr 15	<0.1	<0.1	Apr 13	11.3	<0.1	Apr 16	0.9	<0.1	Apr 16	4.4	<0.1	Apr 12	3.9	<0.1	Apr 23	13.1	0.3
Apr 23	5.3	<0.1	Apr 23	3.6	0.1	Apr 23	8.6	0.1	Apr 23	<0.1	<0.1	Apr 21	0.8	0.2	Apr 23	16.6	0.1	Apr 23	0.3	0.1	Apr 19	12.7	0.3			
Apr	13.2	0.3	Apr	9.6	0.4	Apr	12.4	0.2	Apr	2.6	<0.1	Apr	20.5	0.5	Apr	20.4	0.2	Apr	36.9	0.6	Apr	39.4	0.5	Apr	38.9	0.5
Apr 30	3.2	0.1	Apr 30	1.6	0.1	Apr 30	4.5	0.1	Apr 30	<0.1	<0.1	Apr 28	1.7	<0.1	Apr 30	6.1	<0.1	Apr 30	7.7	0.2	Apr 29	0.9	<0.1	Apr 30	5.8	0.1
May 7	2.7	<0.1	May 7	6.0	0.2	May 7	4.9	<0.1	May 7	1.4	<0.1	May 5	<0.1	<0.1	May 7	2.5	<0.1	May 7	0.4	<0.1	May 10	13.1	<0.1	May 7	1.5	<0.1
May 14	1.2	<0.1	May 14	0.9	<0.1	May 14	12.6	0.2	May 14	1.5	<0.1	May 12	nil	0.1	May 14	13.8	0.2	May 14	0.6	0.1	May 16	6.1	<0.1	May 14	2.4	<0.1
May 21	2.8	<0.1	May 21	0.3	<0.1	May 21	4.3	<0.1	May 21	0.8	<0.1	May 19	0.6	<0.1	May 21	3.6	<0.1	May 21	<0.1	<0.1	May 24	4.4	0.2	May 21	0.5	0.1
May 28	0.8	<0.1	May 28	0.2	<0.1	May 28	6.7	0.3	May 28	0.3	<0.1	May 26	nil	<0.1	May 28	2.0	<0.1	May 28	0.8	<0.1						
May	10.7	0.3	May	9.0	0.4	May	33.0	0.8	May	4.0	0.1	May	2.4	0.2	May	28.0	0.4	May	9.5	0.5	May	24.5	0.3	May	10.2	0.2
Jun 4	3.3	<0.1	Jun 4	4.2	0.1	Jun 4	9.5	0.1	Jun 4	0.5	<0.1	Jun 3	2.5	0.2	Jun 4	0.9	<0.1	Jun 4	4.2	0.1	May 31	3.8	<0.1	May 28	25.3	<0.1
Jun 11	6.8	<0.1	Jun 11	2.9	<0.1	Jun 11	0.9	<0.1	Jun 11	0.3	<0.1	Jun 15	1.0	<0.1	Jun 11	12.1	<0.1	Jun 11	0.4	<0.1	Jun 7	1.4	<0.1	Jun 18	1.8	<0.1
Jun 18	2.8	<0.1	Jun 18	6.5	0.1	Jun 18	0.9	<0.1	Jun 18	5.3	<0.1	Jun 25	nil	<0.1	Jun 18	1.2	<0.1	Jun 18	0.1	<0.1	Jun 14	1.5	<0.1	Jun 25	4.7	<0.1
Jun 25	3.8	<0.1	Jun 25	1.1	<0.1	Jun 25	8.1	0.1	Jun 25	0.3	<0.1				Jun 25	0.5	<0.1	Jun 25	0.2	<0.1	Jun 21	0.8	<0.1			
Jun	16.7	0.2	Jun	14.7	0.3	Jun	19.4	0.3	Jun	6.4	<0.1	Jun	3.5	0.2	Jun	14.7	0.1	Jun	4.9	0.2	Jun	7.5	0.1	Jun	31.8	<0.1

APPENDIX



TABLE 7 (continued)

AUCKLAND			WELLINGTON			HOKITIKA			CHRISTCHURCH			FIJI			SAMOA			TONGA			AITUTAKI			RAROTONGA			
Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	Date	cm	mCi/km <sup>2</sup>	
Jul 2	10.5	<0.1	Jul 2	2.3	<0.1	Jul 2	0.7	<0.1	Jul 2	1.9	<0.1	Jun 30	nil	<0.1	Jul 2	3.2	0.2	Jul 2	0.3	<0.1	Jun 28	1.4	0.1	Jul 2	0.8	0.1	
Jul 9	1.9	<0.1	Jul 9	7.1	N.S.	Jul 9	10.1	0.2	Jul 9	2.4	<0.1	Jul 8	nil	0.3	Jul 9	8.1	<0.1	Jul 9	2.0	<0.1	Jul 6	0.2	<0.1	Jul 9	-	N.S.	
Jul 16	1.6	<0.1	Jul 16	7.6	<0.1	Jul 16	5.3	<0.1	Jul 16	1.5	0.1	Jul 16	0.6	<0.1	Jul 16	6.3	<0.1	Jul 16	1.8	<0.1	Jul 14	0.6	0.3	Jul 16	7.2	0.2	
Jul 23	1.9	<0.1	Jul 23	0.9	<0.1	Jul 23	6.9	0.2	Jul 23	<0.1	<0.1	Jul 26	1.1	<0.1				Jul 23	2.0	0.4	Jul 19	4.5	<0.1	Jul 23	4.3	<0.1	
																		Jul 26	2.4	<0.1							
Jul	15.9	0.2	Jul	17.9	0.2	Jul	23.0	0.4	Jul	5.8	0.3	Jul	1.7	0.4	Jul	17.6	0.3	Jul	6.1	0.6	Jul	9.1	0.5	Jul	12.3	0.3	
Jul 30	3.2	0.1	Jul 30	4.1	<0.1	Jul 30	4.6	<0.1	Jul 30	1.7	<0.1	Aug 2	3.2	<0.1	Jul 29	0.7	<0.1	Jul 30	1.6	<0.1	Aug 2	0.5	0.2	Jul 30	2.9	<0.1	
Aug 6	3.3	<0.1	Aug 6	1.8	<0.1	Aug 6	0.9	<0.1	Aug 6	1.4	<0.1	Aug 10	0.3	<0.1	Aug 6	0.2	0.1	Aug 6	1.1	<0.1	Aug 9	<0.1	<0.1	Aug 6	<0.1	<0.1	
Aug 13	2.1	<0.1	Aug 13	4.5	<0.1	Aug 13	2.0	<0.1	Aug 13	0.4	<0.1	Aug 18	5.5	<0.1	Aug 13	0.4	<0.1	Aug 13	<0.1	<0.1	Aug 16	<0.1	<0.1	Aug 13	<0.1	<0.1	
Aug 20	0.5	<0.1	Aug 20	2.3	<0.1	Aug 20	8.2	0.2	Aug 20	2.2	0.1	Aug 25	nil	<0.1	Aug 20	nil	<0.1	Aug 20	3.1	0.1	Aug 23	2.4	<0.1	Aug 20	11.7	<0.1	
Aug 27	0.8	<0.1	Aug 27	4.1	<0.1	Aug 27	nil	<0.1	Aug 27	2.6	<0.1				Aug 27	2.7	<0.1	Aug 27	1.3	<0.1				Aug 27	4.4	<0.1	
Aug	9.9	0.2	Aug	16.8	0.3	Aug	15.7	0.3	Aug	8.3	0.2	Aug	9.0	<0.1	Aug	4.0	0.3	Aug	7.1	0.2	Aug	3.0	0.4	Aug	19.0	0.2	
Sep 3	8.6	<0.1	Sep 3	1.0	<0.1	Sep 3	0.5	<0.1	Sep 3	2.7	<0.1	Sep 2	16.5	<0.1	Sep 3	<0.1	<0.1	Sep 3	16.5	<0.1	Aug 30	1.2	<0.1	Sep 2	5.4	<0.1	
Sep 9	3.8	<0.1	Sep 10	3.9	<0.1	Sep 10	9.6	<0.1	Sep 10	3.7	<0.1	Sep 7	2.8	<0.1	Sep 10	<0.1	<0.1	Sep 10	nil	<0.1	Sep 6	0.1	<0.1	Sep 10	0.6	<0.1	
Sep 17	0.2	<0.1	Sep 17	0.1	<0.1	Sep 17	1.1	<0.1	Sep 17	<0.1	<0.1	Sep 16	0.3	<0.1	Sep 17	0.1	<0.1	Sep 17	2.7	<0.1	Sep 13	0.3	<0.1	Sep 17	1.4	<0.1	
Sep 24	1.3	<0.1	Sep 24	4.8	<0.1	Sep 24	0.8	<0.1	Sep 24	0.1	<0.1	Sep 22	-	N.S.	Sep 24	0.3	<0.1	Sep 24	5.8	<0.1	Sep 20	3.9	<0.1				
Sep	13.9	0.2	Sep	9.8	0.1	Sep	12.0	<0.1	Sep	6.6	0.1	Sep	19.6	<0.1	Sep	0.4	0.1	Sep	25.0	0.2	Sep	5.5	0.2	Sep	7.4	<0.1	
Oct 1	0.4	<0.1	Oct 1	1.9	<0.1	Oct 1	5.2	<0.1	Oct 1	0.5	<0.1	Sep 30	3.2	<0.1	Oct 1	<0.1	0.2	Oct 1	1.3	<0.1	Oct 4	0.7	<0.1	Oct 1	3.2	0.3	
Oct 8	-	N.S.	Oct 8	1.9	<0.1	Oct 8	7.0	0.1	Oct 8	1.1	<0.1	Oct 6	nil	<0.1	Oct 8	0.8	0.1	Oct 8	0.1	0.1	Oct 11	0.1	<0.1	Oct 8	<0.1	<0.1	
Oct 15	3.7	<0.1	Oct 15	1.9	0.1	Oct 15	8.5	<0.1	Oct 15	2.6	0.1	Oct 13	nil	<0.1	Oct 15	<0.1	<0.1	Oct 15	1.9	<0.1	Oct 18	<0.1	0.1	Oct 15	2.6	0.4	
Oct 22	1.3	0.1	Oct 22	1.3	0.1	Oct 22	4.9	<0.1	Oct 22	1.2	<0.1	Oct 21	1.6	<0.1	Oct 22	0.1	<0.1	Oct 22	6.8	<0.1	Oct 25	8.4	0.2	Oct 22	2.0	<0.1	
Oct	5.4	0.2	Oct	7.0	0.3	Oct	25.6	0.3	Oct	5.4	0.2	Oct	4.8	<0.1	Oct	0.9	0.3	Oct	10.1	0.2	Oct	9.2	0.4	Oct	7.8	0.7	
Oct 29	0.2	<0.1	Oct 29	0.2	<0.1	Oct 29	3.1	<0.1	Oct 29	0.4	<0.1	Oct 28	2.3	0.1	Oct 29	6.6	<0.1	Oct 29	6.9	<0.1	Nov 1	3.7	<0.1	Nov 3	7.7	<0.1	
Nov 5	0.3	<0.1	Nov 5	2.5	<0.1	Nov 5	8.9	0.1	Nov 5	0.7	<0.1	Nov 3	<0.1	<0.1	Nov 5	3.0	0.1	Nov 5	0.7	<0.1	Nov 8	nil	<0.1	Nov 5	6.7	<0.1	
Nov 12	-	N.S.	Nov 12	-	N.S.	Nov 12	<0.1	<0.1	Nov 12	1.4	0.2	Nov 10	7.0	0.1	Nov 12	0.3	<0.1	Nov 12	6.7	0.2	Nov 15	0.4	0.3	Nov 12	4.2	<0.1	
Nov 19	1.5	0.1	Nov 19	0.9	<0.1	Nov 19	1.5	<0.1	Nov 19	0.8	<0.1	Nov 18	nil	0.2	Nov 19	6.1	0.2	Nov 19	8.6	0.3	Nov 22	18.9	0.3	Nov 22	1.7	0.2	
Nov 26	6.6	0.1	Nov 26	2.8	<0.1	Nov 26	6.4	0.1	Nov 26	1.5	0.1	Nov 24	2.2	0.2				Nov 26	7.9	0.2							
Nov	8.6	0.3	Nov	6.4	0.3	Nov	20.0	0.4	Nov	4.8	0.3	Nov	11.5	0.6	Nov	22.0	0.4	Nov	30.8	0.8	Nov	23.0	0.7	Nov	20.3	0.3	
Dec 3	1.3	<0.1	Dec 3	1.0	<0.1	Dec 3	15.2	0.3	Dec 3	1.9	<0.1	Dec 1	1.4	<0.1	Nov 29	18.9	<0.1	Dec 1	0.3	0.1	Nov 29	18.0	0.1	Dec 3	1.9	<0.1	
Dec 10	1.1	<0.1	Dec 10	1.9	<0.1	Dec 10	2.7	<0.1	Dec 10	0.9	<0.1	Dec 8	nil	<0.1	Dec 7	29.4	<0.1	Dec 10	<0.1	<0.1	Dec 6	-	N.S.	Dec 12			
Dec 17	6.7	<0.1	Dec 17	22.4	N.S.	Dec 17	3.1	<0.1	Dec 17	3.7	<0.1	Dec 15	1.0	<0.1	Dec 11	1.9	<0.1	Dec 17	1.6	<0.1	Dec 13	-	N.S.	to			
Dec 24	1.6	<0.1	Dec 24	1.4	<0.1	Dec 24	19.4	0.2	Dec 24	0.5	<0.1	Dec 22	2.3	<0.1	Dec 17	3.0	<0.1	Dec 25	0.3	<0.1	Dec 20	-	N.S.	Jan 5	18.9	<0.1	
															Dec 24	7.0	<0.1										
Dec	10.7	<0.1	Dec	26.7	<0.1	Dec	40.4	0.6	Dec	7.0	<0.1	Dec	4.7	<0.1	Dec	60.2	0.1	Dec	2.2	0.1	Dec	18.0	0.1	Dec	20.8	<0.1	
TOTAL 132	2.5		TOTAL 140	3.0		TOTAL 256	4.3		TOTAL 64	1.7		TOTAL 141	2.8		TOTAL 299	3.0		TOTAL 216	4.1		TOTAL 190	3.8		TOTAL 247	3.0		
Average Concentration (pCi/l)																											
2			2			2			3			2			1			2			2			1			

TABLE 8 - Strontium-90 in Rain 1976 : Rainfall (cm), Deposition ( $\mu\text{Ci}/\text{cm}^2$ ), Concentration (pCi/l).

Station		Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Total	Av.
Kaitia	Rainfall	32.5	2.4	1.8	20.1	6.5	13.0	15.2	14.7	9.4	13.0	13.0	10.2	152	
	Deposition	0.02	0.01	<0.01	0.01	<0.01	0.02	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	0.11	
	Concentration	<0.1	0.4	0.3	<0.1	0.1	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1		<0.1
Auckland	Rainfall	18.5	5.2	3.4	14.0	8.4	15.7	17.9	9.1	14.0	6.4	11.5	10.6	135	
	Deposition	0.01	0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	0.01	<0.01	<0.01	<0.01	0.10	
	Concentration	<0.1	0.2	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1		<0.1
New Plymouth	Rainfall	12.8	10.7	7.8	15.9	12.3	19.1	16.4	12.8	8.4	11.7	14.1	12.0	154	
	Deposition	0.02	0.02	<0.01	0.01	<0.01	0.01	<0.01	0.02	0.01	<0.01	<0.01	<0.01	0.13	
	Concentration	0.1	0.2	<0.1	<0.1	<0.1	<0.1	<0.1	0.1	0.1	<0.1	<0.1	<0.1		<0.1
Havelock North	Rainfall	10.2	21.5	4.1	5.2	0.8	3.8	7.4	10.4	12.6	7.3	5.1	8.6	97	
	Deposition	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	0.07	
	Concentration	<0.1	<0.1	0.1	<0.1	0.5	<0.1	<0.1	0.1	<0.1	<0.1	<0.1	<0.1		<0.1
Wellington	Rainfall	11.0	5.6	8.2	7.5	9.4	21.9	20.8	21.3	12.9	11.5	7.0	42.0	179	
	Deposition	0.01	0.02	0.01	<0.01	<0.01	0.01	0.01	0.02	<0.01	0.03	<0.01	0.01	0.15	
	Concentration	0.1	0.3	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.3	<0.1	<0.1		<0.1
Hokitika	Rainfall	30.0	9.2	13.4	14.0	30.0	23.5	24.5	15.3	12.1	26.5	15.2	45.8	260	
	Deposition	0.02	0.02	0.01	<0.01	0.01	0.02	0.02	0.02	<0.01	0.01	<0.01	0.03	0.19	
	Concentration	<0.1	0.2	0.1	<0.1	<0.1	<0.1	<0.1	0.1	<0.1	<0.1	<0.1	<0.1		<0.1
Christchurch	Rainfall	8.8	5.8	1.6	2.6	3.7	6.7	5.9	8.1	6.6	5.5	4.2	7.5	67	
	Deposition	<0.01	0.01	<0.01	0.01	<0.01	<0.01	0.01	0.02	<0.01	<0.01	<0.01	<0.01	0.09	
	Concentration	<0.1	0.2	0.3	0.2	0.2	<0.1	0.2	0.2	0.1	<0.1	<0.1	<0.1		0.1
Dunedin	Rainfall	2.5	4.0	0.8	2.5	6.8	10.1	4.1	6.8	3.2	5.1	3.4	12.6	62	
	Deposition	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	0.01	0.08	
	Concentration	0.2	0.2	0.6	0.2	<0.1	<0.1	0.2	0.2	0.2	0.1	0.2	0.1		0.1
Invercargill	Rainfall	9.5	2.9	5.7	6.2	14.3	16.0	11.9	4.0	1.5	7.3	6.4	7.9	94	
	Deposition	0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	0.09	
	Concentration	0.1	0.3	0.1	<0.1	<0.1	<0.1	<0.1	0.3	<0.1	<0.1	<0.1	0.1		<0.1
New Zealand Country-wide Average	Rainfall	15.1	7.5	5.2	9.8	10.2	14.4	13.8	11.4	9.0	10.5	8.9	17.5	133	
	Deposition	0.01	0.01	<0.01	<0.01	<0.01	0.01	0.01	0.01	<0.01	<0.01	<0.01	0.01	0.11	
	Concentration	0.1	0.2	0.2	<0.1	0.1	<0.1	<0.1	0.1	<0.1	<0.1	<0.1	<0.1		<0.1
Suva, Fiji	Rainfall	29.8	34.6	28.0	17.1	51.1	17.6	13.7	21.1	31.8	23.0	34.0	45.1	347	
	Deposition	<0.01	<0.01	<0.01	<0.01	0.01	0.01	0.01	0.02	0.01	<0.01	<0.01	0.01	0.12	
	Concentration	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.1	<0.1	<0.1	<0.1	<0.1		<0.1
Rarotonga	Rainfall	24.3	37.9	18.4	37.1	15.1	26.6	4.3	15.6	7.4	6.1	25.4	18.8	237	
	Deposition	0.01	0.01	<0.01*	<0.01	<0.01	0.01	<0.01	0.01	<0.01	<0.01	0.04	0.02	0.12	
	Concentration	<0.1	<0.1	<0.1 *	<0.1	<0.1	<0.1	0.2	<0.1	<0.1	<0.1	0.1	<0.1		<0.1

\* 2-monthly collection

# APPENDIX

**TABLE 9 - Lead-210 in Rain, 1976 : Deposition (mCi/km<sup>2</sup>)**

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Total
Kaitaia	0.28	0.04	0.03	0.13	0.06	0.09	0.12	0.05	0.08	0.22	0.18	0.13	1.41
Auckland	0.17	0.05	0.05	0.09	0.16	0.11	0.13	0.15	0.08	0.10	0.11	(0.13)	1.33
N. Plymouth	0.12	0.11	0.08	0.12	0.09	0.15	0.12	0.14	0.08	0.16	(0.13)	0.13	1.43
Havelock N.	0.11	0.10	0.05	0.05	0.03	0.05	0.04	0.12	0.13	0.10	0.10	0.05	0.93
Wellington	0.17	0.06	0.10	0.05	0.16	0.24	0.11	0.15	0.09	0.15	0.04	0.14	1.46
Hokitika	0.22	0.10	0.16	0.20	0.29	0.27	0.20	0.20	0.09	0.37	0.12	0.51	2.73
Christchurch	0.06	0.06	0.04	0.06	0.05	0.06	0.05	0.08	0.08	0.07	(0.04)	0.05	0.70
Dunedin	0.04	0.05	0.03	0.09	0.07	0.07	0.03	0.08	0.03	0.08	0.03	0.11	0.71
Invercargill	0.06	0.05	0.05	0.12	0.12	0.09	0.06	0.04	0.02	0.04	0.03	0.08	0.76
NZ Average	0.14	0.07	0.07	0.10	0.11	0.13	0.10	0.11	0.08	0.14	0.09	0.15	1.27
Suva	0.11	0.08	0.12	0.08	0.19	0.14	0.17	0.15	0.25	0.32	0.27	0.15	2.03
Rarotonga	0.13	0.17	0.24*	0.08	0.10	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.	0.40	-

N.S. No result available. ( ) Estimate \* 2-monthly collection

**TABLE 10 - Strontium-90 in Milk, 1976 : (pCi/g Ca)**

	1st Quarter	2nd Quarter	3rd Quarter	4th Quarter	Average
Northland	2.9	2.0	3.0	2.4	2.6
Auckland	2.5	2.3	2.7	2.0	2.4
Waikato	2.3	2.5	3.0	2.0	2.5
Taranaki	2.9	3.6	3.6	3.8	3.5
Palmerston North	1.6	1.3	2.1	1.5	1.6
Wellington	2.3	2.1	2.9	2.2	2.4
Westland	6.5	5.7	6.1	5.9	6.1
Christchurch	1.1	0.9	1.2	1.0	1.1
Dunedin	1.0	1.0	1.3	1.0	1.1
NZ Average	2.6	2.4	2.9	2.4	2.6

**TABLE 11 - Caesium-137 in Milk, 1976 : (pCi/g K)**

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Av.
Northland	10	8	5	5	3	5	5	4	4	3	10	6	6
Auckland	7	7	6	5	4	2	<1	4	3	3	10	4	5
Waikato	14	18	15	15	16	N.S.	5	6	6	7	13	8	11
Taranaki	30	23	22	42	29	24	10	13	18	20	25	24	23
Palmerston North	2	3	<1	<1	4	N.S.	<1	9	1	<1	<1	<1	2
Wellington	1	2	<1	1	3	<1	2	<1	<1	7	1	<1	2
Westland	8	5	6	4	3	3	2	1	4	11	3	3	4
Christchurch	<1	3	<1	1	<1	1	<1	<1	<1	<1	<1	3	1
Dunedin	1	5	<1	<1	<1	<1	5	<1	<1	6	6	3	2
NZ Average	8	8	6	8	7	5	3	4	4	6	8	6	6

N.S. No sample.

PART IV

RECENT PUBLICATIONS RELATED TO ENVIRONMENTAL POLLUTION

## Recent Publications Related to Environmental Pollution

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