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GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION

SOME ASPECTS OF FALLOUT FROM THE FIRST AND SECOND CHINESE TESTS

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ABSTRACT

The pattern of movement of fallout debris clouds after the first and second Chinese tests has been studied with the help of wind data at 200 mb level. Samples of fallout debris collected at high altitudes with the help of commercial aircraft have been studied for freshly produced fission products and detailed analysis has been carried out for several shortlived isotopes. Details of analysis used for rapid identification of fissile materials after the two tests are presented. Activity ratios of important shortlived fission products for both of the tests had shown U-235 fission. The results of analysis for Np-239 and U-237 are also given. These analyses provide evidence of production of U-237 by double neutron capture and of Np-239 by neutron activation of U-238. The airborne radioactivity found at Gulmarg after the second Chinese test has been discussed.

SOME ASPECTS OF FALLOUT FROM THE FIRST AND SECOND CHINESE TESTS

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1. Movement of the fallout debris clouds from the first and second Chinese tests

The first and second Chinese tests were conducted on October 16, 1964 and May 14, 1965, respectively. The first test was carried out near Lop Nor in the Sinkiang Province (approximate latitude of 40°N and longitude 90°E) from a tower several hundred feet high (1). The second test was carried out over the remote Western Provinces of China (2). The main radioactive clouds from these tests were, most likely, confined to the troposphere moving rapidly eastwards at an altitude range of 30,000 to 50,000 feet. The wind data was available for the 200 mb level (approx. 40,000 ft.) which is regularly collected in connection with jet aircraft flights. With the help of this data it was possible to trace the initial trajectory of the clouds from these tests. Figure 1 gives the approximate trajectory of the fallout debris cloud from the second Chinese test.

Strong westerlies prevailed at 200 mb level at the time of both of the tests. The wind speed at this level at the time of the first test was around 80 to 100 knots and at the

time of the second test it was in the range of 50 to 70 knots. A remarkable feature of the circulation at this level is a large longitudinal expanse of the westerlies, particularly around 40°N latitude. This gave rise to the rapid circling around the globe of the fallout debris clouds from the two tests.

2. Detection of fresh fallout

Samples of radioactive debris at high altitudes were collected between latitude band of 20° to 50°N, with the help of commercial aircraft. The sampling corridor covered the regions between Tokyo and Bombay, and New York and Bombay. The collections of radioactive particulate matter were made by impaction on surfaces of known area below the wings of the aircraft. The impaction surfaces were thoroughly decontaminated prior to each collection. The samples were obtained by wiping the impaction surface carefully several times with sterlized cotton wool soaked in organic detergents. The flying altitude of the aircraft ranged from 20,000 to 35,000 feet mainly flying between 30° to 50°N latitude.

The samples were collected during October 1964 to December 1964, in connection with the first test and during May 1965 to August 1965, in connection with the second test. The sampling was continued in the intervening period in order to establish the background of old fallout.

Since the sample collection was made merely by impaction during normal flights, these samples did not provide the actual air concentrations of different radionuclides but were found to be extremely useful in providing relative concentrations of the different fission products, induced radioactivities, plutonium, and uranium.

The samples collected before each of the tests showed large concentrations of old fallout debris mainly consisting of Cs-137, Sr-90, Mn-54, Ru-106, Ce-144, Sb-125, Pu-239 and U-238. A 256-channel pulse height analyser was used for rapid scanning of the samples for gamma emitters to detect the presence of shortlived fission products from fresh fallout. The prominent La-140 gamma peak at 1.6 Mev provided most positive indication of fresh fallout as the gamma spectra of old fallout did not show any significant background in this energy region.

Rapid indication of fresh fallout was provided by the samples collected on the commercial flights between Tokyo and Bombay. The fresh fallout from the second Chinese test was first detected from the sample on an aircraft left Tokyo in the morning of May 18. The sample from a similar flight on the previous day did not show any fresh fission products and it was thus deduced that a part of the cloud must have

reached Tokyo late on May 17. The cloud trajectory plotted from the wind data showed the arrival of the radioactivity over northern parts of Japan late on May 16 and the time difference between the arrival of radioactivity in the upper atmosphere over northern Japan and near Tokyo was due to the lateral dispersion of the cloud. Subsequently, fresh fallout was detected in a sample from New York-Bombay flight leaving London on May 26. Based on similar observations after the first Chinese test it was found that whereas it took 12 to 13 days for the cloud to complete the circle around the globe after the first test, the time taken for a complete circle around the globe after the second test was 17 to 18 days.

3. Identification of fissile material

The important shortlived isotopes studied after these tests are Ag-111, I-131, Ba-140, Ce-141, Sr-89 and Zr-95. The longest lived of these isotopes is Zr-95 with a half-life of 65 days. The concentrations of these isotopes in the samples collected before the tests were negligibly small. The relative concentrations of these isotopes were used for finding the type of fissile material used in the new tests. Table 1 gives the activity ratios of important shortlived fission products for Pu-239 and U-235 fission(3).

The last two columns in the table give the observed ratios in the samples from the first and second Chinese tests respectively. These ratios show that the fissile material used in both of these tests was U-235. It is seen from this table that a few of the ratios are very sensitive to the mode of fission. For example, the ratio Ag-111/Sr-89 is 23 times larger for Pu-239 fission than for U-235 fission. Similarly, the ratios I-131/Sr-89 and Ag-111/Ba-140 also show variations by factors of 5 to 15 for the two modes of fission.

The isotopes used for the identification of fissile material were chemically separated after leaching the cotton swipe samples with a mixture of 3M hydrochloric acid and 0.1M hydrofluoric acid. Radiochemical purity of the isotopes was checked by energy measurements and by following the decay. It was also found that great reliance could be placed on carefully planned chemical analysis for rapid identification of the fissile material. After the first detection of fresh activity in a sample, it requires only 10 to 12 hours for the identification of the fissile material by this procedure.

4. Measurement of Np-239 and U-237

The samples were also analysed for Np-239 and U-237

produced by the interaction of neutrons with uranium used in the weapon. Figures 2 and 3 give the decay curves of Np-239 and U-237 respectively, from a typical analysis after the second test. The actual contents of Np-239 and U-237 have been indicated in the figures along with Ba-140 content of the sample for comparison.

Np-239 is produced by the following reaction:

$$\text{U-238}(n,\gamma)\ \text{U-239} \xrightarrow{\beta} \text{Np-239}$$

The neutron capture cross section for the above reaction is fairly large and therefore, the production of Np-239 has been used for estimating the U-238 corresponding to a known number of fissions in the sample. The actual number of fissions in a sample can be estimated from the known concentration and fission yield of a typical fission product Ba-140. Knowing the number of fissions and the fission cross section the number of atoms of U-235 is estimated and compared with the number of atoms of U-238 estimated from Np-239. These estimates on typical samples after the second Chinese test showed that Np-239 could have been produced only from U-238 fraction of enriched uranium.

U-237 can be produced by the following two reactions in an atomic weapon using U-235 as fissile material.

$$\text{U-235}\ (n,\gamma)\quad \text{U-236}\ (n,\gamma)\ \text{U-237}$$

$$\text{U-238}\ (n,2n)\quad \text{U-237}$$

The second reaction depends on fast neutron flux, having a threshold at 6 Mev and maximum cross section at 10 Mev. In the fission neutron spectrum the neutrons in the energy range of 6 to 10 Mev are a very small fraction of the total number of neutrons. Therefore, production of U-237 by this reaction would be small. The most likely mode of production of U-237 detected in the samples is double neutron capture. The production of U-237 by this mode has been calculated knowing the fission and capture cross sections of U-235 and U-236 (Appendix). The values of these cross sections are known fairly accurately from recent literature (4,5). The percentage of fissions giving rise to the formation of U-237 has been calculated and found to be 2.4 per cent which can be used in the same manner as fission yield for calculating the production of U-237. The concentration of U-237 has been compared with the concentration of Ba-140 in a typical sample after the second Chinese test. This sample gave the activity ratio of Ba-140/U-237 = 1.87. This ratio calculated from the fission yield of Ba-140 for U-235 fission and the yield of U-237 by double neutron capture is 1.27. The two ratios show a fairly good agreement.

5. Fallout in India

There are 9 ground-level fallout sampling stations in

India and one in Sikkim State as given in Table 2. Samples are also collected at high altitudes within the country using filter samplers mounted on Canberra aircraft. These tests did not give any significant fallout in India because the testing sites were located towards the north of eastern India and the initial radioactive clouds at high altitude rapidly moved eastwards as discussed earlier. However, small concentrations of fresh fission products were recorded at Gulmarg, the northernmost station in India, within 40 to 50 hours after the second test, caused by an anti-cyclonic movement in upper winds above the region of test, as seen in the trajectory shown in Figure 1. This increase persisted only for 3 to 4 days. The second marked increase in activity was again recorded at Gulmarg on May 31, 1965 i.e. 17 days after the test which indicated the arrival of the cloud over the northern parts of India after completing a circle around the globe. Traces of fresh fission products were also detected in the high altitude samples collected over the central parts of India after May 31, 1965, which showed that the radioactive cloud had become fairly widespread. Figure 4 gives the variations in the level of fission product beta activity at Gulmarg for the period May 13 to June 15, 1965.

Acknowledgments

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Table-1

Activity Ratios of Important Shortlived Fission Products

for First and Second Chinese Tests

Isotope Pairs	Ratio for Pu-239 fission	Ratio for U-235 fission	Observed Ratios First Test	Observed Ratios Second Test	Remarks
Ag-111 / Ba-140	0.163	0.018	0.023	0.010	All the ratios correspond to U-235 fission. Variations in the ratios are likely to be due to isotopic fractionation effects.
Sr-89 / Ag-111	0.45	10.42	8.62	14.74	
Ag-111 / Zr-95	0.80	0.076	0.083	0.043	
Ce-141 / Sr-89	5.10	2.02	-	2.99	
Ba-140 / Sr-89	13.52	5.50	5.04	6.67	
I-131 / Zr-95	7.70	3.75	2.75	-	

Table-2

List of Ground-Level Fallout Sampling Stations

	Station	Altitude (Meters)	Location
1	Bangalore	922	12° 57'N, 77° 30'E
2	Bombay	Sea Level	18° 57'N, 72° 55'E
3	Calcutta	Sea Level	22° 34'N, 88° 25'E
4	Delhi	219	28° 45'N, 77° 20'E
5	Gangtok(Sikkim State)	2000	27° 12'N, 88° 23'E
6	Gulmarg	2743	34° 00'N, 74° 25'E
7	Nagpur	311	21° 12'N, 79° 04'E
8	Naini Tal	1935	29° 30'N, 79° 30'E
9	Ootacamund	2235	11° 23'N, 76° 40'E
10	Srinagar	1598	34° 06'N, 74° 55'E

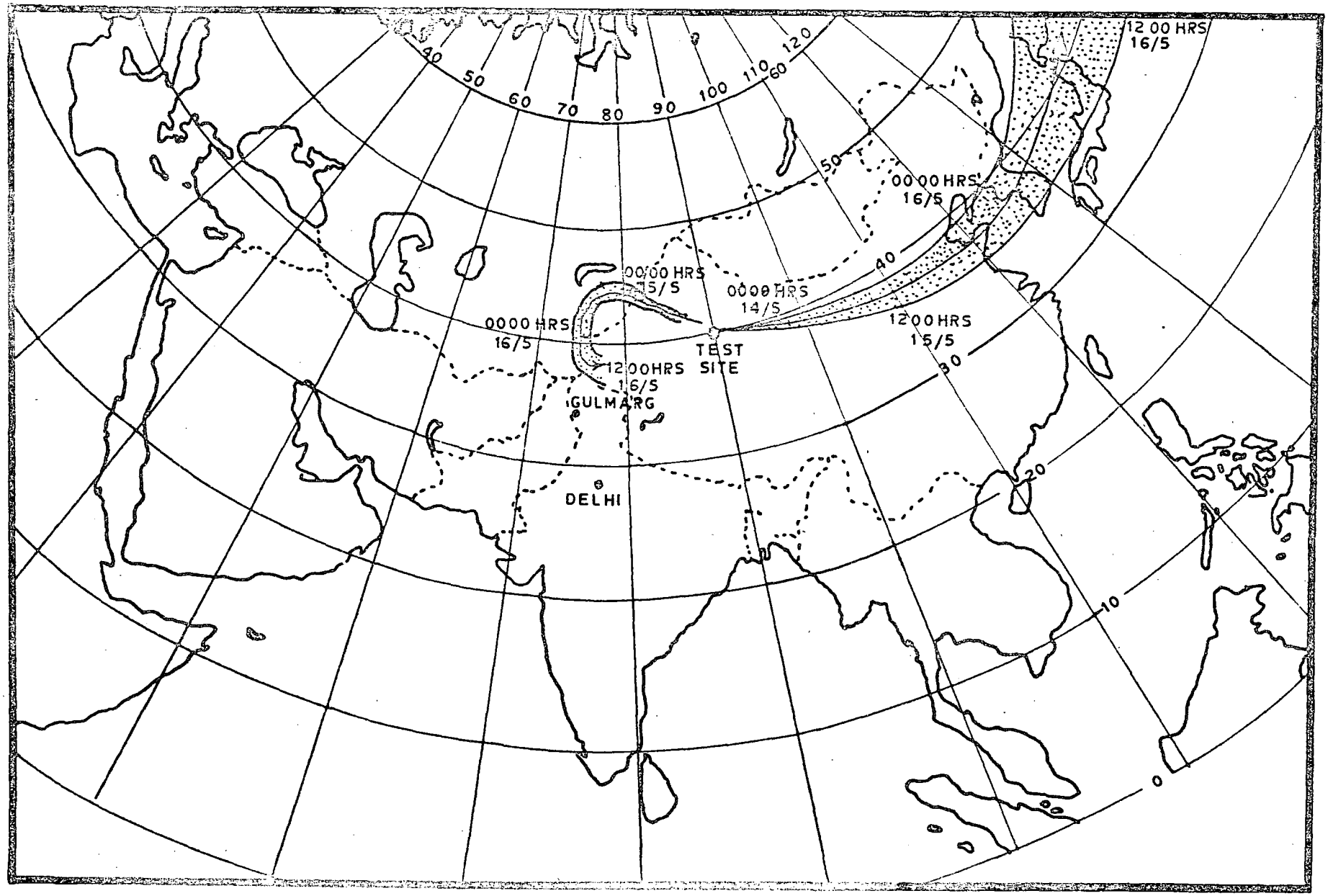

FIGURE-1. TRAJECTORY OF WINDS AT 200MB (40,000FT) FOLLOWING THE SECOND CHINESE TEST AND APPROXIMATE FALLOUT DEBRIS PATTERN.

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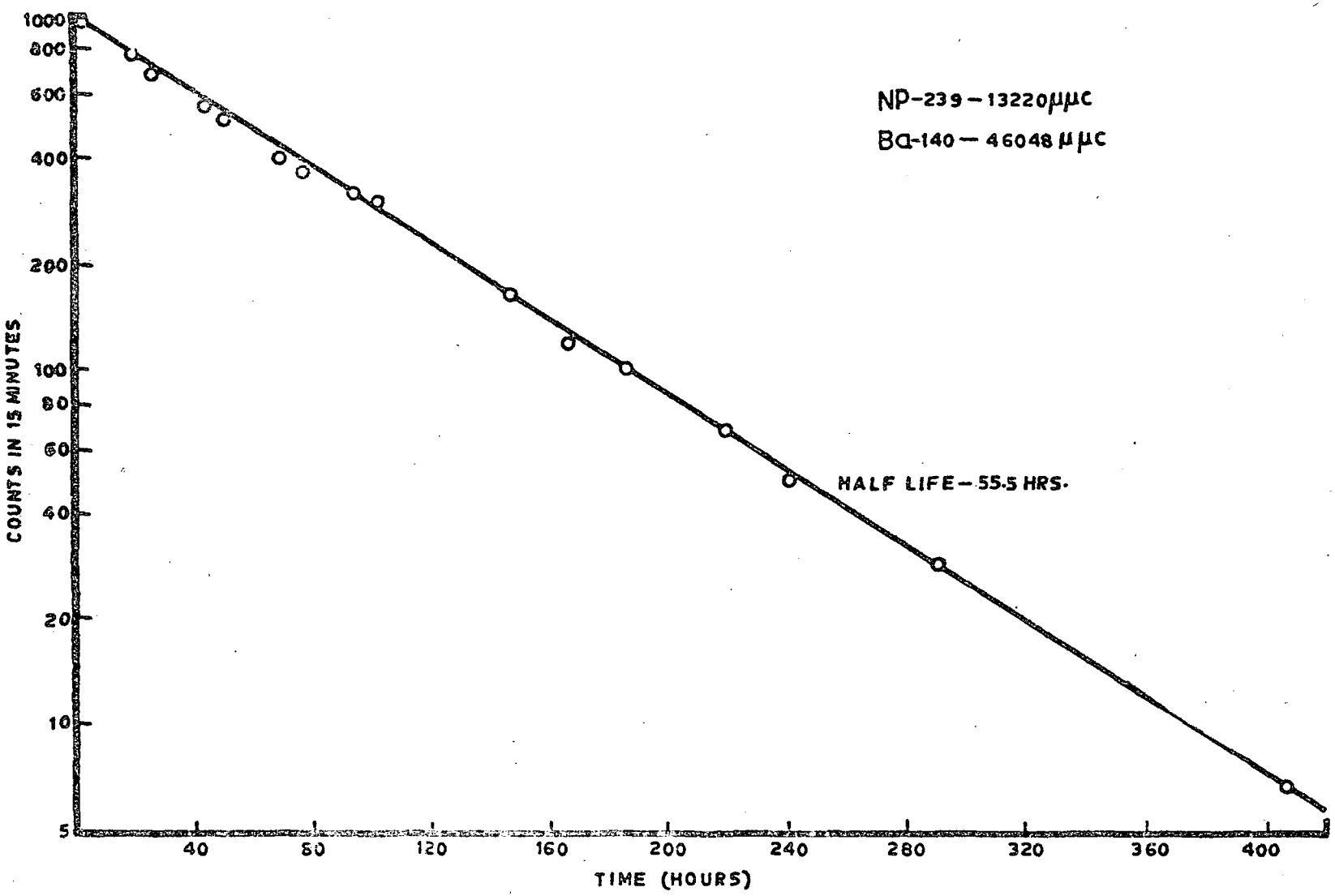

FIGURE-2. DECAY CURVE OF NEPTUNIUM-239 SEPARATED FROM FALLOUT DEBRIS COLLECTED AFTER THE SECOND CHINESE TEST. DATE OF COLLECTION 18.5.1965.

E-5041-G-107

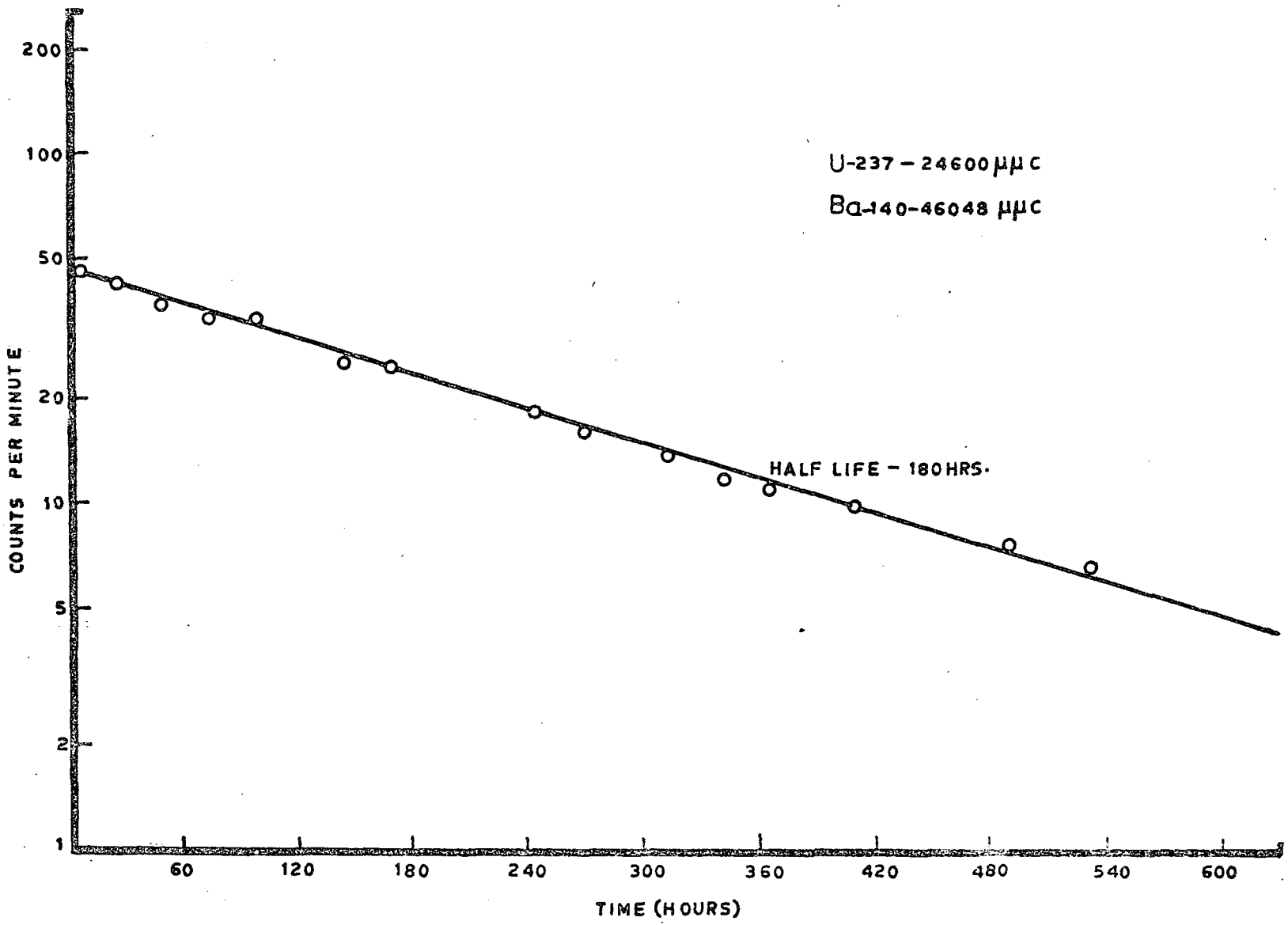

FIGURE-3. DECAY CURVE OF URANIUM-237 SEPARATED FROM FALLOUT DEBRIS COLLECTED AFTER THE SECOND CHINESE TEST. DATE OF COLLECTION 18.5.1965.

E-5041-G-110

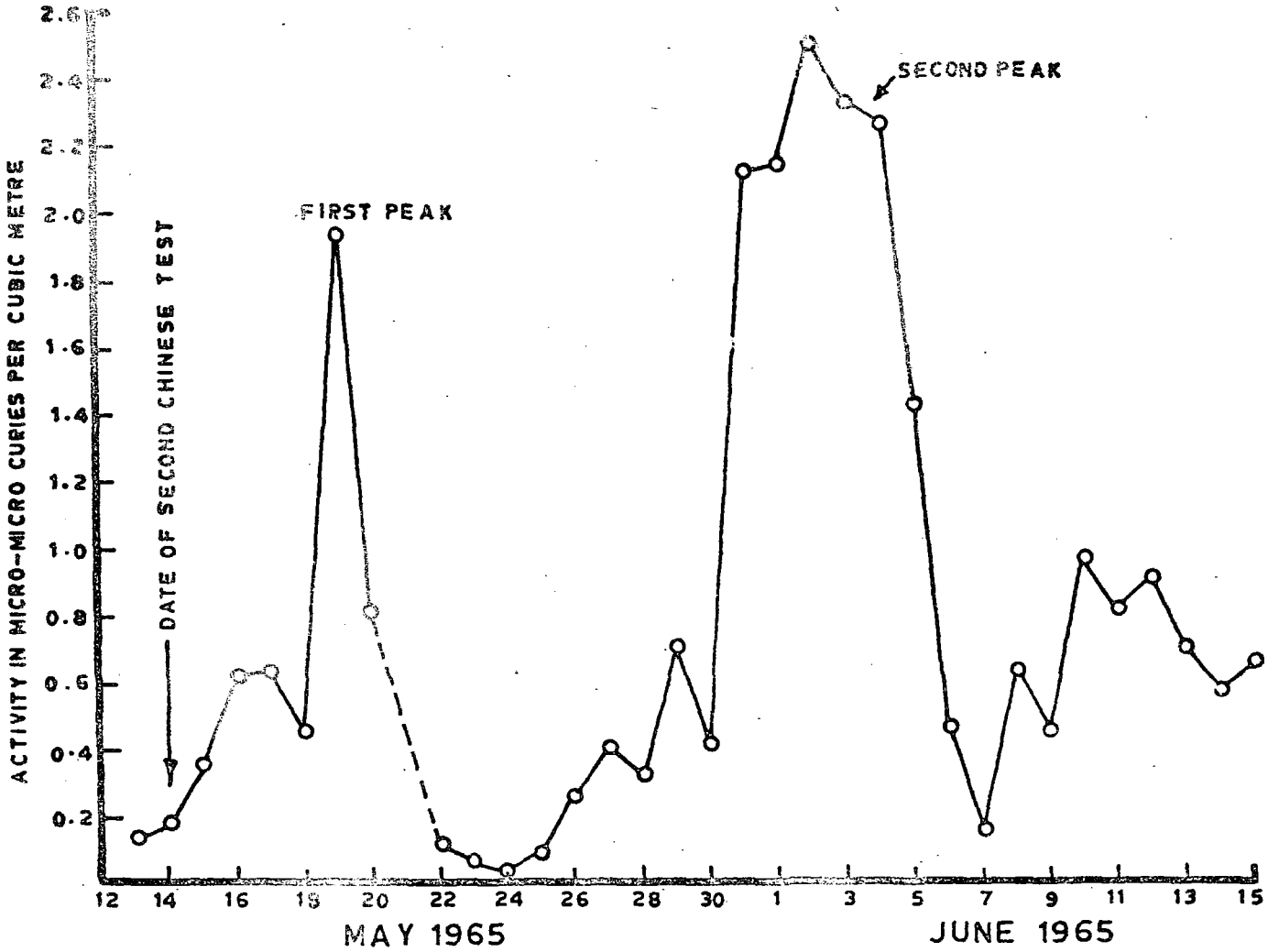

FIGURE-4. FISSION PRODUCT RADIOACTIVITY OF SURFACE AIR AT GULMARG.

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A P P E N D I X

Theoretical Estimates of U-237 Formation by Double Neutron Capture in the case of Nominal Yield Atomic Weapons Using U-235

There are two likely modes of U-237 formation in the case of a nuclear detonation using U-235 as fissile material.

1. U-235 (n, γ) U-236 (n, γ) U-237
2. U-238 (n, 2n) U-237

The second reaction has a threshold at 6 MeV and the cross-section is maximum at 10 MeV. Therefore, for fission spectrum neutrons of nominal yield explosions the production by second reaction is negligibly small. Also, if highly enriched uranium is used, the production is reduced further due to small quantity of U-238 available. However, in the case of megaton weapons where neutron energy has a peak around 14 MeV and large quantity of U-238 is used in third stage of the bomb, the second reaction is the main contributor to U-237 production.

For calculating U-237 by first reaction let us assume

σ_f^5 = fission cross-section of U-235

σ_c^5 = capture cross-section of U-235

σ_T^6 = total cross-section of U-236

σ_c^6 = capture cross-section of U-236

$$\alpha = \frac{\sigma_c^5}{\sigma_f^5}$$

A = number of U-235 atoms initially present

N = number of U-236 atoms formed

R = U-237 fraction of total number of fissions

T = total time of interaction ($\sim$ 0.5 μ. sec)

τ = average time between successive neutron generations ($\sim 6 \times 10^{-9}$ sec)

k = multiplication factor (2 to 2.8)

n_o = initial number of fissions which can be assumed as 1

Then, the number of fissions at any time t is given by

$$n_f(t) = n_o e^{\frac{(k-1)t}{\tau}} \qquad \ldots.(1)$$

In terms of constants for U-235

$$n_f(t) = A.\ \sigma_f^s\ .\ \Phi(t)$$

where $\Phi(t)$ is the flux of neutrons at any time t

Therefore,

$$\Phi(t) = \frac{n_f(t)}{A.\ \sigma_f^s} = \frac{n_o e^{\frac{(k-1)t}{\tau}}}{A.\ \sigma_f^s} \qquad \ldots.(2)$$

The rate of formation of U-236 atoms at any time t is given by

$$\frac{dN(t)}{dt} = A.\ \sigma_c^s\ .\ \Phi(t) = \frac{A.\ \sigma_c^s . n_o e^{\frac{(k-1)t}{\tau}}}{A.\ \sigma_f^s}$$

$$= \alpha . n_o e^{\frac{(k-1)t}{\tau}}$$

The number of U-236 atoms lost by fission or capture at any time t will be given by

$$\sigma_T^6 . N(t) . \Phi(t)$$

and net production of U-236 at time t is given by

$$\frac{dN(t)}{dt} = \alpha . n_o e^{\frac{(k-1)t}{\tau}} - \sigma_T^6 . N(t) . \Phi(t)$$

$$= n_o e^{\frac{(k-1)t}{\tau}} (\alpha - \beta N(t))$$

where $\beta = \frac{\sigma_T^6}{\sigma_f^5 \cdot A}$. If $\frac{k-1}{\tau} = g$ and $n_o = 1$

$$\frac{dN(t)}{dt} = e^{gt} (\alpha - \beta N(t))$$

Integrating,

$$\int_0^N \frac{dN(t)}{\alpha - \beta N(t)} = \int_0^T e^{gt} dt$$

or $N = \frac{\alpha}{\beta} \quad 1 - e^{-(\beta/g)(e^{gT} - 1)}$

$$= \frac{\alpha}{\beta} \text{ for } \quad T >> \tau$$

$$= \frac{\sigma_c^5}{\sigma_T^6} . A \qquad \ldots\ldots(3)$$

Similarly, the rate of formation of U-237 atoms at any time t is given by

$$\frac{dR(t)}{dt} = N(t) . \sigma_c^6 . \Phi(t)$$

$$= \frac{A . \sigma_c^5 . \sigma_c^6}{A . \sigma_f^5 . \sigma_T^6} . e^{\frac{(k-1)t}{\tau}}$$

Therefore, $R(T) = \int_0^{R(T)} dR(t) = \int_0^T e^{\frac{(k-1)t}{\tau}} . \frac{A . \sigma_c^5 . \sigma_c^6 . dt}{A . \sigma_f^5 . \sigma_T^6}$

where R(T) is the number of U-237 atoms formed in time T.

or $R(T) = \frac{\sigma_c^5 . \sigma_c^6}{\sigma_f^5 . \sigma_T^6} \int_0^T e^{\frac{(k-1)t}{\tau}} dt$

and since total number of fissions in time T assuming $n_o = 1$

are given by $\int_0^T e^{\frac{(k-1)t}{\tau}} dt$

The total number of U-237 atoms as fraction of number of fissions is given by

$$R = \frac{\sigma_c^5}{\sigma_f^5} \cdot \frac{\sigma_c^6}{\sigma_T^6} = \alpha_0 \frac{\sigma_c^6}{\sigma_T^6}$$

For fission spectrum neutrons

$\alpha = 0.08$, $\sigma_c^6 = 0.20b$, and $\sigma_T^6 = 0.67b$

Giving, $R = \frac{0.08 \times 0.2}{0.67} = \frac{0.016}{0.67} = 0.024$ (2.4%)

The activity ratio Ba-140/U-237 can be found knowing the fission yield of Ba-140 (5.8%) and half lives of Ba-140 (12.8 days) and U-237 (6.75 days) i.e.

$$\frac{\text{Ba-140}}{\text{U-237}} = \frac{5.8 \times 6.75}{2.4 \times 12.8} = 1.27$$