CANADA

GAMMA-RAY ANALYSIS OF ATMOSPHERIC DUST SAMPLES

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ABSTRACT

Gamma-ray spectra of dust samples from the atmosphere were plotted from measurements with a five-channel analyser. Peaks from fission products Zr-95, Nb-95, Ru-103, Ce-141, Ce-144 and Cs-137, with half-lives of several days and longer, were identified. The same peaks were found in a sample of house dust. Very long-lived fission products such as caesium-137 were not of sufficient activity to permit identification until the samples had decayed for several months.

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(22 pages, 1 Table, 11 Figs.)

INTRODUCTION

In course of flights over the Arctic islands of Canada, aerial studies by the Geological Survey of Canada showed indications of airborne radioactivity measurable with a portable scintillation counter. These areas of activity appeared to shift in conformity with the wind pattern. Thoron and radon are known to occur in the atmosphere. To determine whether any naturally radioactive emitters could be identified, Dr. L. W. Morley, Chief of the Geophysics Division of the Geological Survey, arranged to have dust samples collected in filters mounted in their aircraft. These samples were submitted to the Mines Branch for analysis by the gamma-ray spectrometer.

The first series of samples was collected, beginning in May 1957, from a Catalina aircraft during flights out from Churchill, Manitoba. In 1958 a second series of samples was collected in Eastern Canada, using an Aero Commander aircraft, during one flight from Ottawa to the nearby St. Lawrence Valley and in subsequent flights in the Gulf of St. Lawrence region. Records pertaining to the flights were kept for use in the event that there proved to be significant differences in the samples. Additional dust samples were collected from the ground for purposes of comparison.

The first dust sample, measured in May 1957, was sufficiently active to produce a clearly-defined spectrum; however, to minimize statistical fluctuations, several hours were required to determine the seventy-five spectral intervals, five channels at a time. The resulting

spectrum failed to match any portions of spectra obtained from the thorium or uranium series, or from potassium. Consequently, the question of fission products was considered in view of the series of nuclear explosions which had taken place in Siberia terminating in mid-April 1957, a few weeks before the first sample was collected[1].

In general, other investigations of fall-out activity have been limited to a measurement of the total beta activity; however, such fission products as strontium-89, 90, which emit only beta radiation, have been measured with considerable care in other laboratories after radiochemical concentration. The strontium activity was not detected in the following work which took into account gamma radiation only. A paper by Hunter and Ballou[2] provides a graph which is useful for estimating the relative proportions of the most active fission products remaining at the end of various intervals after fission. Considering the dates when the spectra were first determined (May 24, 29, 1957), and the known dates of the last previous atomic test explosions (up to mid-April, 1957[1]), it is reasonable to assume that at least 40 days had elapsed since the last atomic test. Since much of the activity in a sample was found to decay within a few months, the cumulative effect of tests of previous years was expected to be confined to a few very long-lived nuclides only. Attempts to measure the spectrum of several combined samples failed to show peaks due to caesium-137 (more strictly due to barium-137, the short-lived daughter of Cs-137), and other very long-lived fission products, until the activity of the samples had decayed for several months after collection.

EXPERIMENTAL DETAILS

The spectrometer contained a 2 in. by 2 in. cylindrical sodium iodide crystal as the scintillation detector. The crystal was mounted against a RCA type No. 6655 photomultiplier tube in a magnesium container. A thin aluminum foil window was cemented in that end of the container to which the sample was exposed. The scintillation counter was operated with a five-channel Marconi pulse-height analyser. The dust was collected in a cloth filter 4 in. by 8 in., folded once to 4 in. by 4 in. size. This was wrapped in paper with one thickness separating the filter from the aluminum window in the detector enclosure.

The method of collection employed during the series of flights in 1957 was less efficient than the one adopted during the following year. At that time a scoop directed air from outside the aircraft against or through the filter. In 1958 the filter was mounted directly on the nose of the aircraft. No attempt was made to cover the filter at take-off and on landing, so that a significant proportion of the dust may have been collected at these times.

A few attempts were made at sample concentration to increase the activity in the search for weak long-lived emitters and to produce a more stable sample geometry for half-life determinations. Filters were washed with chloroform which was then evaporated; other filters were dissolved in hot nitric acid; the simplest and most convenient method was to compact a number of fine filters in a thin aluminum sample dish with a tightly fitting cover, using a hydraulic press.

Each filter in the aircraft was exposed at a constant altitude of 1000 ft, during 3 or 4 flights of six hours duration at an airspeed of 180 mph. The dust picked up at take-off and upon landing was not expected to differ in the proportions of the active emitters present; however, to determine whether there was measurable activity, a ground sample was secured from the contents of a household vacuum cleaner bag in use during May and June 1958. The effects of the atom-bomb tests of March 1958 were quite evident, as the total gamma activity of the contents, measuring several rather bulky fractions, was in excess of 7000 counts per minute for a 2 lb sample.

DISCUSSION OF RESULTS

Table 1 shows the contribution to the total activity of the most active gamma emitters 40 to 320 days after fission, and also their principal gamma peaks. Other nuclides, such as strontium-89 and strontium-90, which are known to be produced in fission are not listed as they emit beta particles only. Figures 1 to 10 illustrate spectra of samples from different locations and at various stages of radioactive decay. The activity of the samples differed owing to factors such as exposure time, and meteorological conditions; however, all spectra at the same stage of decay were essentially the same despite the wide separation of the regions in this country where they were collected. Since there is evidence that most of the fall-out activity continues to circulate in latitudes close to that in which the test explosion took place[3], it is improbable that shorter-lived nuclides will re-appear in significant amounts from more recent tests in the Southern hemisphere.

TABLE 1

Radiations from Fission Products

Nuclide	Half life	Percentage of total activity [2] (β and γ), days after fission				Principal gamma-ray levels, keV [5]
		40	80	160	320	
Ce-141	33 d	12	9	4	-	142
Zr-95	65 d	10	14	15	9	723, 756
↓ Nb-95	35 d	7	16	25	18	765
	90 hr					236
Ba-140	13 d	9	3			30, 132, 160, 304, 436, 537
↓ La-140	40 hr	10	3			328(40), 490(50), 815(46), 1600(100) *
Ru-103	40 d	6	8	5	1	498(100), 610(7)
↓ Rh-103	57 min					
Nd-147	11 d	3				92, 530
↓ Pm-147	2.6 yr			1.5	5	121
Ce-144	285 d	3	5	10	23	33.7(7), 81(17), 134(29)
↓ Pr-144	17 min					695(12), 1480(2.3), 2180(5.9)
I-131	8.1 d	2				80(6), 280(6), 364(81), 637(9), 722(2.8)
Xe-133	5.3 d	1				81
Ru-106	1.0 yr				2	No gamma, parent of Rh-106
↓ Rh-106	30 sec					513(100), 624(53), 1045(8)
Cs-137	27 yr				1	No gamma, parent of Ba-137
↓ Ba-137	2.6 min					662

* (Nos. in parentheses indicate rel. intensity for each isotope)

Calibration was carried out in two principal energy ranges, 100 to 1000 keV and 20 to 200 keV, the former using Na-22, Cs-137 and Mn-54, and the latter Cs-137 (32 keV X-ray line) and Co-57, for comparison peaks. The standard calibration samples were separated from the detector sufficiently to have a count-rate of the same order as that from the dust sample, to minimize shift in gain with count-rate[4]. Because of the number of fission products present, each peak was usually composed of more than one unresolved gamma-ray, a factor which added greatly to the problem of identification. A recent list of principal gamma-ray energies has been prepared by Smith and Farmelo[5].

Figure 1 presents the complete gamma-ray spectrum of a typical dust sample above 100 keV, with a comparison of peak counts at three successive dates. Two main peaks of energies 500 and 740 keV are clearly evident. The widths of the two peaks are greater than would be obtained from a monoenergetic gamma-ray in the same energy region; namely, the 662 keV line from Cs-137, for which 8% resolution was obtained. The rate of decay of the 500 keV peak appeared to be more rapid initially. This might be attributed to 13-day Ba-140 emitting at 436 and 537 keV; additional evidence is obtained in Figure 10, where there is an early decay in the 160 keV region of the 1958 samples. Most of the activity is, however, due to 40-day Ru-103 at 498 keV; this peak was evident for about one year.

The other major peak is 65-day Zr-95 (723, 756 keV) in combination with its daughter element, 35-day Nb-95 (765 keV).

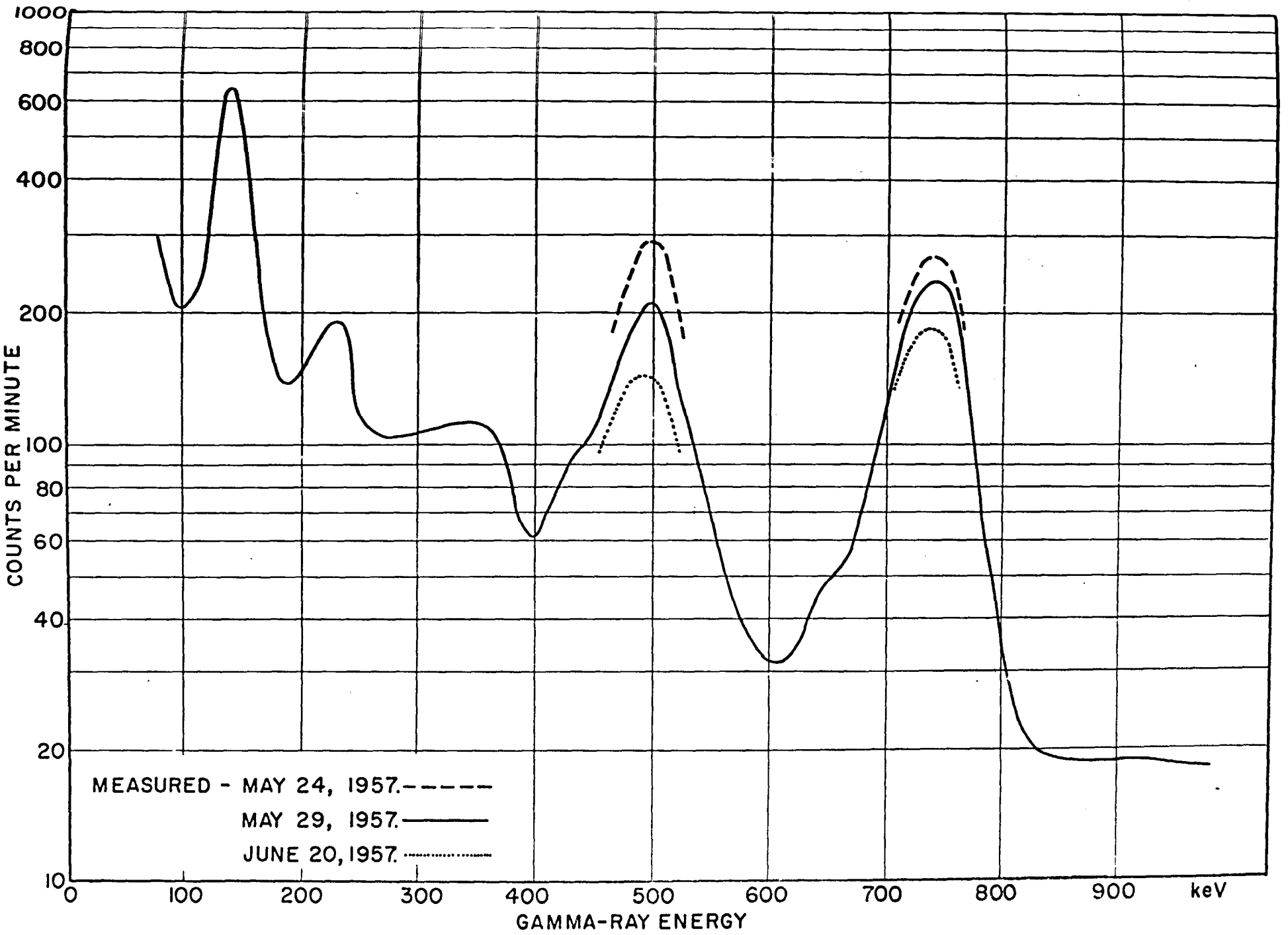

Figure 1 - Sample 1-1957, three measurements shortly after collection

The low energy region is indicated more fully in Figure 2 which shows the complete spectrum over the range from 20 keV to 900 keV for a 1957 sample taken 6 months after collection. In Figures 1 and 2 the 662 keV peak of Cs-137 possibly shows as the slight hump on the lower edge of the 740 keV peak; it does not show up distinctly, however, until the sample is more than one year old: see Figure 3, measured August 12, 1958, in which the 662 keV ray has emerged as the dominant activity.

The similarity in shape between the spectrum of the laboratory dust sample, shown in Figures 4 and 7, and the spectrum in Figure 2 indicates that the same nuclides are responsible for both. This very sensitive test of dust obtained from a large volume of air which had passed through the filter in front of the cooling fan in the pulse height analyser showed no evidence of contamination by the tracer isotopes used in the laboratory. An additional "ground" sample of house dust collected in a canister vacuum cleaner also exhibited a spectrum of the same general shape; this is shown in Figure 5.

The spectra in Figures 6 and 7 were obtained with a 100-channel pulse-height analyser designed by Atomic Energy of Canada Ltd., Chalk River, Ontario, and used on temporary loan from Computing Devices of Canada, Ltd. With this equipment a complete spectrum could be measured in a single run, very greatly reducing the work required. The counting periods could, therefore, be extended considerably for weak samples, and more frequent checks could be made of peak heights for half-life determinations.

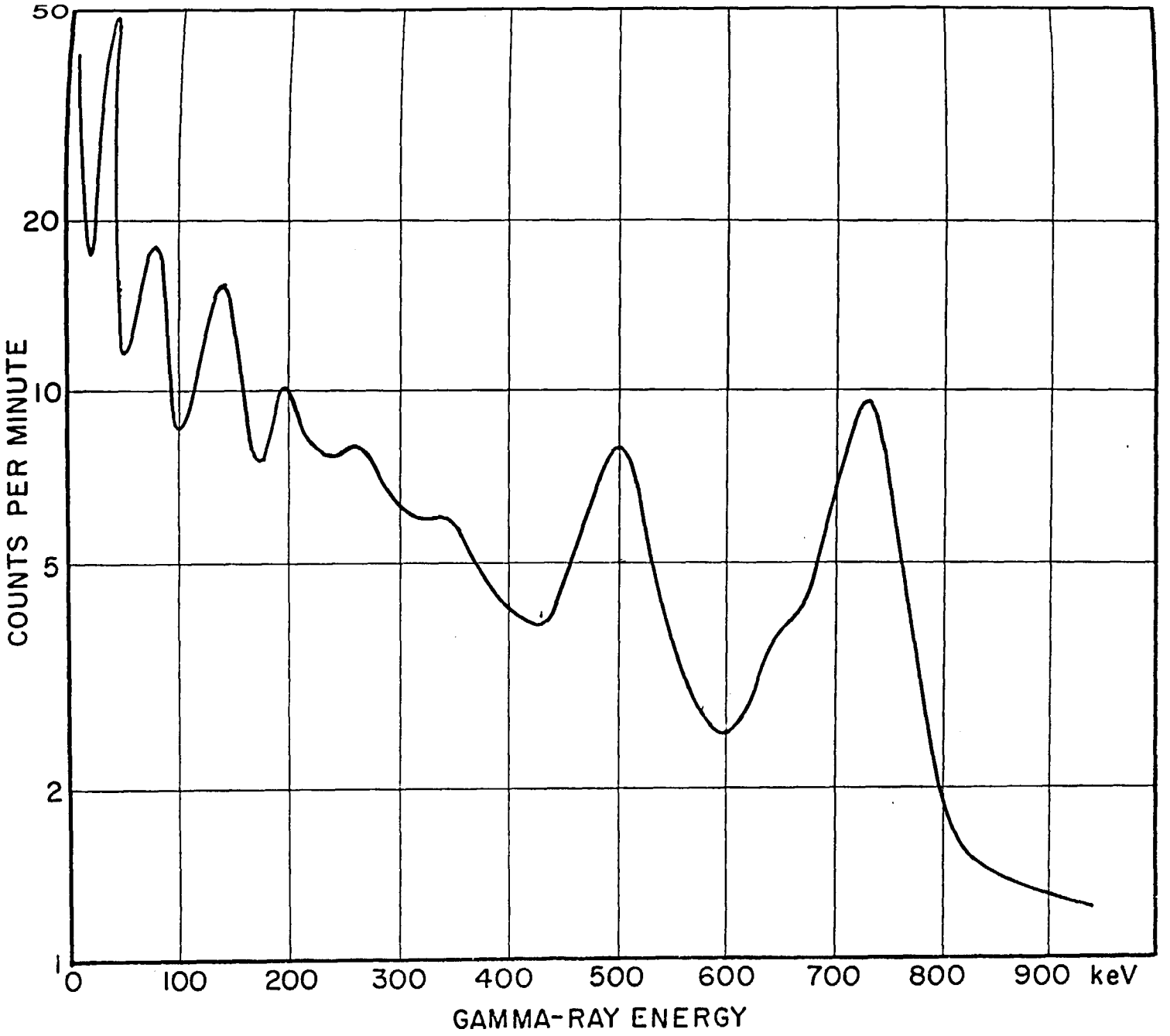

Figure 2 - Samples 12 to 15 - 1957 measured December 17, 1957
(Count-rate normalized to about that of a single sample for purposes of comparison)

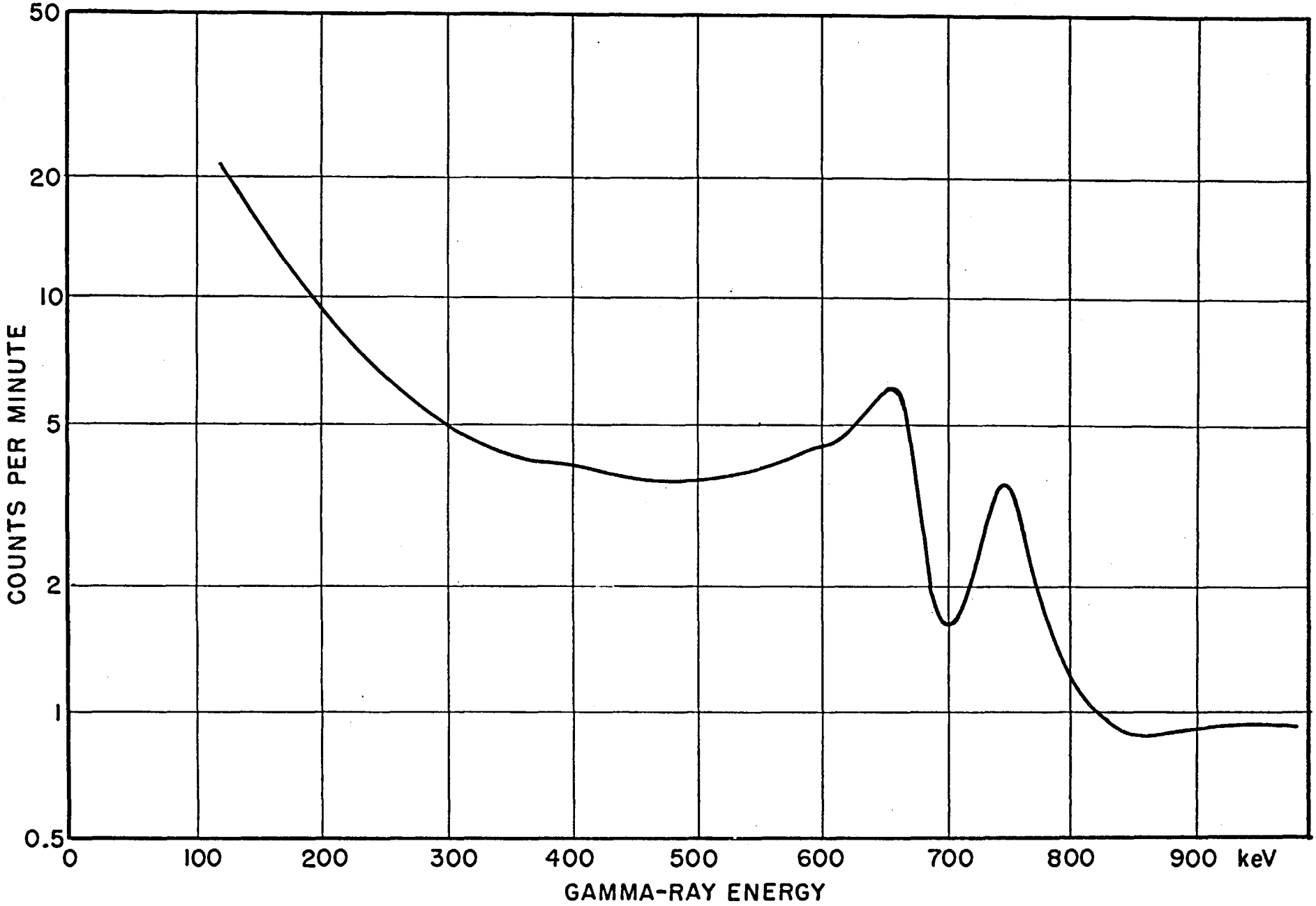

Figure 3 - Samples 4 to 15 - 1957 measured August 12, 1958
(Count-rate normalized to about that of a single sample for purposes of comparison)

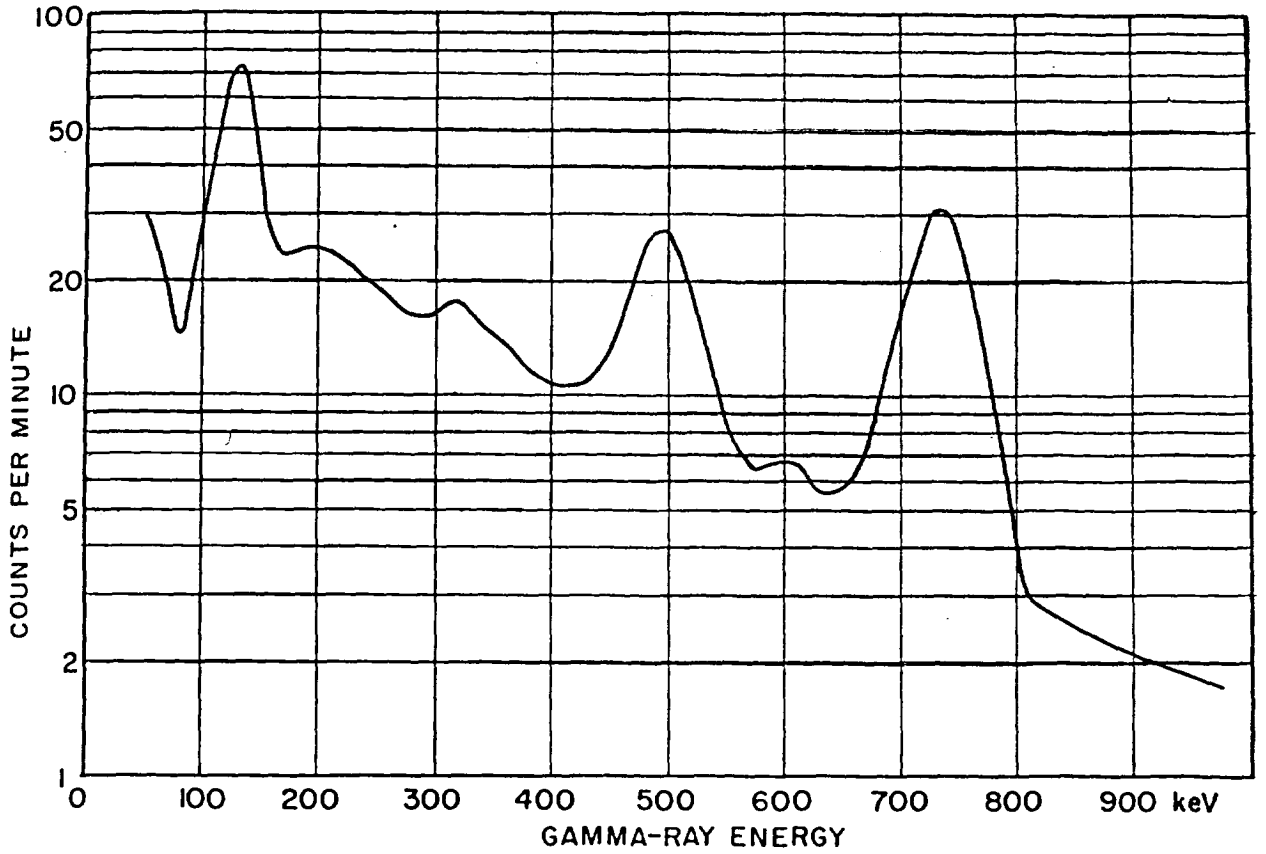

Figure 4 - Sample of laboratory dust measured November 7, 1957

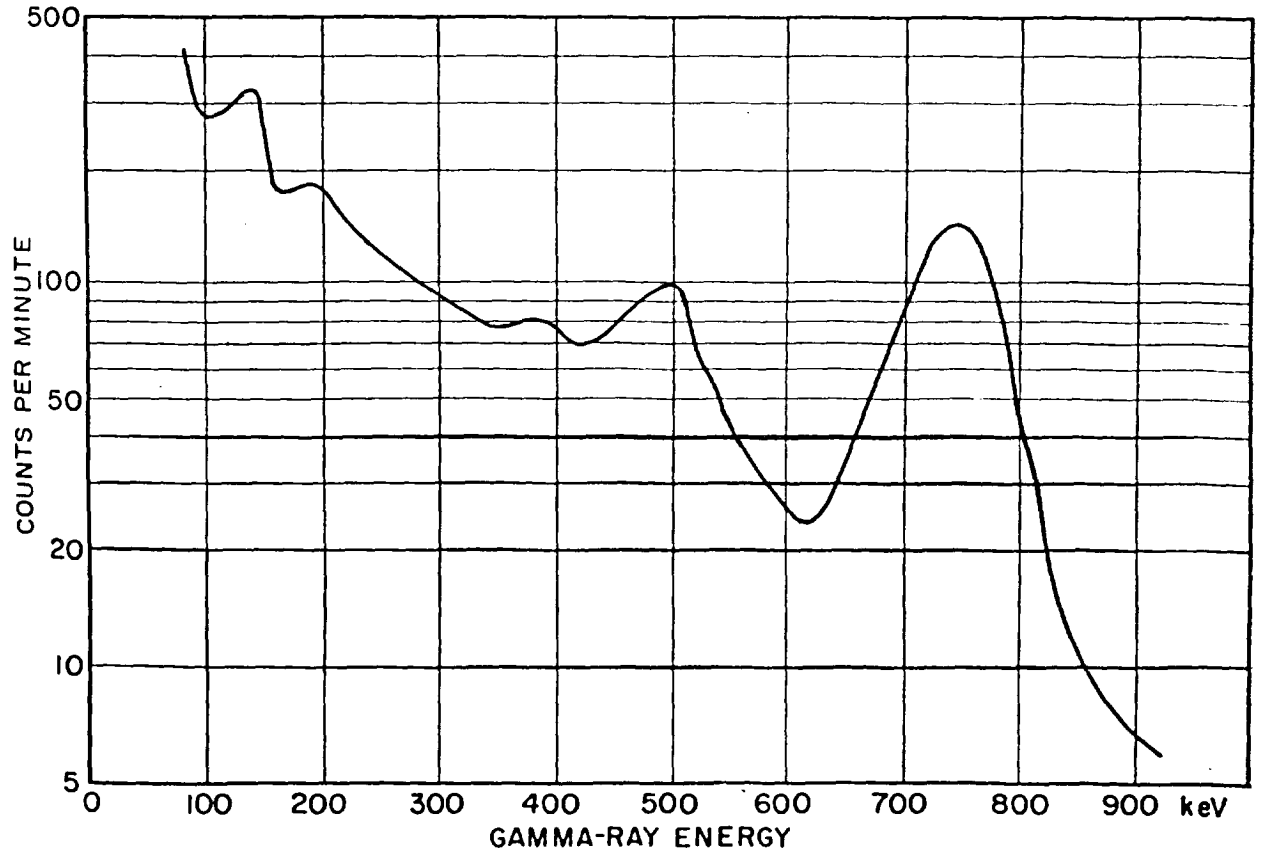

Figure 5 - Sample of house dust measured July 11, 1958

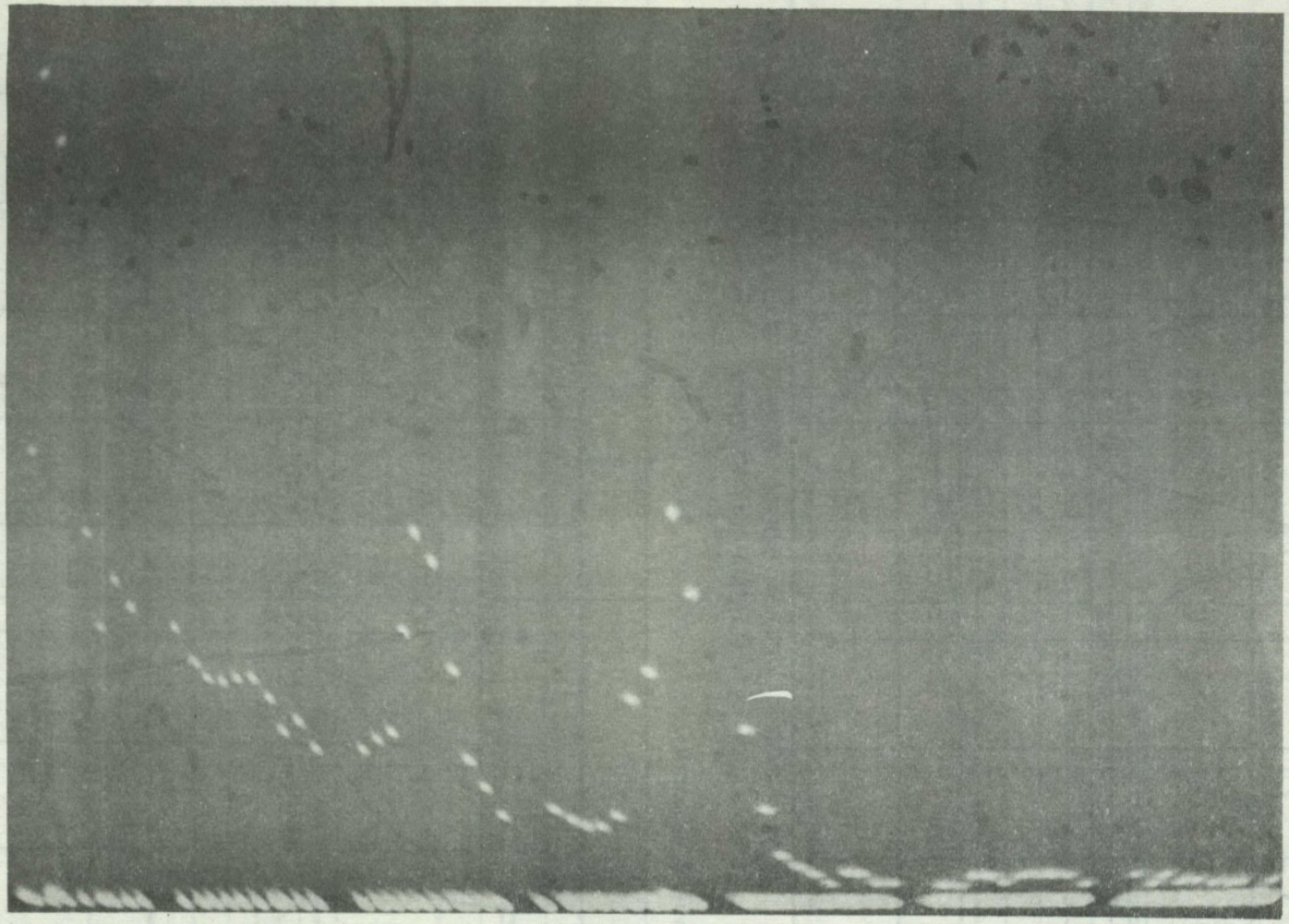

Figure 6 - Spectrum of an atmospheric dust sample as shown on a 100-channel, pulse-height analyser, November 7, 1957.

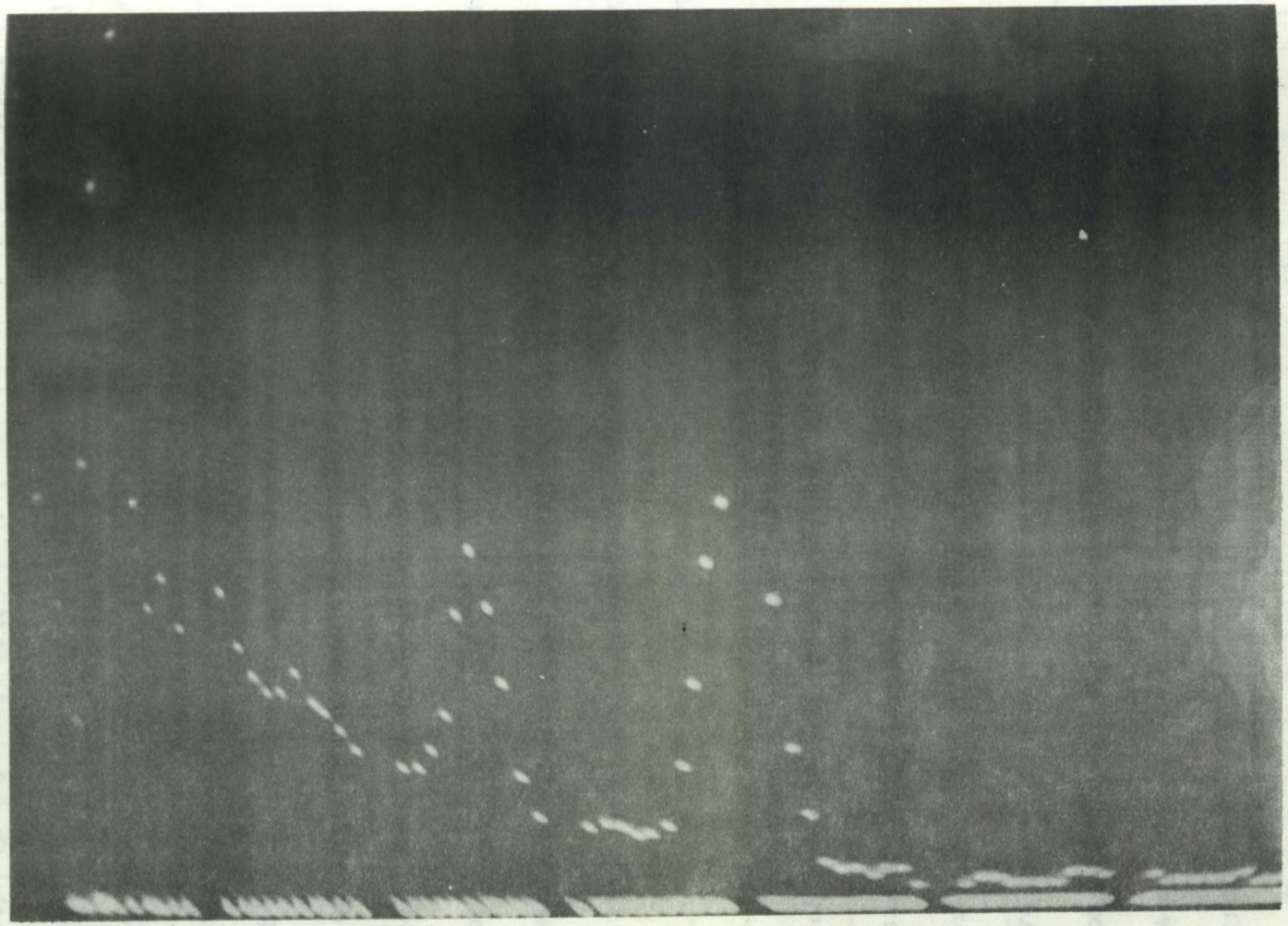

Figure 7 - Spectrum of the laboratory dust sample (see Figure 4) as shown on a 100-channel, pulse-height analyser, November 7, 1957.

The spectra shown in Figure 8 are those for the first 1958 sample, which was obtained over Eastern Ontario. The remainder of the 1958 samples, obtained over the Gulf of St. Lawrence, all show the same spectral shape. The spectrum in Figure 9, extended to 2750 keV, shows by the absence of peaks at 1800 and 2600 that measurable quantities of thorium and uranium dust (in secular equilibrium) are not present. As there is no evidence at all of the 1600 keV peak of La-140, the daughter of Ba-140, in these spectra, the fall-out atoms in the samples were probably two to three months old (see Appendix).

Spectra obtained in the low energy region are shown in Figure 10. The early decay in the 1958 sample around 160 keV has already been noted. In the same general peak the decay rate at 142 keV matches that of 33-day Ce-141, and the residual peak in the 1957 sample at about the same energy is identified as 285-day Ce-144 at 134 keV. These peaks cannot be resolved without radiochemical separation; it is therefore necessary to observe the gradual shift in energy of the combined peak and to rely on half-life estimates.

A peak due to X-rays from lead at 72.6 keV is clearly shown in all the low energy curves in Figure 10, except that of July 14, 1958, when a small block of lead used to press the sample down against the crystal enclosure was omitted. In this case a thin lead sheet (1/16 in.) placed over the sample caused the peak to reappear at close to its previous intensity. The spectrum of the background clearly shows the peak where higher energy background radiation is producing X-rays in the walls of the lead castle.

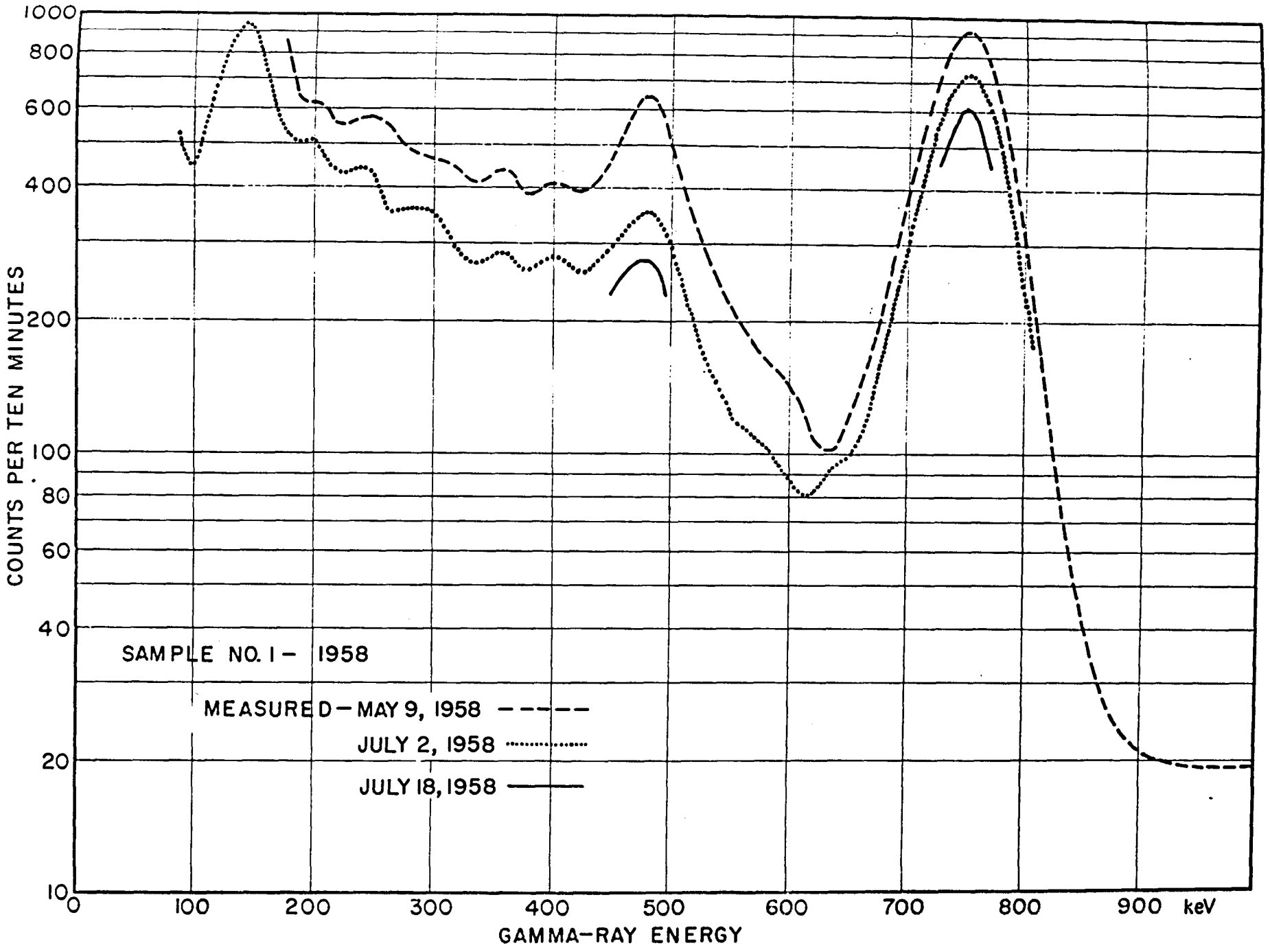

Figure 8 - Sample 1 - 1958, the first three determinations

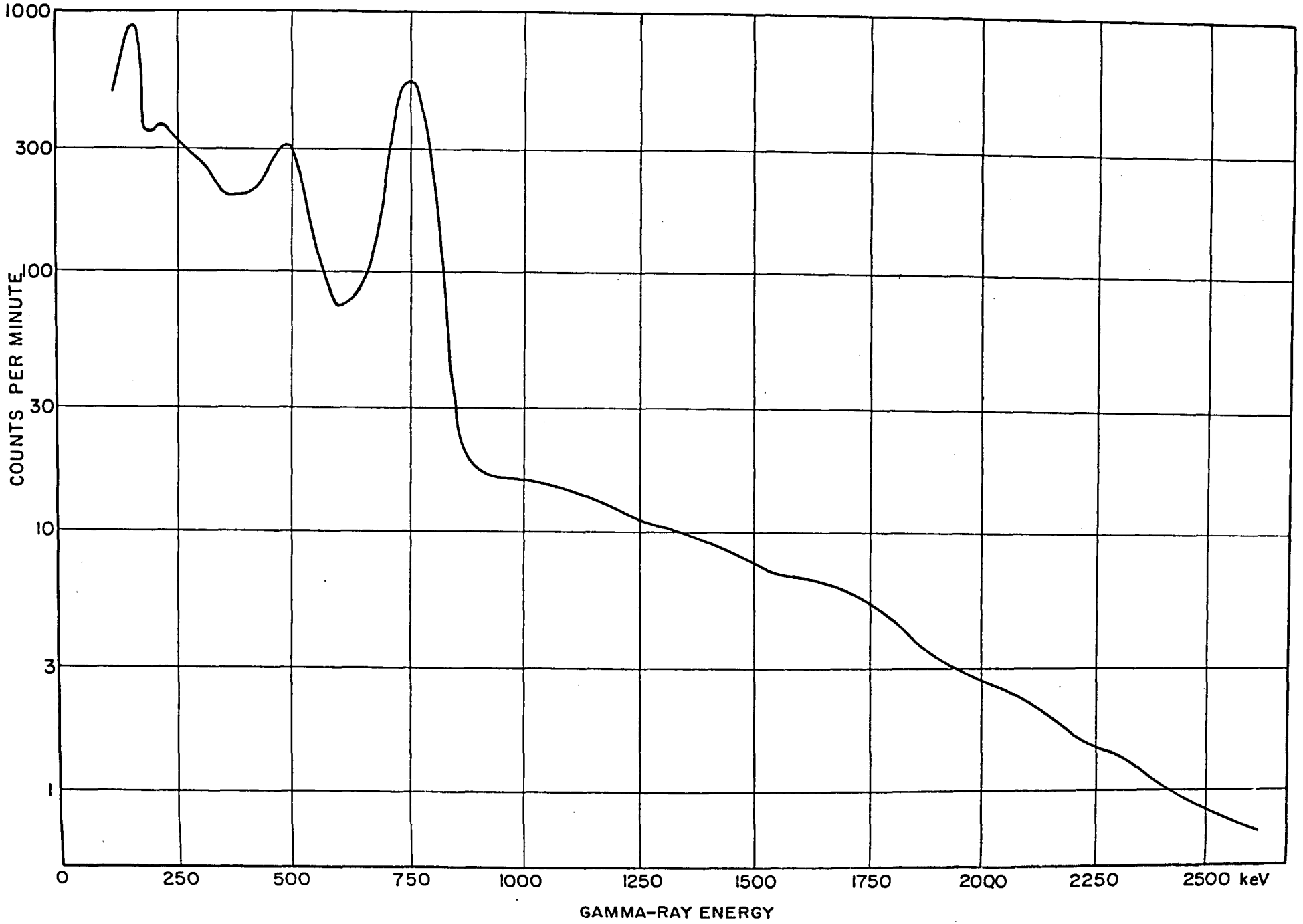

Figure 9 - Samples 1, 2, and 3 - 1958, extended spectrum, June 16, 1958

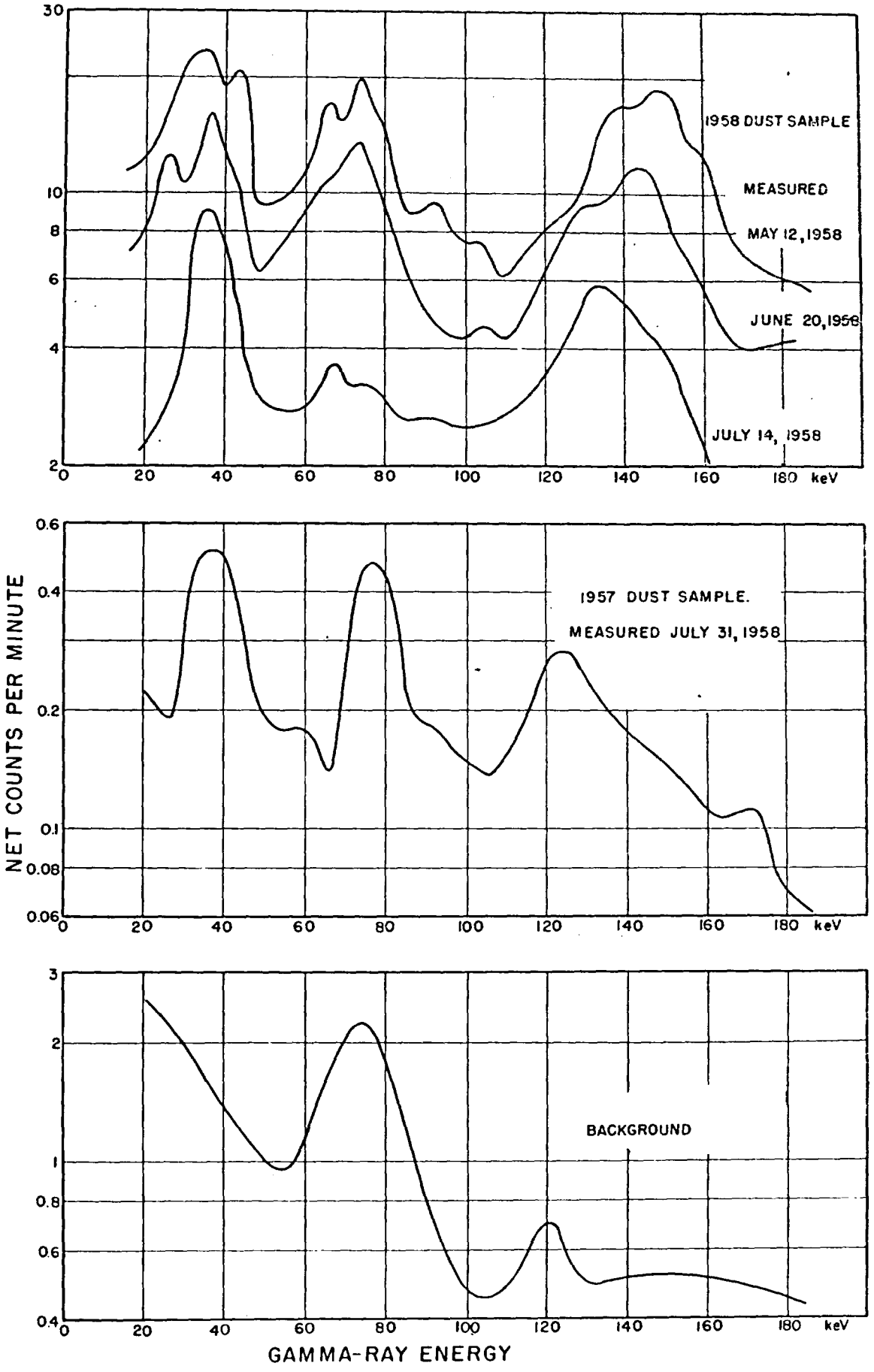

Figure 10 - Low energy spectra

Figure 9 - Samples 1, 2, and 3 - 1958, extended spectrum, June 16, 1958

The total activity in the principal peaks of Ru-103 and Zr-95 in the region 250 to 850 keV, for five samples combined, was found to be equivalent to 0.1 microcurie of Cs-137 shortly after the time of collection in mid-June 1958. The five filters, each having an area of about 0.2 sq ft, intercepted and retained a portion of the dust from 2×10^7 cu ft of air. This work supplements other recent beta determinations of atmospheric dust activity (1, 6).

ACKNOWLEDGMENTS

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APPENDIX

The following spectrum (see Figure 11), obtained on October 29, 1958, from a recent sample (collected on October 14 over the Gulf of St. Lawrence, on a flight to Ottawa on October 17, returning to the Maritimes on October 21, and over the Gulf on October 22), clearly indicated fission products which are only a few weeks old. The 1600 keV peak of lanthanum-140, daughter of 13-day barium-140, is quite evident in regard to energy: the nearest calibration peak was potassium-40 at 1460 keV; and in regard to half-life, the relative change in height of this peak in comparison with the other main peaks was noticeable after two days. The principal peaks are compared after a 12-day, and a 27-day, interval on the figure; the height of the 1600 keV peak above the continuum has decreased with a half-life value of 13 days.

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(Figure 11 follows,)
(on page 20.)

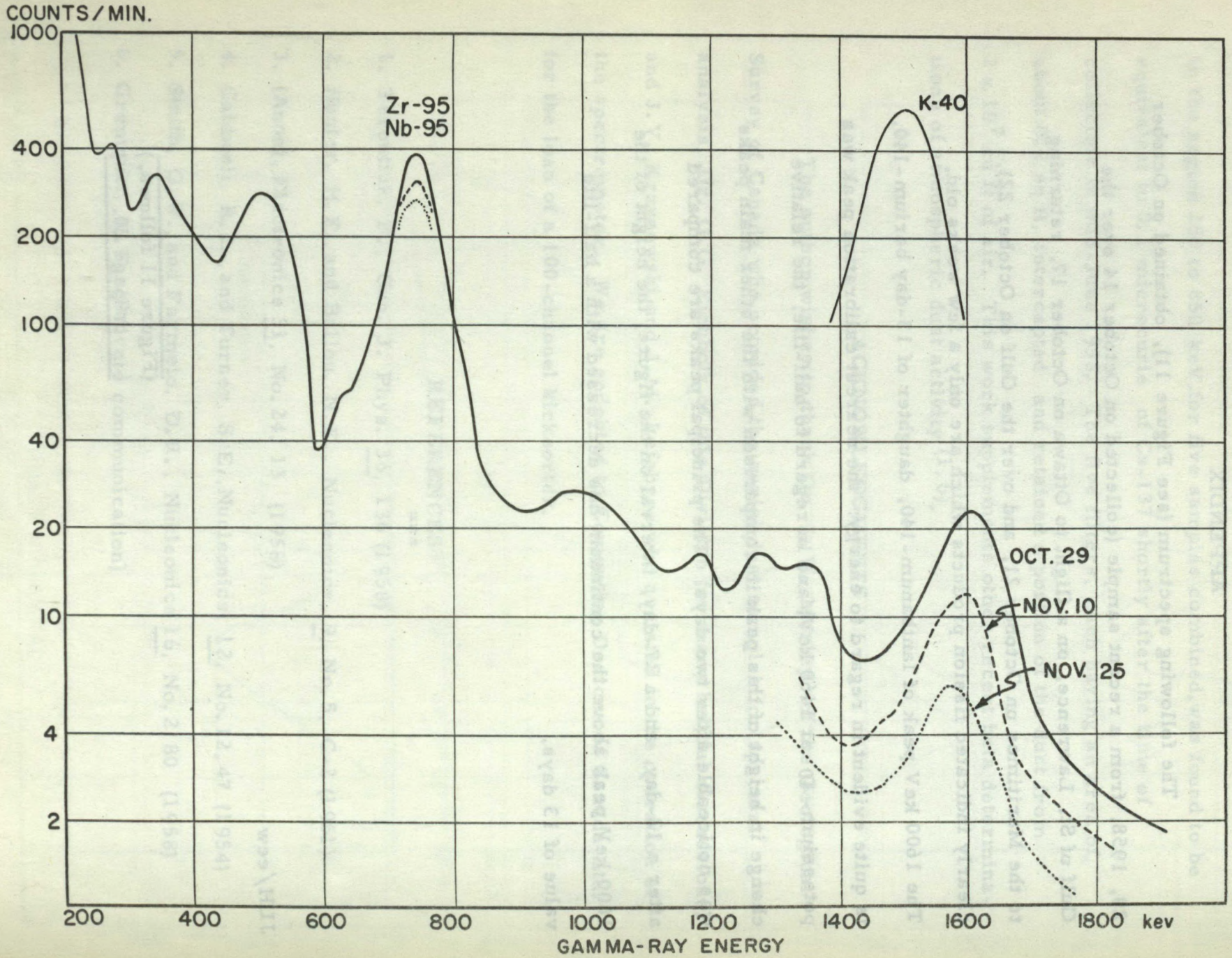

FIG. 11 - SPECTRUM OF DUST SAMPLE SHOWING La-140 PEAK.