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COMPARISON OF Ge(Li) AND ANTI-COMPTON SYSTEMS FOR MEASUREMENTS OF ENVIRONMENTAL SAMPLES

N. A. Wogman
Battelle, Pacific Northwest Laboratories
Richland, Washington

MASTER

Summary

There are numerous reasons for performing environmental measurements for routine health or safety monitoring and to determine the movement of trace elements or radionuclides through our environment to man. This is often a requirement for the licensing of nuclear power reactors, as well as many other meteorological or environmental research experiments. In this paper a variety of sensitive low-level counting systems have been discussed from an analyst's viewpoint, centering on a variety of NaI(Tl) and Ge(Li) gamma ray spectrometers. The coincident gamma-ray emitters are most sensitively detected through NaI(Tl) multidimensional gamma ray spectrometry, while single gamma ray emitters are very sensitively detected with Ge(Li) detector systems. NaI(Tl) detector systems are superior in general for environmental measurements.

Introduction

As the study of radionuclides in the environment is examined, a myriad of problems appear, some requiring very sophisticated electronics and counting systems, others requiring very simple analysis techniques. Of forty different problems in the reactor, fuel reprocessing, and associated nuclear industries, there are probably forty different solutions. When the measurement of environmental radionuclides is considered, the analyst is immediately forced to consider air, water, plants, animals, and man. In considering the design[1] of a radionuclide counting system, one of the first items that comes to mind is the size of the samples. If airborne radionuclide concentrations are to be measured, an air sampler capable of 1000 standard cfm pumping capacity could be used for periods of two weeks. For the sampling of water, one might think of evaporating a few liters of water, or of pumping the water through filter beds and ion exchange resins to remove the radionuclides from 1000 gallons of fresh or seawater. When the measurement of radionuclides in tissues or vegetables is required, an ashing procedure or freeze drying procedure is often utilized to reduce the bulk specimens to a few hundred grams. With all of these samples, an attempt is being made to measure environmental radionuclides in bulk materials. With this primary consideration in mind, the counting system must be designed such that it has a capability for the measurement of the many environmental radionuclides in large environmental samples. Some of the radionuclides which may be in these environmental materials are listed in Table I. An inspection of the decay schemes of these radionuclides immediately illustrates that they emit coincident gamma rays, multiple gamma rays, and single gamma rays. For these three cases, one would therefore design gamm-ray counting systems to measure the radionuclides of interest by different techniques, taking advantage of the various decay schemes. The factors governing the minimum activity level of these various detector systems from a specific gamma ray-emitting source are the presence of contaminating gamma-ray activity[2], the photopeak counting efficiency, and Compton and natural background under the photopeak of interest. These three factors help define the problem.

Development of NaI(Tl) Spectrometry

Gamma-ray spectrometry started in the late 1950's as NaI(Tl) crystals became available, as well as the necessary multi-channel analyzers, amplifiers, and analog to digital convertors. These new developments were of major importance to the measurement of radionuclides by their gamma-ray emissions. As is shown by the black line in Figure 1, ^{46}Sc from reactor effluents could easily be detected if it was chemically isolated and then analyzed by a single NaI(Tl) detector. However, if contaminating activity which one normally finds in the environment with ^{46}Sc is included, such as ^{65}Zn, ^{59}Fe, and ^{60}Co, then the gamma-ray peaks cannot be resolved one from another with the single NaI(Tl) detector system. This was typically the case for gamma-ray spectrometry on into the early 1960's. However, NaI(Tl) crystals in the early 1960's became available in fairly large sizes such that the radiochemist could attempt to complete his measurment and answer his problem by using a sum technique[3,4] -- that of total absorption of the gamma ray energies emitted in coincidence as they are deposited in a crystal. Figure 2 illustrates the results of this type of measurement where the radiochemist is measuring ^{60}Co, ^{46}Sc, and ^{65}Zn in an environmental water sample with little interference. One might note, however, that very small amounts of ^{65}Zn or ^{46}Sc would be lost in the Compton, which is derived from the sum peak of the 2.50 MeV ^{60}Co, if its concentrations were orders of magnitude greater than that of the ^{65}Zn or ^{46}Sc. A considerable number of the environmental radionuclides listed in Table I decay by coincident gamma ray emission; thus, as the crystals and memory cores became available in the mid-1960's, the radiochemist was able to develop multidimensional gamma ray spectrometers[5-10] which utilize coincidence techniques for radionuclide selectivity. For the first time, the environmental samples could be analyzed for their multiple radionuclide concentrations with some degree of sensitivity. As is shown in Figure 3, this type of system[5,6] is composed of two NaI(Tl) crystals which are 180° opposed. Between these crystals is placed a thick lead absorber with a sample hole. This absorber serves to attenuate scattered radiation between the top and bottom crystals. The entire primary NaI(Tl) detector system is anticoincidence shielded by either NaI(Tl) crystals or plastic phosphors. This results in a low background and reduced Compton

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interference. The resulting signals from either single gamma ray emitters or coincidence gammay ray emitters are fed normally to a 4096 channel hard-wired memory operated in a 64 by 64 channel array. Systems such as these use primary NaI(Tl) detectors ranging in size from 4" thick by 6" dia to 12" dia by 11" thick.[10] In Figure 4 the ^{46}Sc and ^{60}Co which as was noted earlier in Figure 1 had unresolved photopeaks as measured by a single detector, are now clearly resolved in the energy spectrum of the multidimensional gamma ray spectrometer even if their concentrations are quite different. The energy deposited in crystal 1 is on the axis designated Energy 1. The energy deposited in crystal 2 is on the axis denoted Energy 2. All coincidence events such as those illustrated for ^{46}Sc and ^{60}Co are in the energy-energy plane. The counts that are illustrated in the lines from the designated Sc or Co photopeaks to the Energy 1 or 2 axis areas are the Compton events which are minimized in this specific multidimensional crystal system by its anticoincidence shield.[6] Figure 5 illustrates the energy-energy coincidence areas and single gamma-ray photopeak areas for a multidimensional gamma ray spectrometer spectrum of environmental radionuclides in a separated sample of ocean water. It can be seen that a considerable number of radionuclides may be measured by their coincident gamma ray emission without any interference from neighboring photopeaks. This system allows some 20-25 radionuclides to be measured without chemistry in air filters. It allows 10-15 radionuclides to be measured in separated ocean water using ion exchange methods. Similar types of radionuclides can also be measured in foodstuffs and in man. In this manner, one can therefore investigate the radionuclides in an entire ecosystem from their point of origin through their transport and delivery to their uptake by man.

In considering the sensitivity of any gamma ray counting system, one must keep in mind the background that can be achieved with any specific system. In Figure 6 is an illustration of the axis backgrounds of a single 9" diameter by 8" thick NaI(Tl) gamma ray spectrometer.[9] This system illustrates the moderately low background achievable with thick concrete and lead shields. Typical backgrounds for a well-shielded system range from 0.3 to 10 counts per minute per 47 keV channel. The major components of the background are ^{40}K, decay products of ^{232}Th and ^{238}U, and cosmic ray events. Of course, a multidimensional system utilizes coincidence techniques between the primary detectors to lower the background even further. In Figure 7 a pair of crystals like the ones used to make the measurements in Figure 6 have been placed in a variety of anticoincidence and massive shield modes. The equal energy coincidence gamma backgrounds in this anticoincidence shielded spectrometer with 110 meters of overburden are of the order of 0.1 to 10 counts per thousand minutes per 47 keV by 47 keV hannel in the energy range from 0.250 to .5 MeV. The peaks that are illustrated are rom ^{40}K, ^{208}Tl, a daughter of ^{232}Th, and ^{214}Bi, a daughter of ^{238}U. The ^{40}K peak is at 0.73 MeV, which is from an equal energy disposition due to scatter of the single 1.46 MeV gamma in both crystals. The anticoincidence utilization alone on this type spectrometer system provides a background reduction of the order of 6-10. Lower backgrounds have been obtained utilizing beta-gamma-gamma coincidence systems[10]; these usually are of the order of 1 count in 30,000-100,000 minutes for a photopeak area. Some triple or quadruple gamma ray coincident requirement systems also have exceedingly low backgrounds.[11] Thus, in a period of some ten years the gamma ray spectroscopists have lowered their backgrounds as shown in Figure 6 from 1 to 100 counts per minute per 47 keV channel, to fractions of a count per 1000 minutes per 47 keV coincidence energy interval as shown in Figure 7.

Figure 8 illustrates the background obtainable for gamma rays in coincidence with gamma energy created by positron annihilation.[9] This is the coincidence background for 0.511 MeV deposition of energy in crystal 1 with 0 to 3 MeV deposition of energy in crystal 2. Backgrounds obtainable along this energy line are also of the order of 0.1 to a few counts per thousand minutes per 47 x 47 keV channel. A tenfold background reduction occurs by placing the spectrometer 110 meters underground and using a lead absorber to inhibit scattered radiation between crystal 1 and 2.

One might think that through the use of the aforementioned coincidence techniques a gamm-ray spectrometer system would lose its efficiency as the background was decreased by some 10,000-fold. However, for large NaI(Tl) crystal spectrometers, the absolute coincidence efficiencies as shown in Figure 9 are not less than 10% at the 1 MeV energy deposition point. The efficiency of the larger detector system for a point source of radioactivity is compared with that of a 6" thick by 11" dia. and 3" thick by 3" dia. NaI(Tl) crystal. The counting efficiency, E, is defined as the fraction of the gamma rays emitted by a source that are totally absorbed in one detector. The total coincidence counting efficiency, $2E_1E_2$, is defined as the probability of total absorption of two gamma rays, one in each of the two detectors. The coincidence counting efficiency of the 9" dia. by 8" thick system is 27 times greater than that of the 3" system and about equal to the 6" thick by 1" dia. system. The coincidence efficiencies for the two large crystal systems are only three- to fourfold lower than for one of their single crystals, yet they are still twofold higher than the efficiency of the single 3" crystal. The sum of the efficiency for the two large single crystals is between 50% and 90% over an energy range of 1500 to 300 keV respectively.

For environmental samples, the scientist must keep in mind the size of sample and the efficiency of the system which he hopes to utilize for analyses. A major advantage of the large detector systems compared with smaller systems is their high counting efficiency for large diameter samples. Both their single and coincidence photon counting efficiencies decline rather slowly with increasing source diameter. Figure 10 illustrates the decrease in efficiency for ^{60}Co for two counting systems. The first, which is a 6" thick by 5" dia. multidimensional gamma ray spectrometer crystal system has a decrease in efficiency from 1 to 0.4 for a changing sample size of a point source to a 4" dia. sample, while the large 11" dia. by 6" thick crystal system maintains its coincidence efficiency for this sample size at about 90%. The counting efficiency drops by about 20% in going from a point source to a 1" thick by 6" dia. source. Thus for large environmental samples one should go to a crystal system that has a fairly large area yet is thick enough to absorb the eminating gamma rays. This allows the direct counting of many large, low-activity samples without ashing or other concentrating techniques.

Development of Ge(Li) Gamma-Ray Spectrometry

NaI(Tl) detectors are not the only route which the scientist has available to him for the sensitive analysis of radionuclides in environmental materials. Lithium-drifted germanium [Ge(Li)] gamma-ray detectors are used extensively in many fields requiring the quantitative analysis of gamma rays. Over the past few years, a large variety of shapes and sizes of Ge(Li) detectors and various special configurations utilizing these detectors have been fabricated.[12-20] Along with this tremendous variety has come a considerable amount of uncertainty in their evaluation. In the past, the question of which detector to use for a particular problem was generally answered with intuition which was sufficient for many problems. However, this intuitive approach is not always sufficient and some problems such as the design of high-sensitivity gamma ray spectrometers require a more quantitative analysis.

Figure 11 shows the gamma-ray spectrum of a lichen analyzed first with a 5" dia. by 3" thick NaI(Tl) detector, then with a 20 cc, 11 mm deep Ge(Li) detector. This is not the ultimate in Ge(Li) detector systems; however, it is one which many universities or electric power companies might use for the analysis of some of their environmental samples.[12] This figure illustrates the high resolution of the Ge(Li) detector system as compared to a NaI(Tl) system, as well as the 100- to 1000-fold efficiency differences. Many of the radionuclides with significant concentrations analyzed with the Ge(Li) could not be seen with the NaI(Tl) system due to photopeak interferences. However, for low-activity material the Ge(Li) would not have sufficient efficiency for the analysis. Figure 12 shows the efficiency difference for a point source sample on a large 70 cc Ge(Li) detector and on an 11" dia. by 6" thick NaI(Tl) detector operated in coincidence or sum mode of gamma ray analysis. The overall efficiency difference varies from 10- to 100-fold. A Marinelli beaker sample system which would surround a Ge(Li) detector would lie somewhat lower than these extremes. Even though it has a lower efficiency, one might want to utilize a Ge(Li) system for its increased specificity in an anticoincidence mode[13-15] as shown in Figure 13. In this latter system, the Ge(Li) detector is surrounded with a NaI(Tl)[19] or a plastic phosphor anticoincidence shield.[13-16] In addition, the radioanalyst might wish to utilize two germanium detectors[16] or a Ge(Li)-NaI(Tl) coincidence arrangement[19] to lower background and specific radionuclide interferences. A good illustration of the utilization of an anticoincidence shielded Ge(Li) diode system is shown in Figure 14 of the analysis of a Bikini tuna sample. The upper spectrum is a result of a gamm-ray coincidence between the Ge(Li) and a plastic phosphor shield; the lower spectra are those events which were not in coincidence. The lower left-hand corner of this figure illustrates four energy areas: ^{65}Zn, ^{54}Mn, ^{137}Cs and ^{57}Co. It is obvious that the Ge(Li) detector utilized for this analysis could not resolve ^{57}Co, ^{54}Mn, or ^{65}Zn if it was just used by itself. However, in its anticoincidence operational mode it did a good job of resolving these three peaks, as well as improving the statistics for ^{137}Cs analysis. If one were to consider this type of anticoincidence analysis further, it would become obvious that a combination of NaI(Tl) and Ge(Li) coincidence techniques might yield a high-efficiency, high-resolution system. This spectrometer would be constructed like that shown in Figure 13 except that the lower Ge(Li) detector would be replaced by a NaI(Tl) crystal.[19] In this system one requires a gamma ray to deposit its energy in in the NaI(Tl) detector. This would then open a gate allowing analysis of the second coincident gamma ray which has then entered the Ge(Li) detector. In Table II is an illustration of the radionuclides to be measured by this type of window arrangement. For example, in grouping B, there are five energy windows for coincident events. This would require a 6000 channel analyzer at the minimum. The first 1000 channels would be utilized to store those gamma ray events from the Ge(Li) detector which were not in coincidence with anything. The second 1000 would be those events in coincidence with an energy deposition of from 500 to 700 keV in the NaI(Tl) detector. The third 1000 would be those events from 700 to 950 keV of energy deposited in the NaI(Tl) detector, and so forth; thus, some radionuclides would appear in multiple spectra. The 0.511 MeV peak from ^{22}Na would trigger the 500-700 keV gate of the NaI(Tl) detector, and the 1275 keV gamma of the ^{22}Na would then be deposited in the Ge(Li) coincidence spectra from this gate. In addition, the 1022 keV sum from ^{22}Na would trigger the 950-1250 gate; again the 1275 keV gamma would be recorded in this spectra. The utilization of this type of system can be seen in Figure 15, a gamma ray spectra of caribou liver. In the operation of the detector system to analyze this material, a 1022 keV coincidence gated spectrum with a window width of 300 keV was utilized. In the upper curve the normal Ge(Li) spectrum has had the gated coincidence spectrum subtracted. The upper spectrum shows only ^{40}K, ^{22}Na, and ^{137}Cs. However, the gated spectrum contains ^{60}Co and ^{108m}Ag in addition. This spectrum illustrates the first data obtainable for ^{108m}Ag in an environmental specimen. It was this anticoincidence-coincidence gated system that allowed this type of sample to be analyzed for this specific radionuclide.

Comparison of NaI(Tl) and Ge(Li) Gamma-Ray Spectrometry

Having been presented with the general capabilities of single NaI(Tl) systems, total absorption NaI(Tl) systems, multidimensional NaI(Tl) systems, as well as a variety of Ge(Li) systems, the scientist might well be confused as to which specific detector would complete his environmental job. Only through a comparison of these systems can the best spectrophotometer for the study be chosen.[21] In Figure 16 is an illustration comparing the background counting rates of large NaI(Tl) detectors and Ge(Li) systems. The highest background is for two 11" dia. by 6" thick NaI(Tl) detectors which are being used as a total absorption spectrometer; they have no anticoincidence shields. The summed background varies from 10 to 100 counts per minute per peak area in the range from 0 to 3 MeV. This compares to 10 to 100 counts per 1000 minutes for their coincidence background total peak area. A 70 cc Ge(Li) detector has a background which is comparable if not a little higher than the NaI(Tl) coincidence spectra. Figure 17 illustrates a comparison of the counting efficiency of ^{60}Co as a function of source diameter for these large NaI(Tl) and Ge(Li) detectors. The highest efficiency of 40% to 50% is for the gamma ray sum mode of operation of two of the large 11" dia. by 6" thick NaI(Tl) crystals. The coincidence efficiency is on the order of 10% for this system when source diameter ranges from 0 to 10 inches. The 70 cc Ge(Li) detector varies from 2% to 0.2% efficiency for the same range of source sizes, much lower than the NaI(Tl) coincidence or sum efficiencies. In Figure 18 is a rough figure of merit comparison utilizing the efficiency from Figure 17 and background from Figure 16 for large NaI(Tl) and Ge(Li) systems. The efficiencies used for the NaI(Tl) detector systems were strictly for a simple coincidence system, not for the exotic anticoincidence shielded, multidimensional gamma ray spectrometer systems discussed earlier in this paper. The highest figure of merit obtainable was for the coincidence technique utilizing the multidimensional gamma ray spectrometer system; second was utilization of the 70 cc Ge(Li) diode; and last, the utilization of a gamma ray sum technique for the analysis of a point source specimen. This latter figure would thus indicate that for large environmental samples and the analysis of coincidence gamma ray emitters, the large NaI(Tl) multidimensional gamma ray spectrometer would be the detector of choice. Based upon the earlier discussion, the detector of choice for a point source of single gamma ray-emitting radionuclides would be a simple Ge(Li) detector with an efficiency of about 15%. If either of these systems has a considerable amount of interfering radionuclide activity, it is going to require an anlytical radiochemical approach to determine which system will yield the most data. In practice both systems should often be utilized if an entire series of 20-25 radionuclides are to be analyzed in environmental materials ranging from air and water to vegetation and animal tissue to man.

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TABLE I

ENVIRONMENTAL RADIONUCLIDES

SINGLE γ-RAY	MULTIPLE γ-RAYS	COINCIDENCE γ-RAYS
^{7}Be	^{57}Co	^{22}Na
^{40}K	^{58}Co	^{46}Sc
^{51}Cr	^{59}Fe	^{60}Co
^{54}Mn	^{65}Zn	^{88}Y
^{95}Nb	^{95}Zr	^{110m}Ag
^{103}Ru	^{106}Rh	^{124}Sb
^{137}Cs	^{125}Sb	^{134}Cs
^{141}Ce	^{131}I	^{140}La
^{144}Ce	^{140}Ba	^{152}Eu
	^{144}Pr	^{207}Bi
	^{155}Eu	

TABLE II

COINCIDENT GAMMA-RAY ENERGY WINDOWS FOR ENVIRONMENTAL RADIONUCLIDES

ENERGY WINDOW (keV) GROUPING A	ENERGY WINDOW (keV) GROUPING B	γ-RAY DETECTED IN NaI(Tl) "WELL"	RADIONUCLIDE	COINC. γ-RAY DETECTED IN Ge(Li)
500	500	511	^{22}Na	1275
		570	^{207}Bi	1063
		603	^{124}Sb	1692
		605	^{134}Cs	796
		615	^{108m}Ag	434, 723
		658	^{110m}Ag	885
	700	723	^{108m}Ag	434, 615
		796	^{134}Cs	605
		815 SUM	^{140}La	1595
		885	^{110m}Ag	658
		889	^{46}Sc	1120
		898	^{88}Y	1836
950	950	1022 SUM	^{22}Na	1275
		1049 SUM	^{108m}Ag	723
		1063	^{207}Bi	570
		1120	^{46}Sc	809
		1157 SUM	^{108m}Ag	615
		1172	^{60}Co	1332
	1250	1275	^{22}Na	511
		1332	^{60}Co	1172
		1338 SUM	^{108m}Ag	434
1450	1450	1595	^{140}La	328, 487, 815
		1692	^{124}Sb	603
		1786 SUM	^{22}Na	511
		1836	^{88}Y	898
1950	1950			

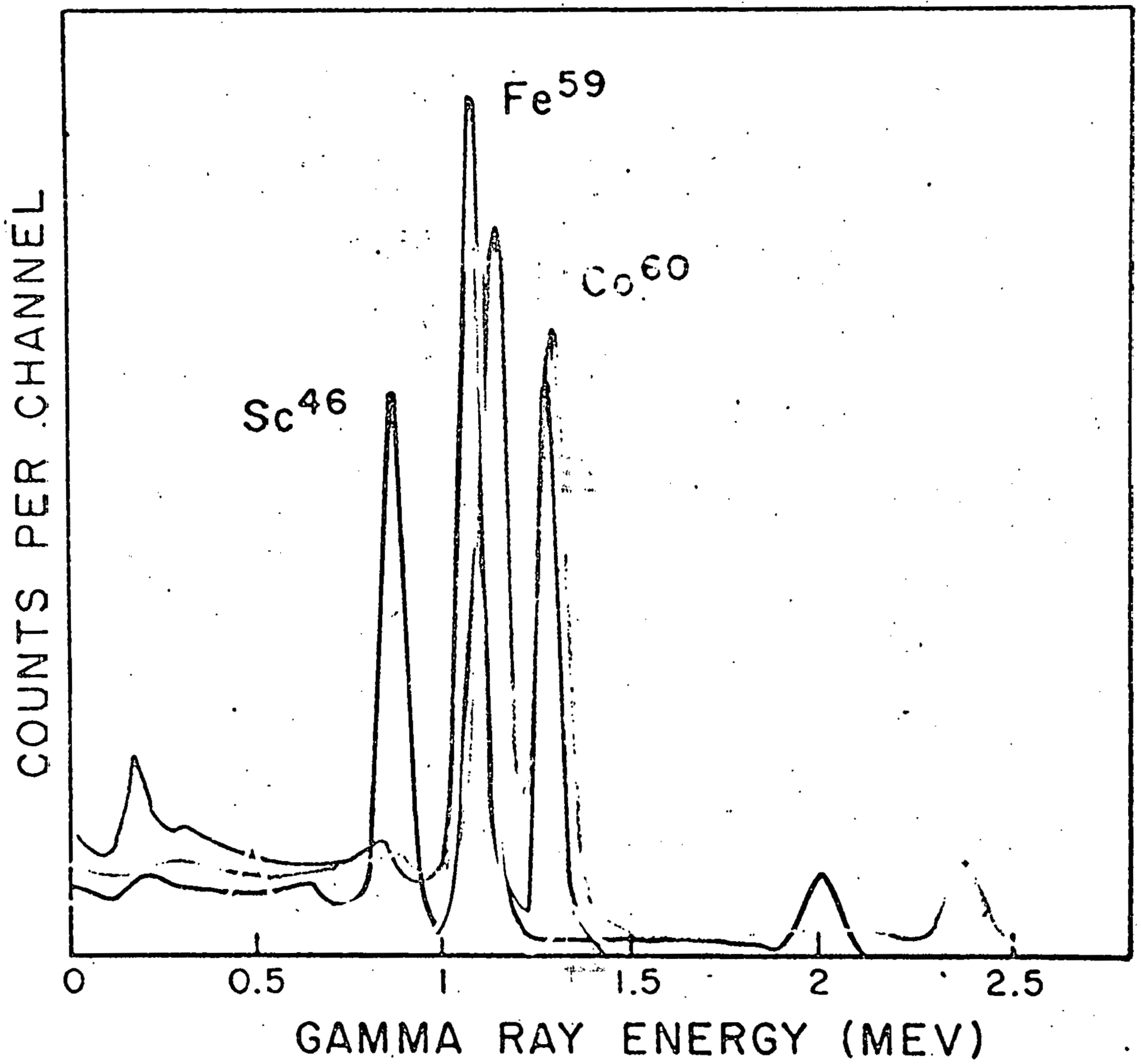

Fig. 1. Gamma ray spectra of ^{46}Sc, ^{65}Zn, ^{59}Fe and ^{60}Co taken with a single 3 inch diameter by 3 inch thick NaI(Tl) crystal.

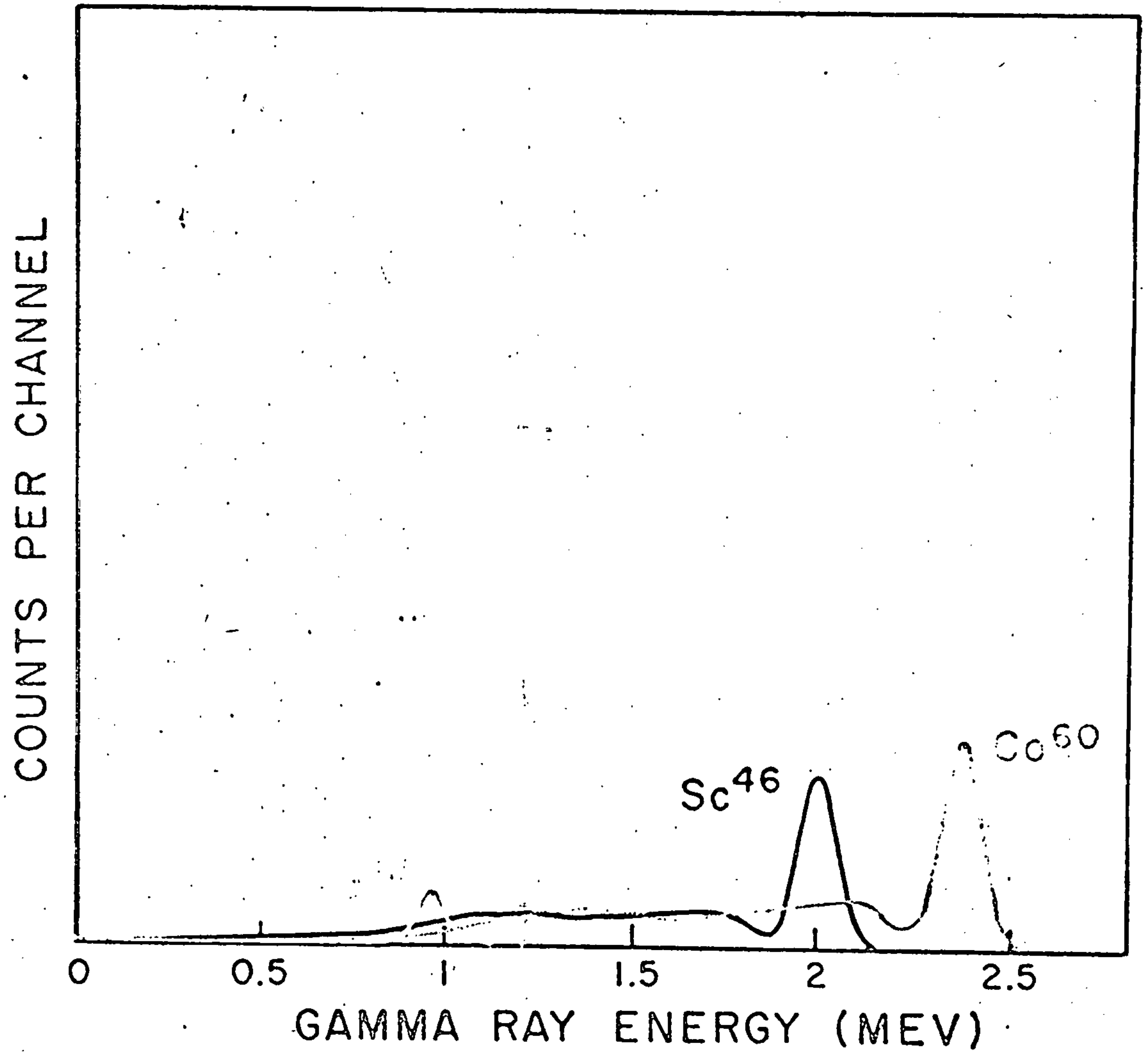

Fig. 2. Gamma-ray spectra of ^{65}Zn, ^{46}Sc and ^{60}Co in a 9 inch diameter by 9 inch thick large NaI(Tl) well crystal.

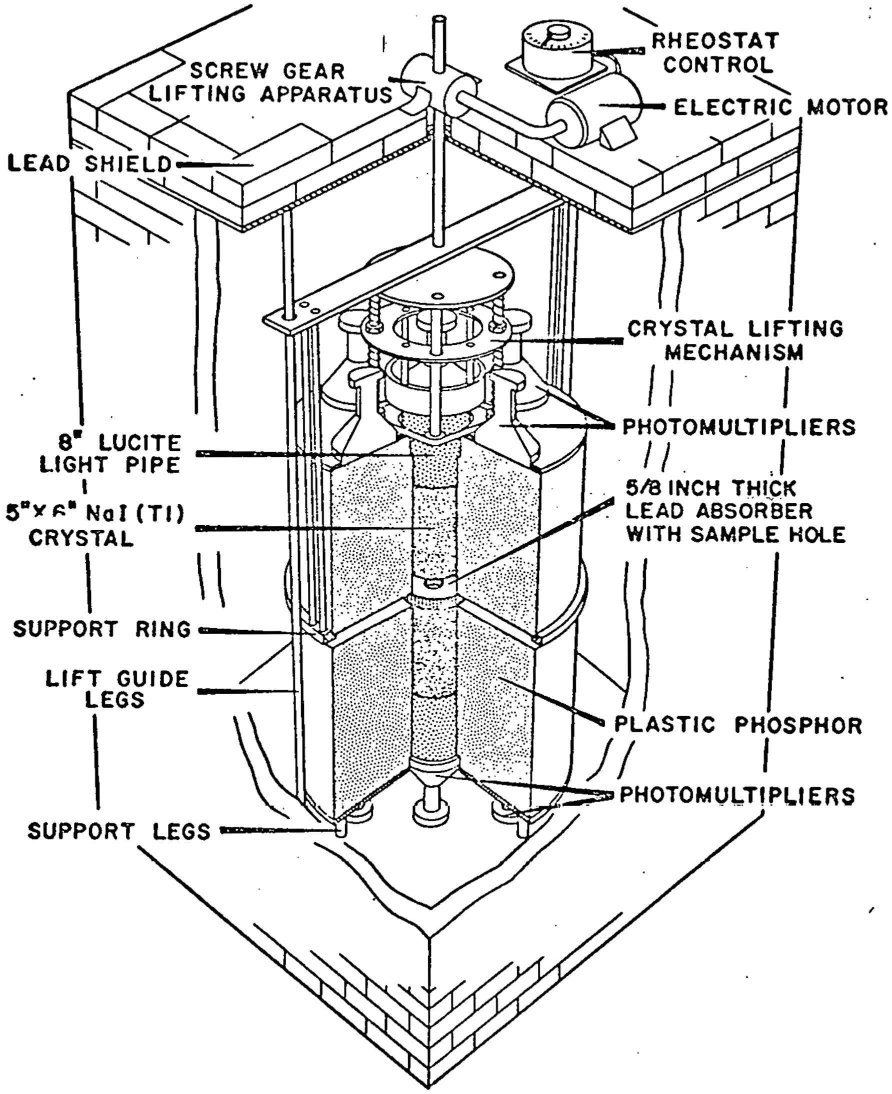

Fig. 3. Sodium iodide-plastic phosphor detector system for multidimensional gamma ray spectrometry.

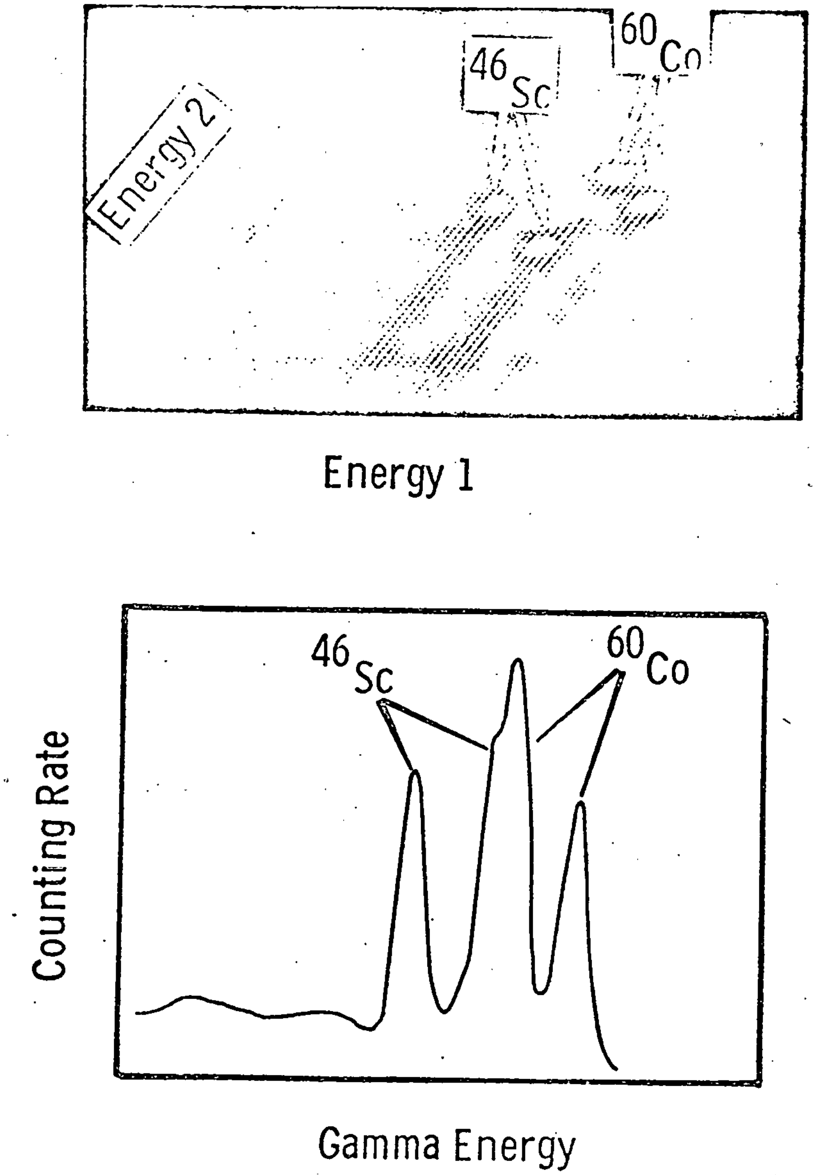

Fig. 4. Comparison of multidimensional gamma-ray and single crystal spectra.

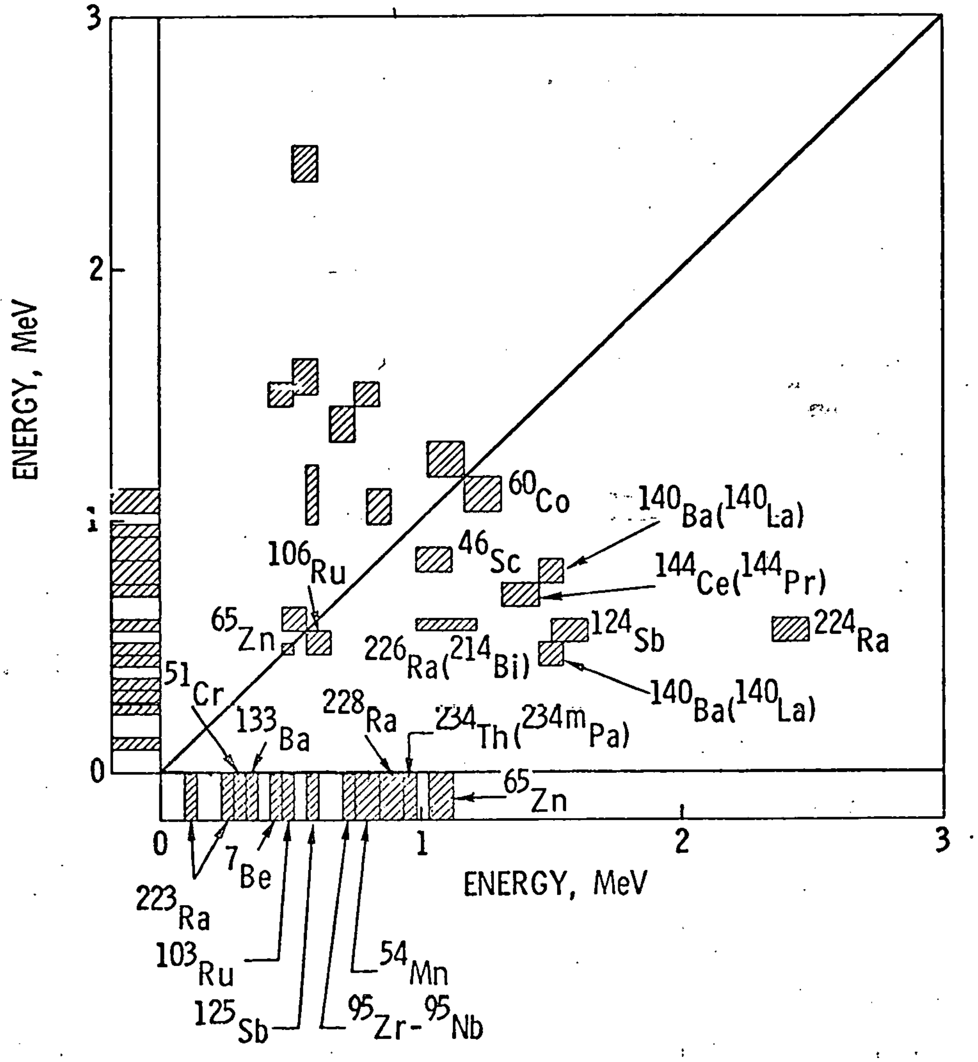

Fig. 5. Radioisotope matrix areas for use in oceanography.

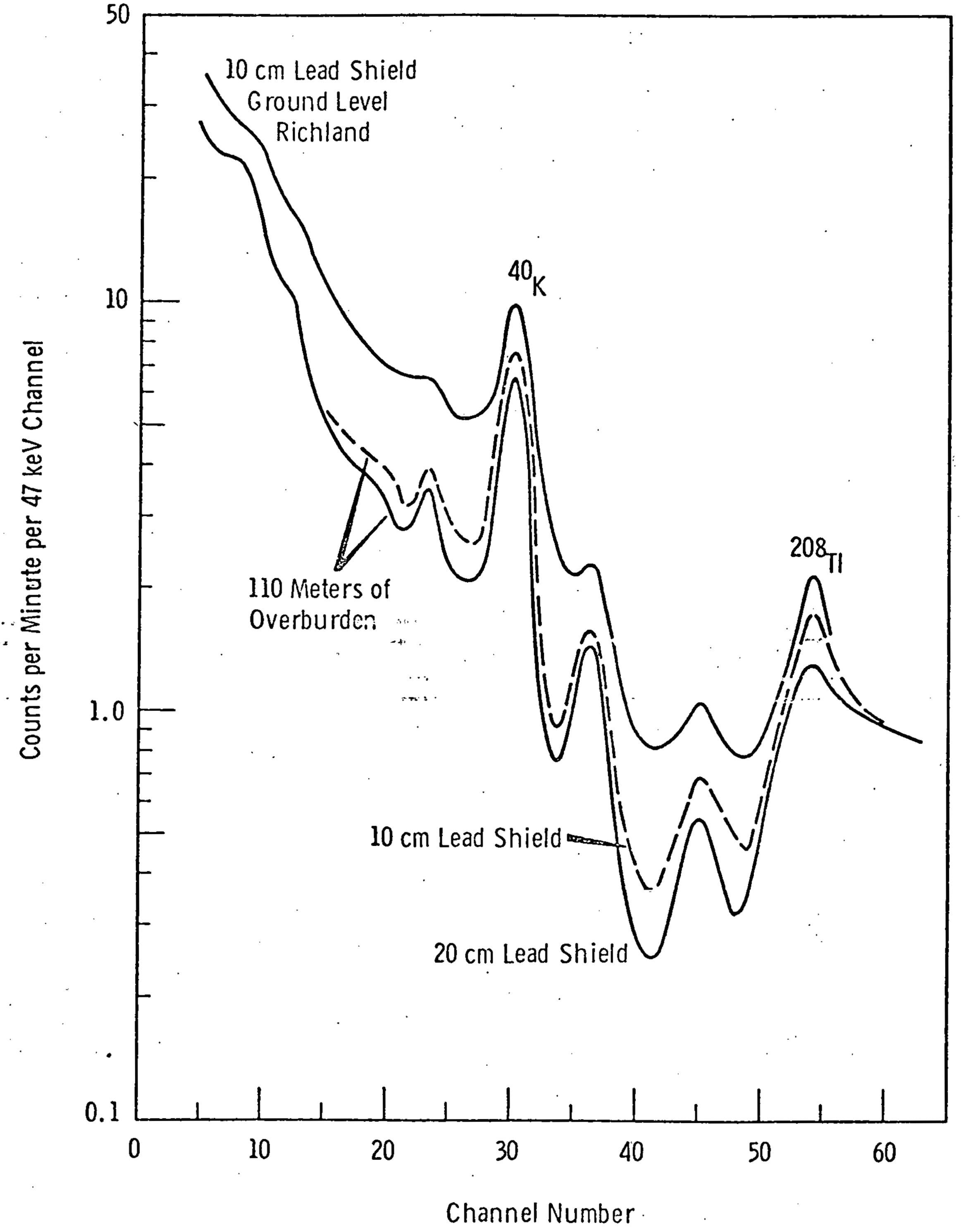

Fig. 6. Comparison of single crystal background response to cave shielding mass.

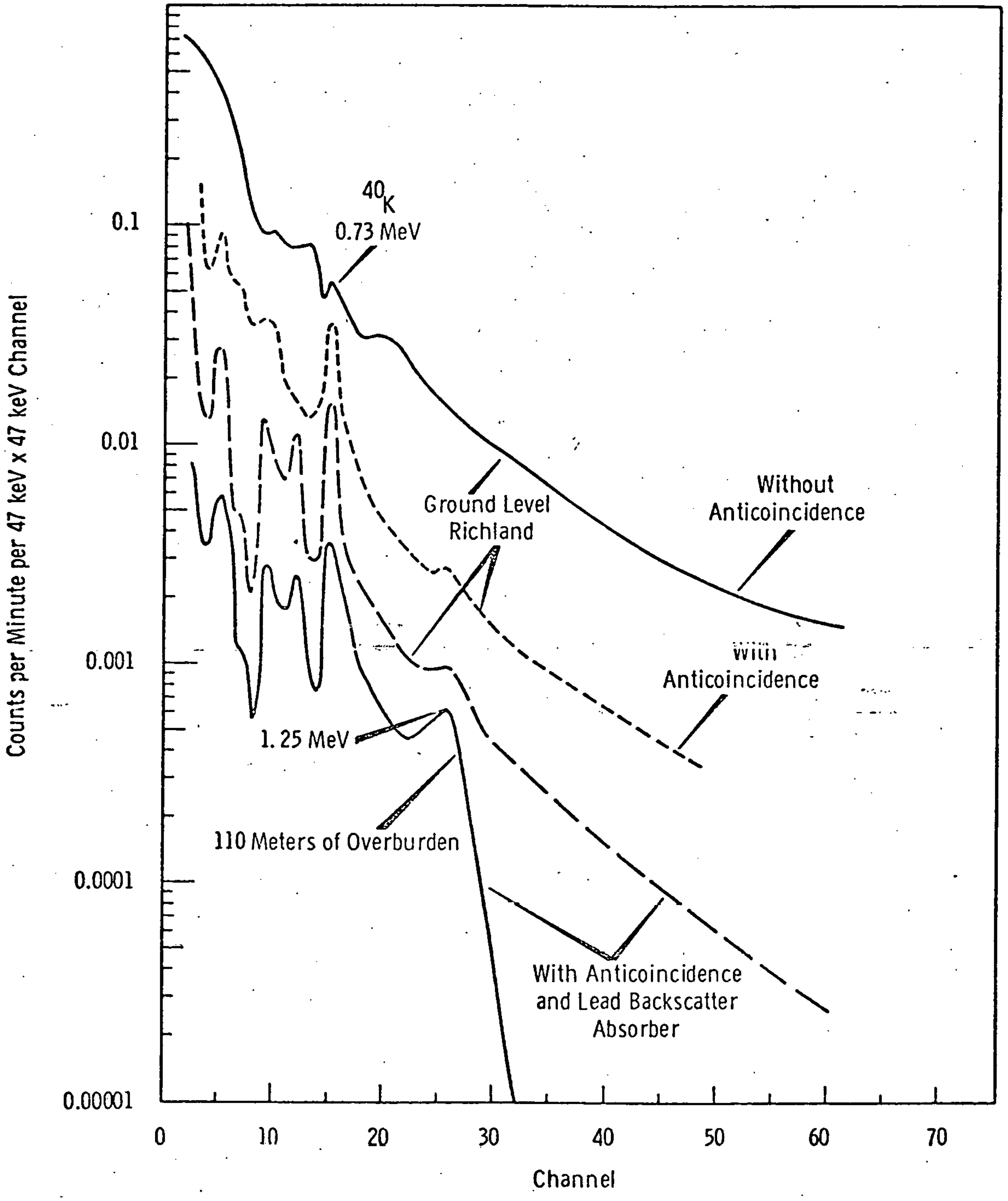

Fig. 7. Comparison of the equal energy coincidence background response to various mass, anti-backscatter, and anticoincidence shielding conditions.

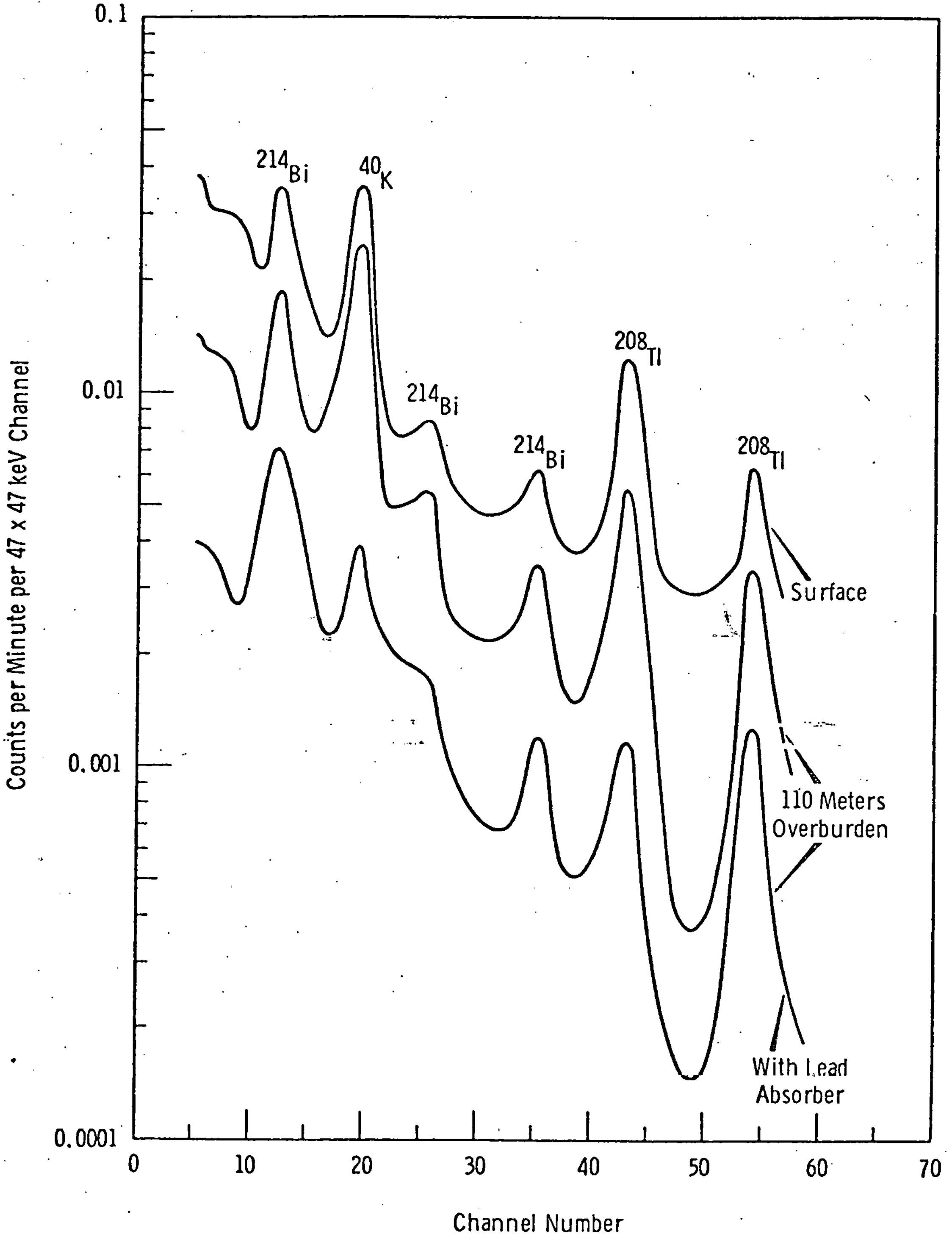

Fig. 8. Comparison of the background gamma spectrum in coincidence with annihilation radiation to various mass shielding conditions.

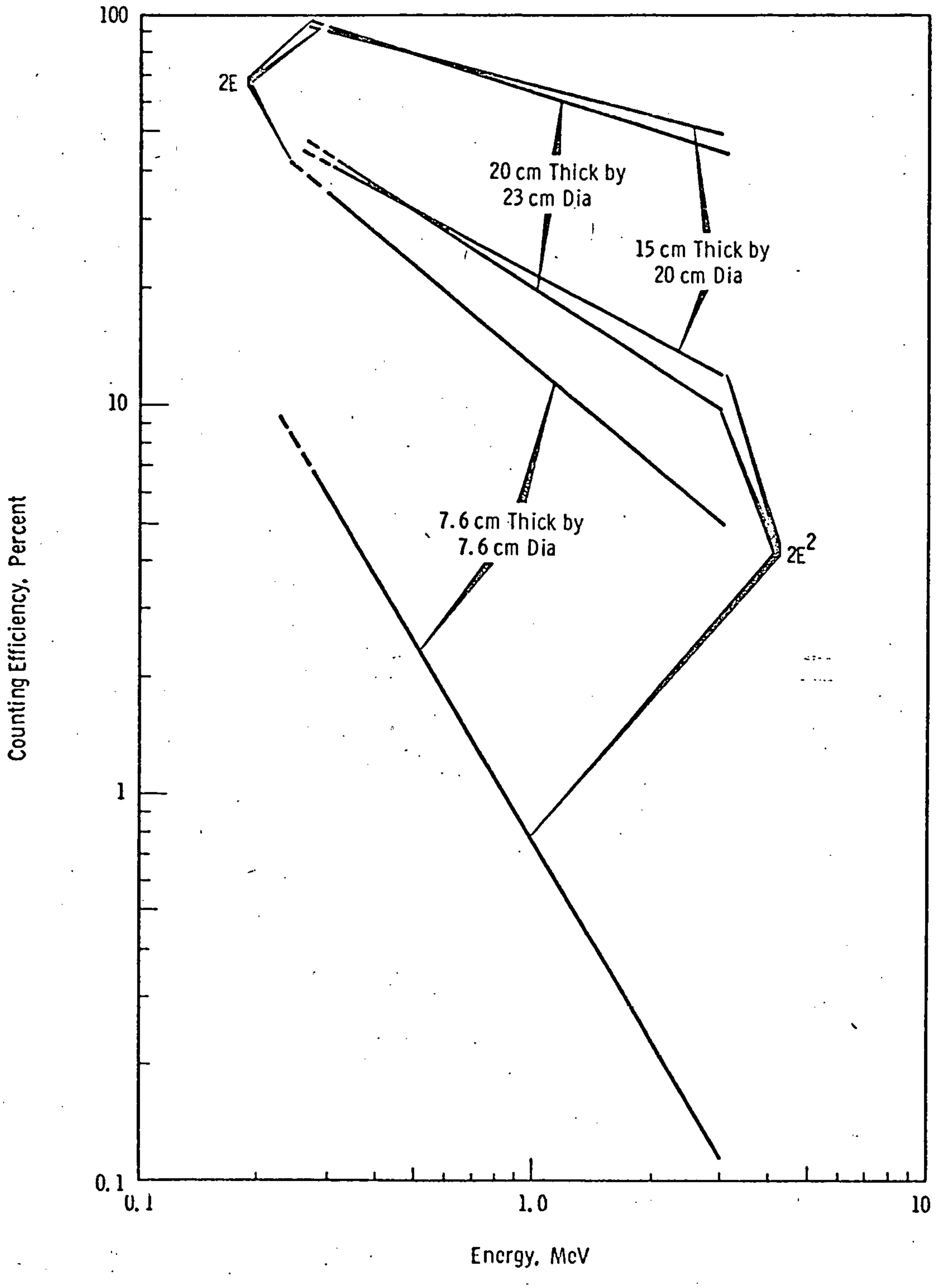

Fig. 9. Total counting efficiencies and total coincidence counting efficiencies of three multidimensional gamma-ray detector systems.

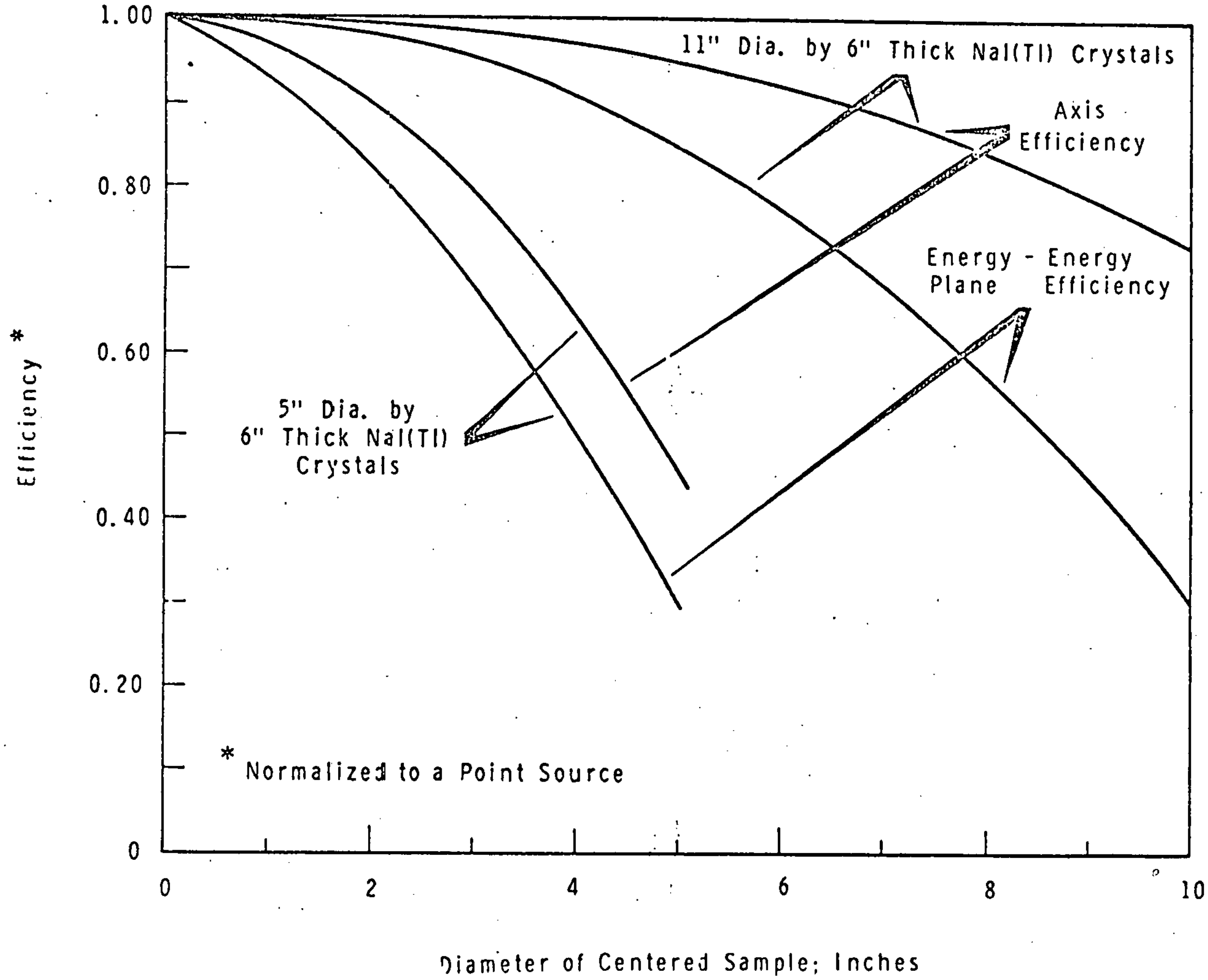

Fig. 10. Comparison of the counting efficiency of ^{60}Co as a function of source diameter for 2 multidimensional detector systems.

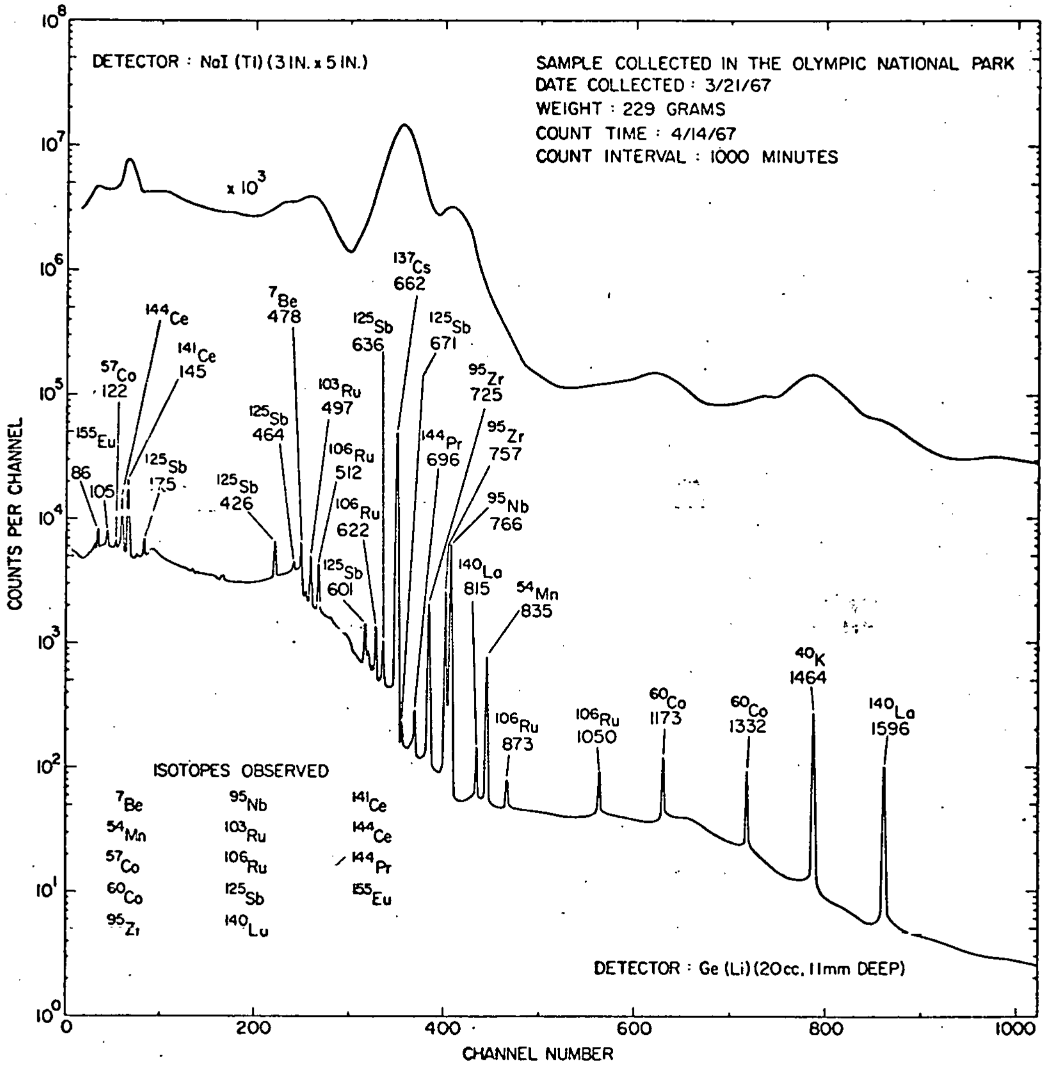

Fig. 11. Gamma-ray spectrum of lichen.

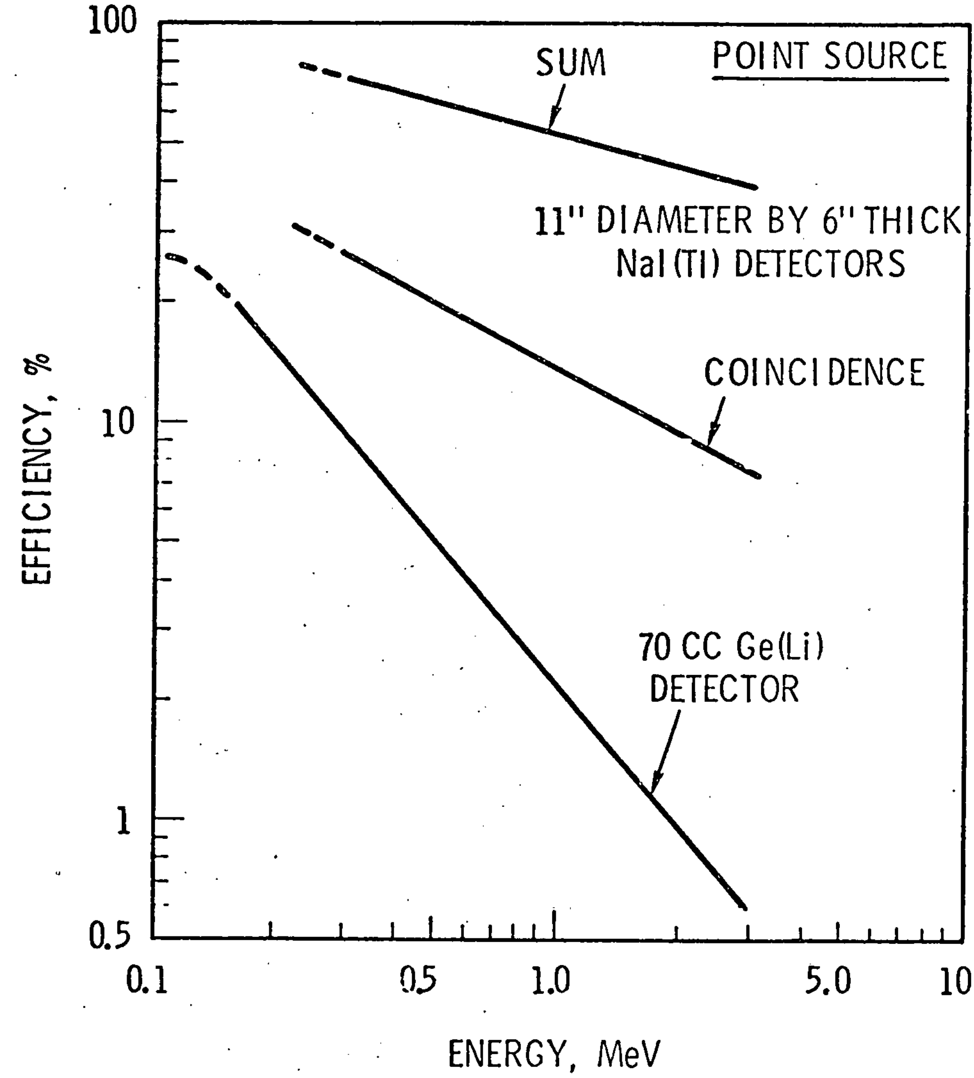

Fig. 12. Gamma-ray counting efficiencies for large NaI(Tl) and Ge(Li) detectors.

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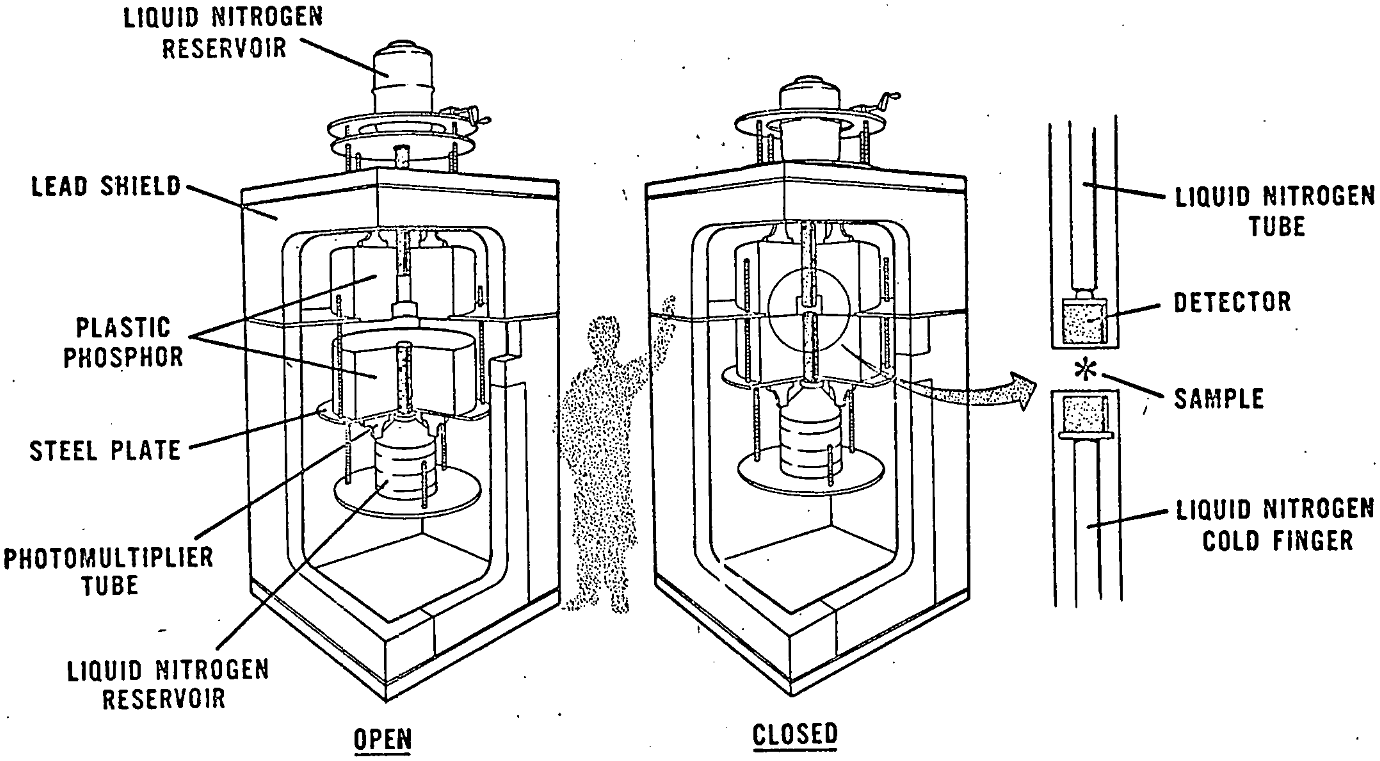

Fig. 13. Anticoincidence spectrometer system.

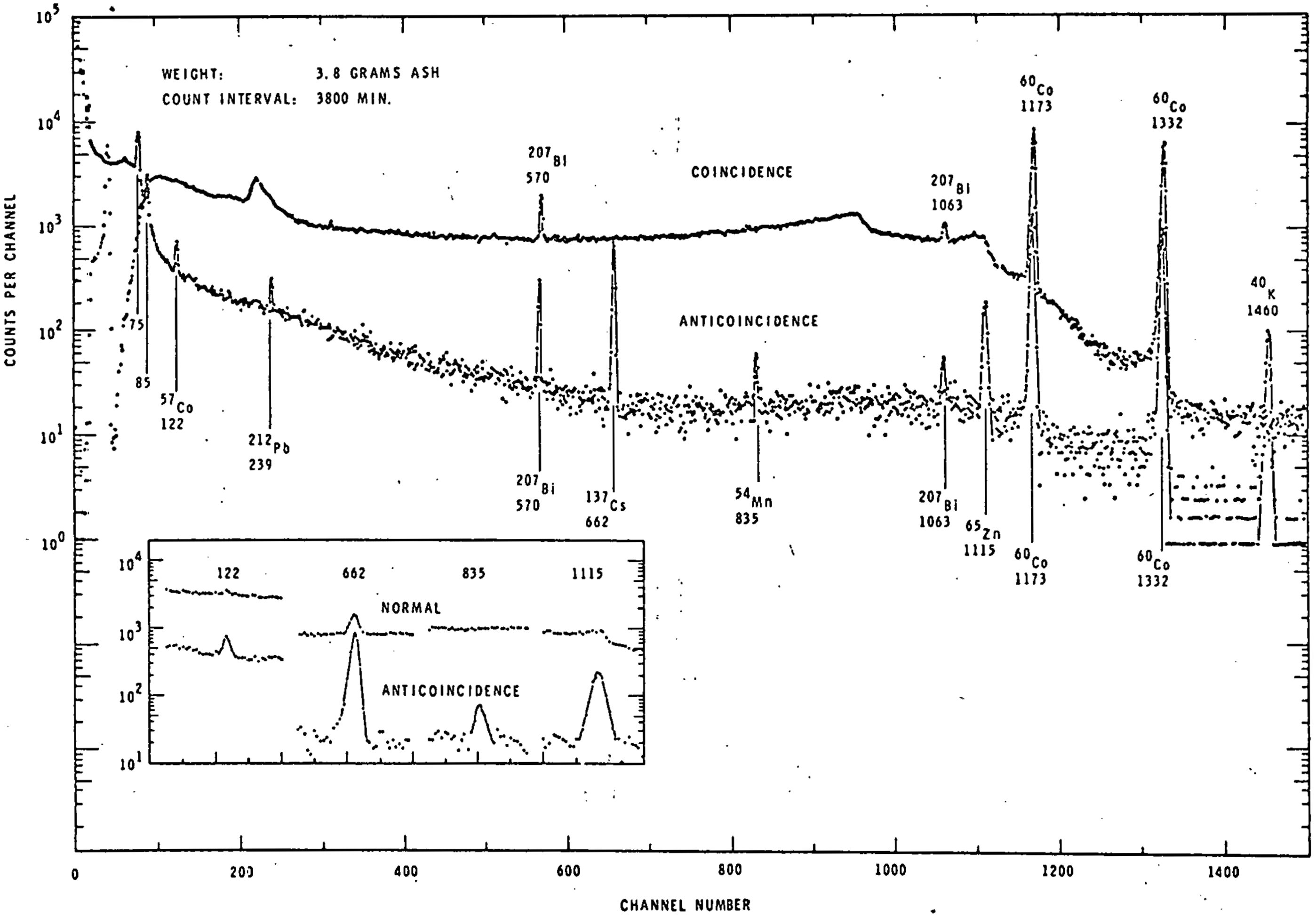

Fig. 14. Gamma-ray spectra of Bikini tuna.

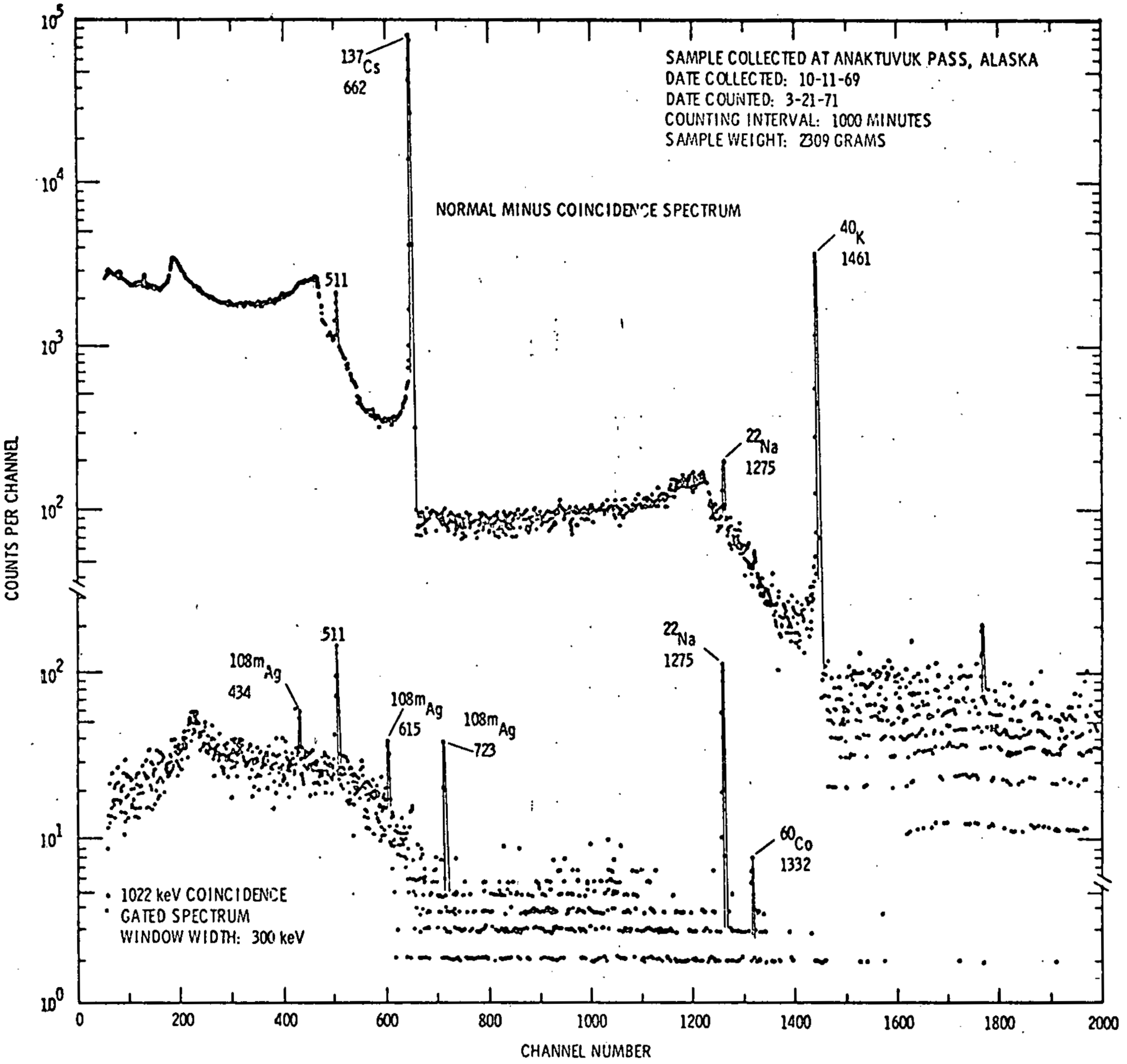

Fig. 15. Gamma-ray spectra of caribou liver.

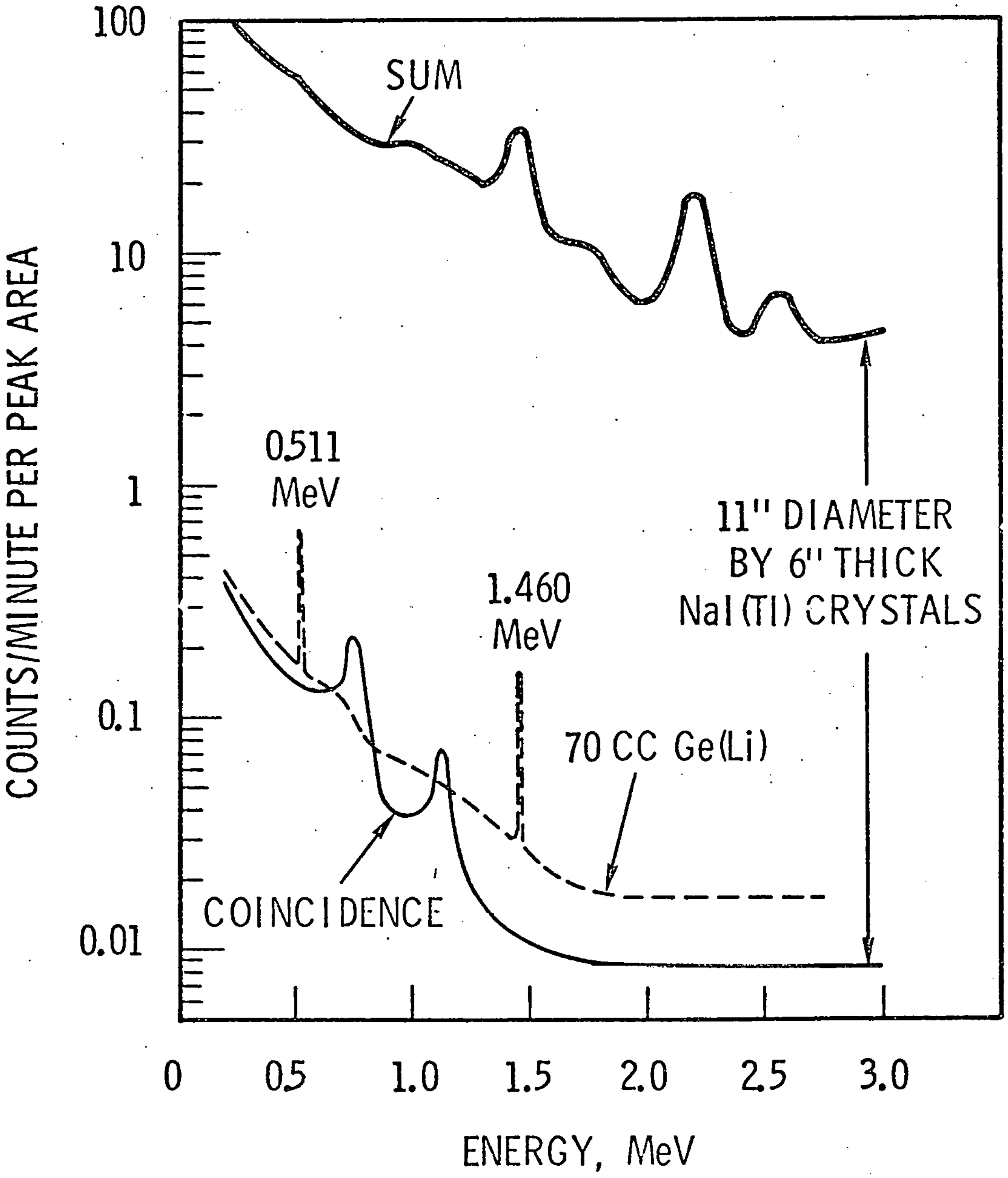

Fig. 16. Background counting rates of large NaI(Tl) and Ge(Li) detectors.

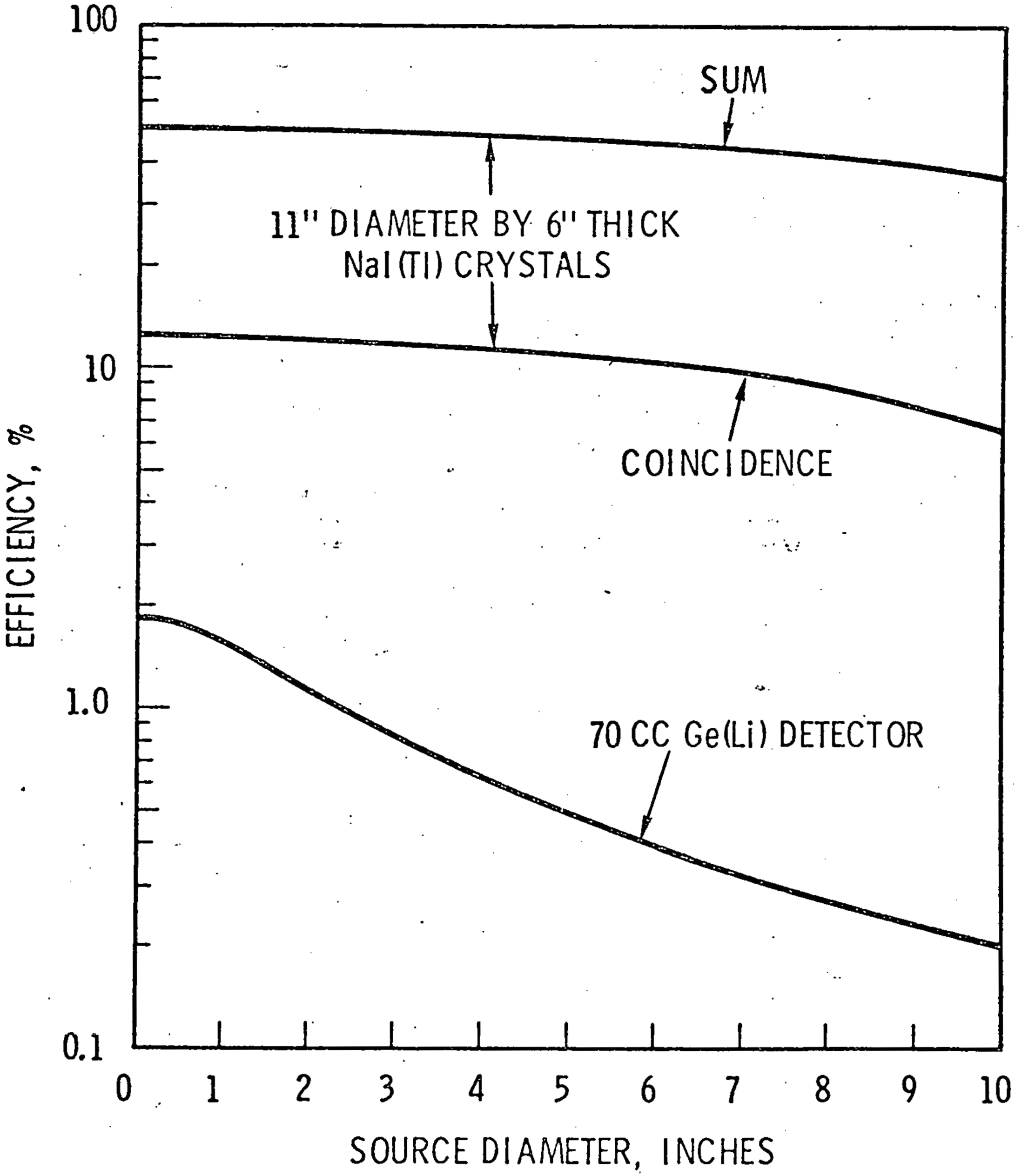

Fig. 17. Comparison of the counting efficiency of ^{60}Co as a function of source diameter for large NaI(Tl) and Ge(Li) detectors.

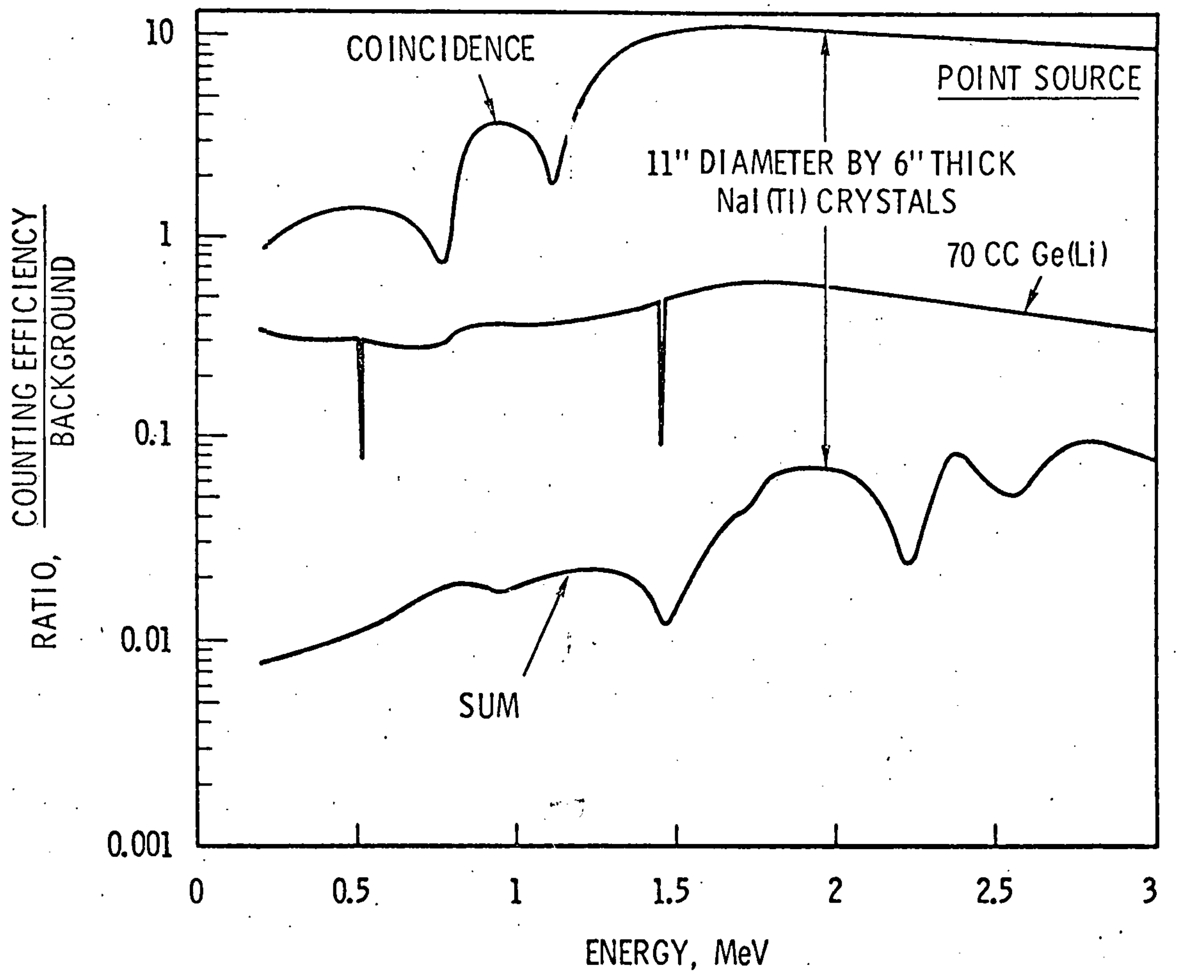

Fig. 18. Ratio of counting efficiency to background for large NaI(TlL and Ge(Li) detectors.