

## The Benefits of Using Super-Large Germanium Gamma-Ray Detectors for the Quantitative Determination of Environmental Radionuclides

R. M. Keyser, T. R. Twomey, and S. E. Wagner, ORTEC

*An experimental comparison of a number of large and super-large HPGe gamma-ray spectrometers demonstrates that, from the standpoint of either sample throughput or detection limit, the largest detectors provide a benefit greater than what might be expected from just their higher efficiency. At a given MDA — one 90% efficiency detector can count as many samples as four 30% efficiency detectors. Alternatively, one 90% detector, while processing the same number of samples as three 30% detectors, can achieve a significantly lower MDA on each sample. These advantages are tangible ones for the environmental spectroscopist, because larger detectors cost less per percent efficiency than smaller detectors.*

### INTRODUCTION

The gamma-ray spectroscopist — whether performing low-level environmental measurements, searching for rare events during in-beam experiments, or doing in-vivo bioassay — usually encounters the problem of maximizing peak counts while minimizing background counts.

In this note the marked and, perhaps, unexpected advantages of high-efficiency germanium detectors are discussed from the viewpoint of the environmentalist; however, the arguments are just as relevant for other detector users.

As larger detectors have become available, those performing environmental gamma spectroscopy have purchased progressively larger units in the belief that “bigger must be better.”

It is obvious that larger detectors must be able to accumulate the same number of counts in shorter times than smaller detectors, but what is the overall benefit? Is a single large detector a better solution than two detectors; each with one-half the efficiency? From the point of view of the user, how does the cost/benefit change with detector efficiency?

G. Bellia<sup>1</sup> determined experimentally that large Ge detectors, with their higher peak-to-Compton ratio, offer increased capability in the discrimination of a full-energy peak in a complex spectrum. He also noted that the more favorable peak-to-total ratio allowed a noticeable gain in the useful-to-useless event ratio in experiments involving multiple gamma-gamma coincidence events.

Cooper<sup>2</sup>, in a definitive paper, derived from first principles a formula for the “sensitivity” (or minimum detectable activity),  $D_m(E_1)$ , of a detector to radiation of energy  $E$  emitted by a specific radionuclide, in the presence of higher-energy emitters:

$$D_m(E_1) = \frac{A_m \left\{ \sqrt{2bR(E_1)[\bar{B}_{c2}(E_1) + \bar{B}_N(E_1)] + \frac{A_m^2}{4} + \frac{A_m}{2}} \right\}}{\epsilon(E_1) \cdot f_1 \cdot t} \quad (1)$$

Where:

$A_m$  is the reciprocal of the fractional error (often termed user-set sensitivity),

$b$  in channels/keV is the reciprocal of the gain, such that  $b \cdot R(E_1)$  equals the number of channels in the peak,

$R(E_1)$  is the resolution at energy ( $E_1$ ),

$\bar{B}_{c2}(E_1)$  is the Compton background interference at gamma-ray energy one ( $E_1$ ) from gamma-ray two at some higher energy. (If more than one gamma ray contributes to this background, then  $\bar{B}_{c2}(E_1)$  is the sum of the individual contributions),

$\bar{B}_N(E_1)$  is the natural background interference at the energy of  $\gamma_1$ ,

$\epsilon(E_1)$  is the peak efficiency of the detector for detecting gamma rays of energy  $E_1$ ,

$f_1$  is the fraction of disintegrations of the source which results in the emission of the gamma ray of energy  $E_1$ ,

$t$  is the measurement interval.

There are two cases that must be considered: the more common one, in which the source-induced background dominates, and the one in which the external background dominates.

Cooper noted that for the case in which the Compton background from other sources in the sample is much greater than the natural background (as is often the case with environmental samples), the sensitivity [ $D_m(E_1)$ ] is dominated by the term:

$$\frac{\{R(E_1)[\bar{B}_{c2}(E_1)]\}^{1/2}}{\epsilon(E_1)} \quad (2)$$

for a given  $A_m$ ,  $b$ , and  $t$ .

Cooper then observed that the Compton background at  $\gamma_1$  must be constant for a given source geometry intensity and a particular  $\gamma_2$ . He therefore proposed that it could be described by a Compton efficiency at  $E_1$  for  $\gamma_2$ :  $\epsilon_{c2}(E_1)$ .

Substituting this efficiency in Equation (2), Cooper proposed a figure of merit (FOM):

$$F_2(E_1) = \frac{\epsilon(E_1)}{[R(E_1)\epsilon_{c2}(E_1)]^{1/2}} \quad (3)$$

where  $F_2(E_1)$  is the detector FOM for detecting  $\gamma_1$  in the presence of  $\gamma_2$ .

The detector with the highest FOM will have the highest sensitivity (lowest limit of detection) for detecting gamma rays of energy  $E_1$  in the presence of higher-energy  $\gamma_2$ .

In general, the FOM allows the comparison of two detectors from the point of view of their detection limit for one nuclide in the presence of one or more higher-energy nuclides.

Examining the FOM, one sees:

(a) As one might expect, the FOM is proportional to the efficiency  $\varepsilon(E_1)$ , confirming that larger detectors do improve throughput.

(b) Since the resolution appears in the denominator under the square-root sign, the effect of improving resolution from 2.0 keV to 1.7 keV, for example, has only a modest effect on improving the FOM (~8%).

(c) The appearance of the Compton efficiency  $\varepsilon_{c2}$  in the denominator, inside the square root, is an interesting feature: a reduction in the Compton efficiency will lead to an increase in the FOM and in sample throughput. A reduction in the Compton efficiency is equivalent to increasing the detector peak-to-Compton ratio (p/C), since this translates into fewer Compton-scattered events being lost from the primary photopeak and contributing to background at lower energies.

In the case of ultra-low-level sources and in the case of higher energy lines not being present in the source being measured, the natural background from all sources (outside world, lead shield, and cryostat) may be the background that will determine the minimum detectable activity. To determine whether ultra-large detectors are advantageous in all situations, experimental consideration was given to both cases: (a) source background dominating and (b) natural background dominating.

## Experimental Details

Figure 1 shows a detector background spectrum of two 55% relative efficiency p-type coaxial germanium detectors. The measurement was made in a shield consisting of 20-cm low-background lead and 6-cm copper. While not completely “state of the art” from the standpoint of shielding typically used in environmental counting, it is representative.\*

Thirty detectors, covering a wide range of relative efficiency (10% to 100%) for  $^{60}\text{Co}$  at 1333 keV (defined according to IEEE Standard 325–1996) were placed, in turn, inside the shield, and background spectra (i.e., with no source present) accumulated for 100,000 seconds.

The non-peak background count-rate per channel per second was measured for each detector at 464 keV, 1445 keV, and 2335 keV. These results are plotted as a function of relative efficiency in Figures 2 (a) through (c). For each measurement, the background was determined from the average over 20 channels at the nominal energy specified.

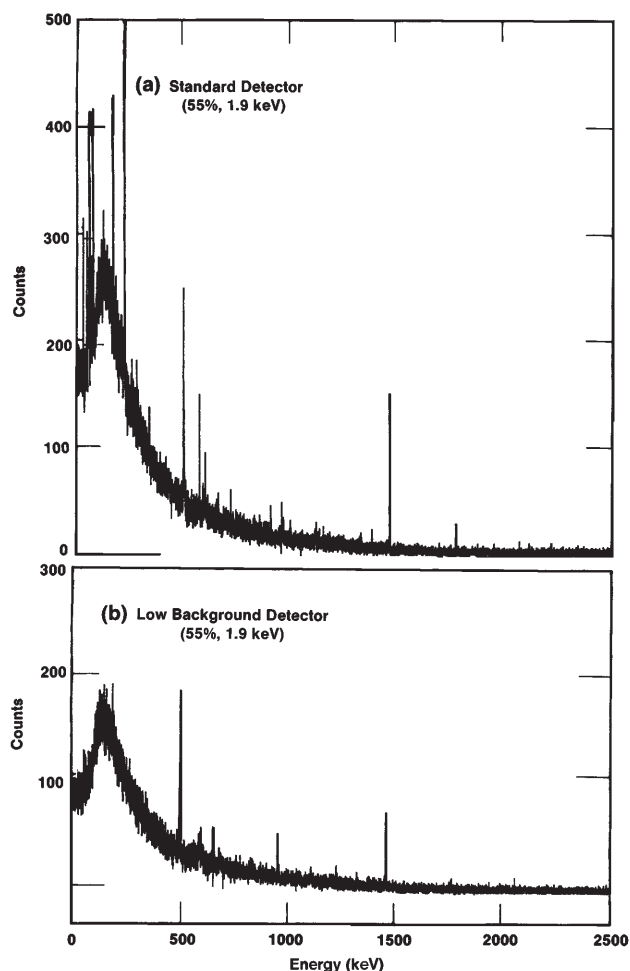


Fig. 1. Background Spectra for Two 55% Efficiency HPGe Detectors: (a) a “Standard” Cryostat and (b) a Low-Background Cryostat. (Spectra acquired for 100,000 seconds.)

On the same detectors, similar measurements were made of the non-peak background at 325, 540, 765, and 936 keV, resulting from the presence of a mixed  $^{152}\text{Eu}/^{154}\text{Eu}/^{125}\text{Sb}$  point-source calibration standard, placed 10 cm from the detector endcap. Non-peak background count-rate per channel per second vs. detector efficiency is plotted in Figure 3.\*\* Compare, for example, the count rate for a 90% detector to that of a 30% detector at each energy. Non-peak background count-rate per channel per second **per percent efficiency** is

\*Since the source of the 1460.75 keV ( $^{40}\text{K}$ ) peak is external to the cryostat, one would not expect a reduction in its size simply from using low-background cryostat materials; a lead back-shield inside the cryostat is the cause of the reduction.

\*\*This background is related to the total background under a peak by the peak width. In the 300-keV region, the detector FWHM is ~1.1 keV (~4 channels at the 0.33 keV/channel conversion gain employed); in the 936-keV region, the detector FWHM is ~1.5 keV (~5-1/2 channels).

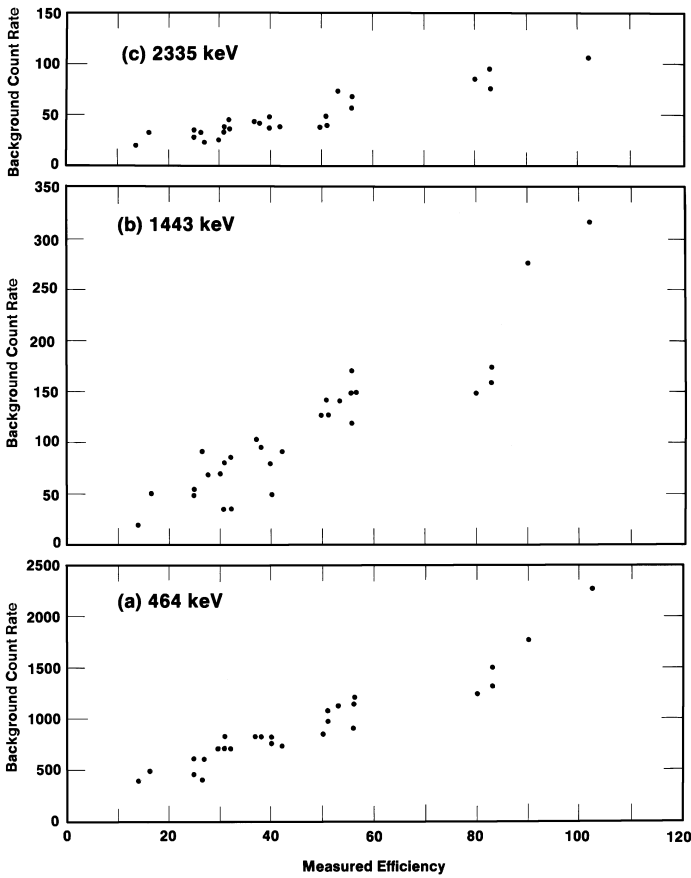


Fig. 2. Three 100,000-Second Background Spectra Count Rate vs. Detector Relative Efficiency at (a) 464 keV, (b) 1443 keV, and (c) 2335 keV. (Conversion gain is 0.33 keV/channel.)

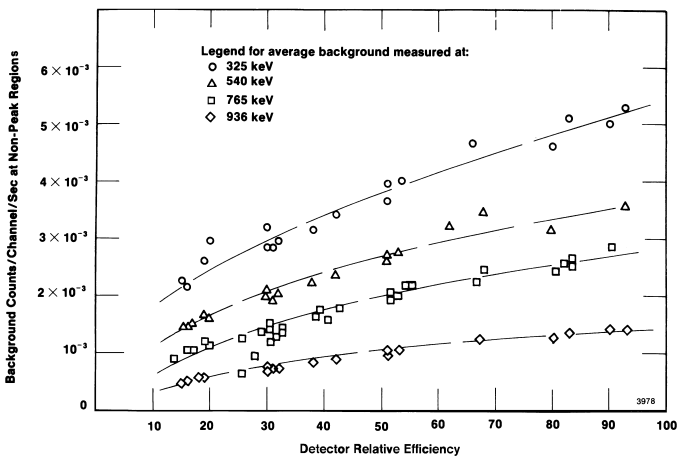


Fig. 3. Background Count Rates in the Presence of a Mixed  $^{152}\text{Eu}/^{154}\text{Eu}/^{125}\text{Sb}$  Source vs. Detector Efficiency. (Conversion gain is 0.33 keV/channel.)

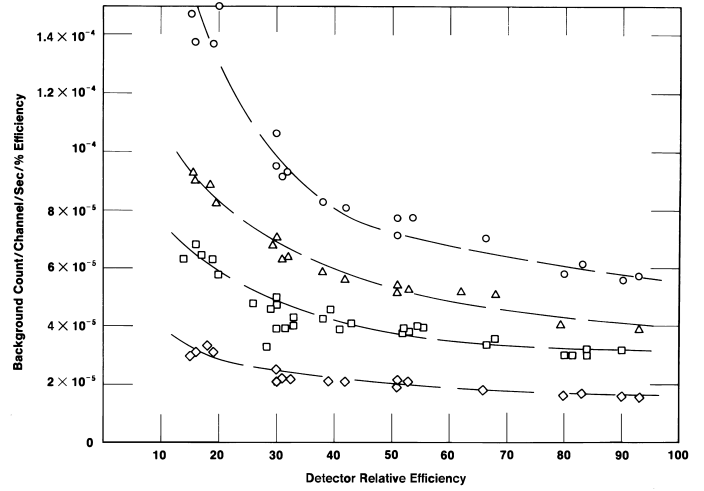


Fig. 4. Background Count Rates per Percent Efficiency in the Presence of  $^{152}\text{Eu}/^{154}\text{Eu}/^{125}\text{Sb}$  Source vs. Detector Efficiency. (Conversion gain is 0.33 keV/channel.)

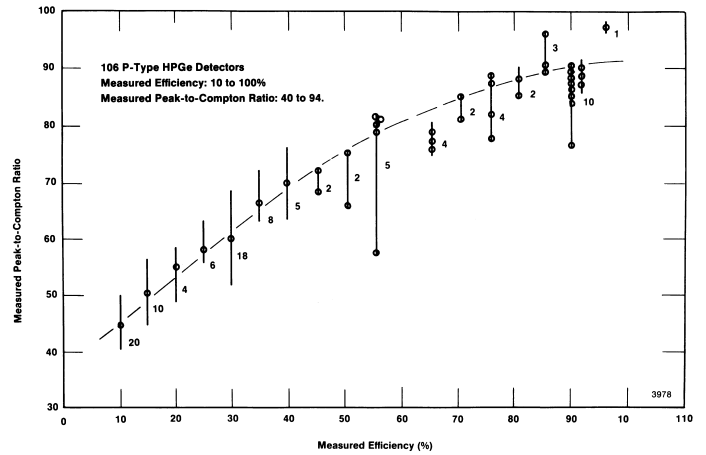


Fig. 5. Peak-to-Compton Ratio vs. Efficiency for 106 P-Type Coaxial Detectors.

plotted vs. detector efficiency in Figure 4. Note that at each energy this non-peak background count rate decreases **per percent of detector efficiency**.

Figure 5 shows the increase of measured peak-to-Compton ratio vs. efficiency for 106 detectors<sup>3</sup> recently manufactured by ORTEC. Compare, particularly, the 90% detector's peak-to-Compton ratio with that of the 30% unit.

The FOM in Equation (3) increases with improved performance. But since, in a discussion of detection limits, it is customary to refer to minimum detectable activity (MDA), the inverse of the FOM has been calculated for these measurements. Since the Compton efficiency is proportional to the background, the relative MDA,  $MDA_R$  is:

$$MDA_R = \frac{[R(E_1)B(E_1)]^{1/2}}{\epsilon(E_1)} \quad (4)$$

where we use the experimentally determined background  $B(E_1)$  instead of the Compton efficiency.

Cooper<sup>4</sup> determined FOMs for a wide range of germanium detector types; in all cases the FOM for 662 keV and 1333 keV were within 10% of each other. On this basis, the background measured at 765 keV obtained from the thirty-detector experimental group was chosen to calculate an  $MDA_R$ , in relative units, as a function of measurement interval,  $t$ . Curves of relative MDA vs. relative counting time were plotted (Figure 6) for three representative detector efficiencies: 30%, 60%, and 90%. Note that these curves are generated from averages of the MDA obtained for several detectors of each efficiency.

## Discussion of Results

Figures 2 (a) through (c), 3, 4, and 6 show that, as the relative efficiency at 1333 keV increases, the non-peak background **per percent efficiency** decreases. Reasoning simply that the higher the efficiency, the greater the number of events in the spectrum, a conclusion might have been drawn that the

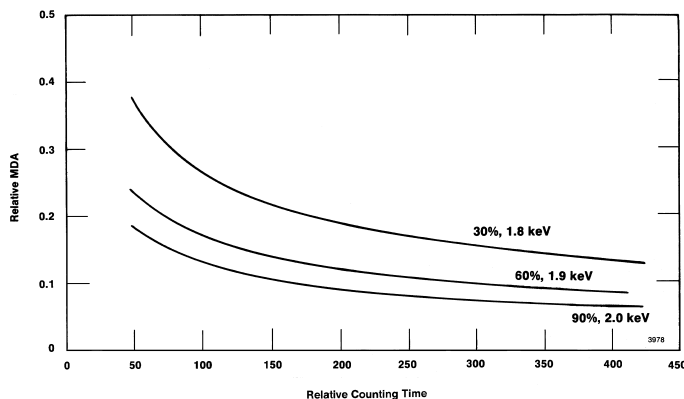


Fig. 6. Relative Detection Limit (MDA) vs. Counting Time at 765 keV for Typical 30, 60, and 90% Ge Coaxial Detectors.

background would be directly proportional to the efficiency. The slight degradation of detector resolution with increasing efficiency also points in the same direction. However, the experimental evidence contradicts this. **Clearly, this must be a consequence of the increase in peak-to-Compton ratio (p/C) with increasing relative efficiency.** (See Figure 5.)

For typical shielding configurations, the fact that the background, measured with no source present, decreases per percent of detector efficiency means that the most dominant background component is Compton scatter from specific lines rather than from a cosmic background continuum.

*Note: There is, of course, a corresponding increase, with increasing detector size, of the background photopeaks themselves, and therefore it is clear that for the determination of low levels of nuclides normally present in Ge detector backgrounds, or for the determination of low levels of nuclides with photopeaks close (within about 1 keV) to such peaks, a detector fabricated from selected low-background materials and a quality lead shield are desirable.*

The  $MDA_R^*$  defined in Equation (4) differs from the reciprocal of Cooper's figure of merit  $F_2$  in the use of "background," as in Equation (2), rather than "Compton efficiency," as in Equation (3). For convenience, the two background terms in Equation (1) have been summed as total background  $B(E_1)$ .

Figure 7 shows, at a fixed counting time, how relative MDA relates to detector efficiency. Since a user knows the MDA that can be obtained — for a particular counting time with his present detector — the curve allows a quick estimate of the improvement in MDA that can be expected by increasing the efficiency to any particular larger value.

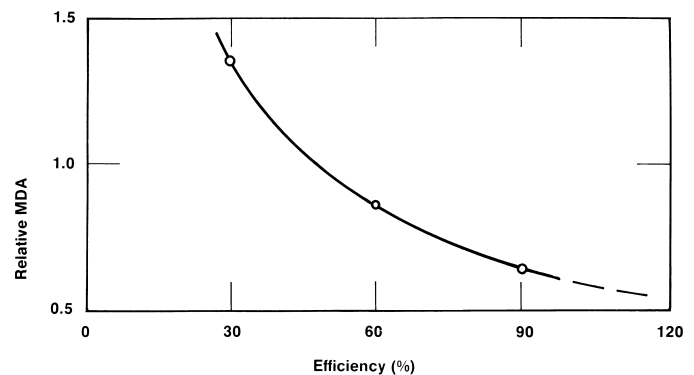


Fig. 7. Relative MDA, for 765 keV, vs. Efficiency for Fixed Counting Times.

\*The expression for  $MDA_R$  is consistent with widely-used definitions such as (a) the  $L_c$  defined by Currie<sup>5</sup> and (b) the KTA definition of detection limit<sup>6</sup> employed by the Nuclear Power plant regulatory agency in the Federal Republic of Germany. The definition of  $L_c$  is:

$$L_c = 2.33 [R(E_1) \cdot B(E_1)]^{1/2} \epsilon(E_1) \cdot f_1 \cdot t;$$

the definition of  $MDA_{KTA}$  is:  $MDA_{KTA} = [R(E_1) \cdot B(E_1)]^{1/2} \epsilon(E_1) \cdot f_1 \cdot t$ , where the symbols have their previous meanings.

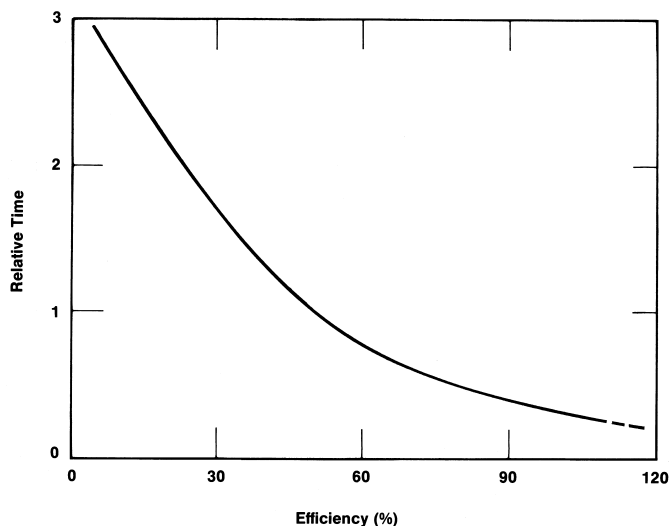


Fig. 8. Relative Time vs. Efficiency, for 765 keV, at Fixed MDA.

Figure 8 shows, for a fixed MDA requirement, the counting time decrease with increasing efficiency. Thus, this curve allows estimating the reduction in counting time (i.e., relative throughput increase) that can be expected by increasing the efficiency to a particular value.

[The term “relative throughput” is used because the reciprocal of the measurement interval for a given MDA is proportional to the number of samples that can be processed to a given detection limit in, for example, one day or one week.]

Figure 9 displays relative sample throughput (for fixed MDA, at 765 keV) vs. efficiency for multiple 30% detectors and single 60% and 90% detectors. Figure 10 shows typical relative list price (normalized at 30% efficiency) per percent efficiency: the larger the detector, the lower the price per percent efficiency.

Figures 9 and 10 lead to the conclusion that, apart from the question of detector redundancy, multiple low-efficiency detectors are not a good choice for processing large numbers of samples. **A single large detector costs less than several smaller detectors of the same total efficiency and yet permits greater sample throughput.**

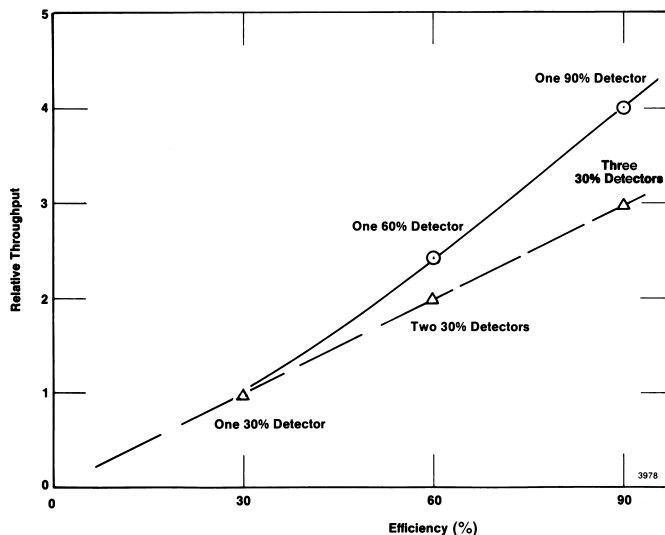


Fig. 9. Relative Sample Throughput at 765 keV, vs. Efficiency.

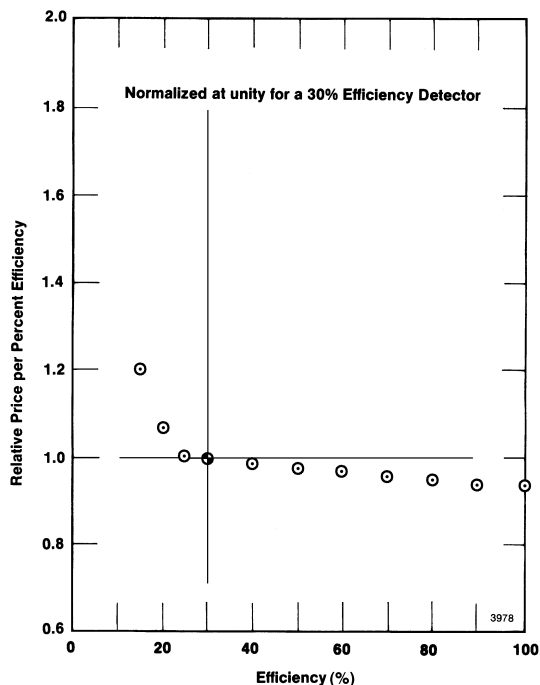


Fig. 10. Relative Price per Percent Efficiency vs. Efficiency.

## Applicability at Lower Energies?

This discussion of relative throughput has been based on the presumption that the line of interest to be quantified is ~765 keV. At substantially lower energies (~<350 keV), the greater diameter of higher-efficiency detector crystals is valuable, but the greater length is of less value. This is because the crystal is black to lower-energy radiation (there is more than enough detector thickness to stop virtually all the incident low-energy photons). However, the “extra” germanium continues to reduce the Compton background at all energies; moreover, at lower energies, there is more Compton background to reduce because there are progressively more higher-energy peaks that are contributing to the background.

At energies around 300 keV, the improvement is less: ~10% additional throughput when a single 90% detector is compared to three 30% detectors. However, since the cost of the 90% detector is ~10% less than the cost of three 30% detectors — even without the added electronics cost — it is clear that, at any energy, there is a substantial advantage in cost per sample processed when using a single large detector.

## Summary

Lower detection limits can be obtained with higher-efficiency Ge detectors than with smaller ones. A single large detector can result in substantially higher sample throughput than multiple smaller detectors of the same total efficiency. This translates directly into cost-per-sample saving from the viewpoint of operating an environmental laboratory.

## References

- <sup>1</sup>G. Bellia et al., “Performances of Large Volume p-Type HPGe Detectors,” *Nuclear Instruments and Methods in Physics Research* **A284** (1989) 399–404.
- <sup>2</sup>J.A. Cooper, “Factors Determining the Ultimate Sensitivity of Ge(Li) Gamma-Ray Spectrometers,” *Nuclear Instruments and Methods in Physics Research* **82** (1970) 273–277, North Holland Publishing Company.
- <sup>3</sup>T.J. Paulus and R.M. Keyser, “Enhancement of Peak-to-Total Ratio in Gamma-Ray Spectroscopy,” *Nuclear Instruments and Methods in Physics Research* **A286** (1990) 364–368, Amsterdam: North Holland Publishing Company.
- <sup>4</sup>J.A. Cooper, “Figure-of-Merit Measurements for Ge(Li) Detectors,” *Nuclear Instruments and Methods in Physics Research* **94** (1971), 289–296, North Holland Publishing Company.
- <sup>5</sup>Lloyd A. Currie, “Limits for Qualitative Detection and Quantitative Determination: Application to Radiochemistry,” *Analytical Chemistry*, **40**, No. 3, (1968), 586–593.
- <sup>6</sup>“The Limit of Detection Concept,” <<Kerntechnischer Ausschuss (KTA), Messung und Überwachung der Ableitung gasförmiger und aerosolgebundener radioaktiver Stoffe, Fassung: 2/79 Teil 1 KTA 1503.1>>.