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Counting efficiency in gamma-ray spectrometry with different sample volumes for the same geometry

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ABSTRACT

The aim of this study is to determine an easy and fast method to calculate efficiencies in different volumes, in the same counting geometry, for gamma-ray spectrometry technique. Reference Material Soil IAEA 326 was packed in a 100 mL capacity polyethylene bottle with different masses and volumes, and sealed for about four weeks, prior measurement, in order to ensure that radioactive equilibrium had been reached between ^{226}Ra and its progeny. After this time, they were measured by gamma-ray spectrometry with a hyper-pure germanium detector. The masses of the reference material used were 25, 60, 80, 95 and 128g. The energies of gamma-rays used in this paper are recommended due to the considerations: gamma intensity value, peak quality, spectral region without interference and the gamma- ray energies of the ^{238}U and ^{232}Th series very important for determining the natural radioactivity. The efficiency values obtained compared to the adjusted efficiency values were similar and presented a good correlation coefficient. The performance was acceptable for all different masses studied, indicating results consistent for the method. The proposed method could be useful as a tool for laboratories, dealing with of samples on a routine basis, by reducing the cost on the purchase of another counting geometry and optimizing the use of the detection system, thus improving their performance.

Keywords: **gamma-ray spectrometry, counting efficiency, environmental samples.**

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1. INTRODUCTION

The technique of gamma-ray spectrometry is a powerful tool for determining the contents of gamma-ray emitter radionuclides in environmental samples (soil, sediment, water, and biological samples). As the result of applying this technique, a count vs. energy spectrum is obtained [1].

To ensure that high-quality spectra are obtained, good practices for their acquisition must be established, including physical setup (detector, shielding and appropriate laboratory), electronic settings (associated electronics), counting conditions, correction for the background radiation in the laboratory (corrections for unwanted sources of radiation) [2] and validation by proficiency tests.

The fundamental step in gamma-ray spectrometry technique is obtaining a correct efficiency curve, which basically depends on the counting geometry (geometric configuration of the container used for conditioning samples), solid angle (position of the counting geometry relative to the detector), quantity (volume of sample used in the counting geometry) and density of the sample [3].

For the gamma-ray spectrometry technique, it is necessary to know very well the efficiency of the measurement system, that is usually obtained through calibrated standards prepared in the same geometry used in the samples counting. Widely used standards for the efficiency curve are certified reference materials for soil, sediment, vegetation, among other standards. Another option for the laboratory is the preparation of a standard calibration solution, which consists of diluting a standard radioactive solution with known activity of the interest radionuclides using the counting geometry of the samples [4]. Monte Carlo code can also be used to obtain calibration efficiency in the gamma spectrometry technique.

Depending on the condition of the sample to be analyzed, it is necessary to reduce the volume of samples for the counting geometry. The conditions are high density, sample activity, insufficient amount of sample, among other factors. Samples with high density makes it difficult to determine self-absorption, resulting in an incorrect counting; high activity in the sample increases the dead time of the detection system, impairing the resolution of the spectrum peaks and increasing the uncertainty in determining the activity; the insufficient amount of sample would be for specific cases when the mass is insufficient for the counting geometry used. For these types of samples described, a specific efficiency curve that considers the sample mass is required. In case the sample to be analyzed has a density and composition different from the certified reference material used in the efficiency curve, it is necessary to correct the self-absorption in the result obtained [5]. The dependence on efficiency is directly related to the volume, density and composition of the sample, which can be easily and quickly corrected with the self-absorption test [4].

The aim of this study is to determine an easy and fast method to calculate efficiencies for different volumes in the same geometry.

2. MATERIALS AND METHODS

The geometry used was a 100 mL capacity polyethylene bottle (F100), a geometry well known and tested in several intercomparisons, in our laboratory. Reference Material Soil IAEA 326 was packed in the F100 bottle in different masses, ranging from 25g to 128g, to obtain different sample volumes from 19.5mL to 100 mL and sealed for about four weeks prior to measurement in order to ensure that radioactive equilibrium had been reached between ^{226}Ra and its progeny. Table 1 presents the masses and volumes of the reference material samples in the F100 geometry used in determining the efficiency curves.

Table 1. Mass and volume of reference material in F100 geometry

mass (g)	volume (ml)
25	19.5
60	46.9
80	62.5
95	74.2
128	100

After this time, the bottles with the reference material were measured by gamma-ray spectrometry with a hyper-pure germanium detector (HPGe) Canberra model XtRa, 25% relative efficiency with associated electronics and coupled to a microcomputer. Multichannel Maestro A65-I model software [6] was employed for spectrum acquisition. Interwinner 6.0 from Eurisys Measurements Incorporation [7] software was used for personal computer analysis of gamma-ray spectra from HPGe detector and for making the efficiency curve. The blank samples for background determination were prepared with super pure water.

The performance of the gamma-ray spectrometry measurements was evaluated by participation in proficiency tests for Brazilian laboratories, organized by Instituto de Radioproteção e Dosimetria (IRD), which is available on a routine basis [8].

3. RESULTS AND DISCUSSION

The efficiency was measured for a HPGe detector using reference material soil IAEA 326. The energies of gamma-rays used in this paper are recommended by considering: gamma intensity value, peak quality, and spectral region without interference [9]. Selected gamma-ray energies were: 46.5 keV from ^{210}Pb, 295.2 and 351.9 keV from ^{214}Pb and 609.3 keV from ^{214}Bi of the ^{238}U series; for the ^{232}Th series gamma-ray emissions were: 238.6 keV from ^{212}Pb and 911.1 keV from ^{228}Ac.

Table 2 shows results obtained in efficiencies, efficiencies adjusted by the linear regression and difference between efficiencies for energy of interest radionuclides from the of the ^{238}U and ^{232}Th series.

Table 2. Efficiencies, efficiencies adjusted and difference between efficiencies for energy of interest

	46.5 keV (^{210}Pb)			**238.6 keV (^{212}Pb)**			**295.2 keV (^{214}Pb)**		
Mass	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %
25 g	13.143±1.314	12.657±0.465	-3,7	5.340±0.323	5.189±0.130	-2.8	3.391±0.207	3.399±0.089	0.2
60 g	9.018±0.902	9.794±0.288	8,6	4.330±0.262	4.444±0.081	2.6	3.147±0.192	3.028±0.056	-3.8
80 g	8.012±0.801	8.158±0.258	1,8	3.902±0.236	4.018±0.072	3.0	2.679±0.163	2.816±0.050	5.1
95 g	7.225±0.723	6.931±0.287	-4,1	3.629±0.220	3.698±0.080	1.9	2.643±0.161	2.657±0.056	0.5
128 g	4.374±0.437	4.316±0.446	-3,3	3.142±0.190	2.995±0.126	-4.7	2.353±0.143	2.308±0.087	-1.9
	351.9 keV (^{214}Pb)			**609.3 keV (^{214}Bi)**			**911.2 keV (^{228}Ac)**		
Mass	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %
25 g	3.087±0.192	3.024±0.120	-2.0	1.581±0.104	1.588±0.058	0.5	1.271±0.062	1.322±0.055	4.0
60 g	2.745±0.171	2.674±0.076	-2.6	1.474±0.097	1.392±0.036	-5.5	1.243±0.060	1.154±0.034	-7.2
80 g	2.306±0.171	2.474±0.068	7.3	1.236±0.097	1.280±0.033	3.6	1.082±0.060	1.058±0.031	-2.2
95 g	2.232±0.143	2.324±0.076	4.1	1.136±0.081	1.196±0.037	5.3	0.941±0.052	0.986±0.035	4.8
128 g	2.126±0.139	1.994±0.117	-6.2	1.061±0.075	1.012±0.057	-4.7	0.824±0.046	0.828±0.054	0.4

Fig. 1 presents the fitting equations using linear regression of the counting efficiencies, with mass ranging from 25 to 128 g for each energy studied.

(^{210}Pb) **(^{212}Pb)**

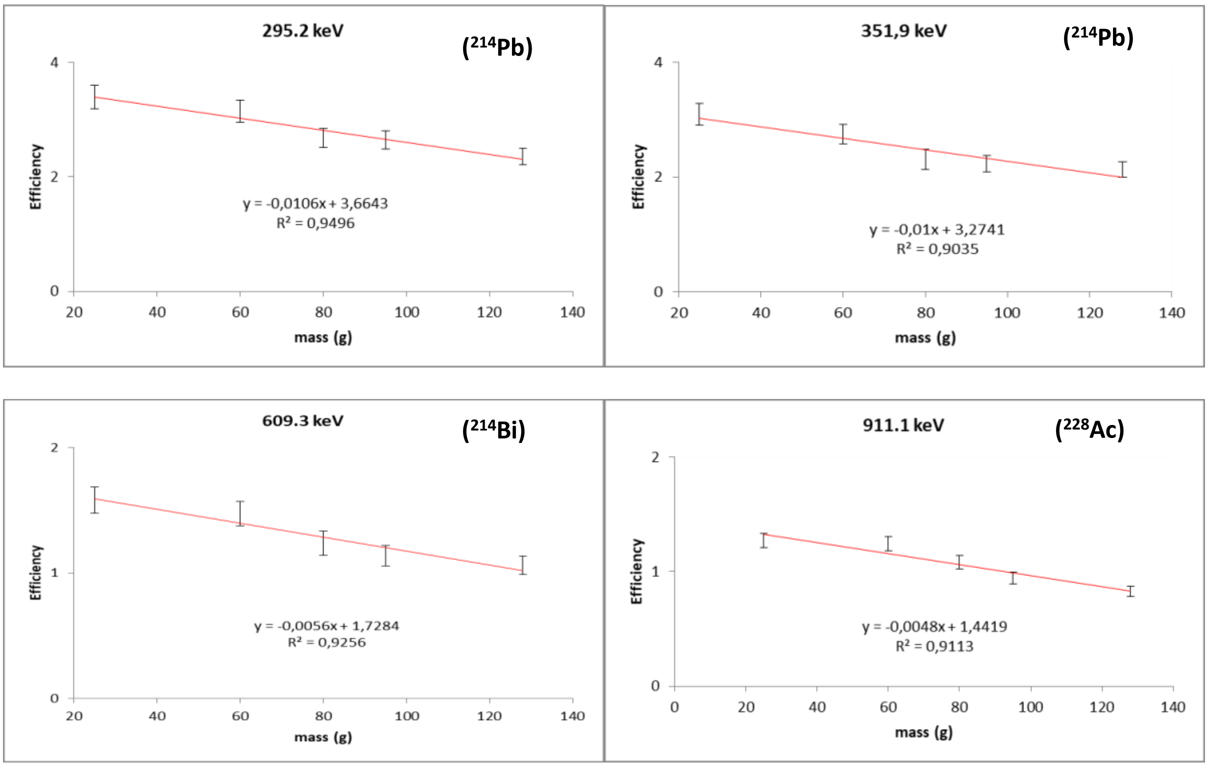

Fig. 1. Linear regression considering the range of mass from 25 to 128 g

The efficiency values obtained compared to the adjusted efficiency values were similar and showed a good correlation coefficient. The uncertainty of the adjusted efficiency was estimated using the correlation between the linear coefficients [10]. Table 3 presents the results obtained of the efficiencies and efficiencies adjusted by the exponential decay (first order) equation and difference between efficiencies for each quantity of sample studied.

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Table 3. Efficiencies, efficiencies adjusted and difference between efficiencies in different sample quantities.

	25 g			**60 g**			**80 g**			**95 g**			**128 g**		
keV	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %	Eff %	Eff % $_{\text{adjust}}$	Diff. %
238.6	5.340±0.323	4.945±0.106	-7.4	4.330±0.262	4.216±0.102	-2.6	3.902±0.236	3.773±0.080	-3.3	3.629±0.220	3.560±0.076	-1.9	3.142±0.190	3.081±0.082	-1.9
295.2	3.391±0.207	3.742±0.101	10.3	3.147±0.192	3.295±0.100	4.7	2.679±0.163	2.831±0.077	5.7	2.643±0.161	2.736±0.075	3.5	2.353±0.143	2.472±0.081	5.0
351.9	3.087±0.192	2.931±0.098	-5.1	2.745±0.171	2.654±0.099	-3.3	2.306±0.171	2.215±0.075	-3.9	2.232±0.143	2.167±0.074	-2.9	2.126±0.139	2.021±0.081	-4.9
609.3	1.581±0.104	1.540±0.095	-2.6	1.474±0.097	1.474±0.098	0.0	1.236±0.097	1.233±0.073	-0.2	1.136±0.081	1.140±0.074	0.3	1.061±0.075	1.074±0.081	1.2
911.2	1.271±0.062	1.298±0.095	2.1	1.243±0.060	1.236±0.098	-0.6	1.082±0.060	1.087±0.073	0.4	0.941±0.052	0.940±0.074	-0.1	0.824±0.046	0.819±0.081	-0.6

Figure 2 presents the adjustment used by the exponential decay (first order) equation considering the energy range from 238.6 to 911.2 keV.

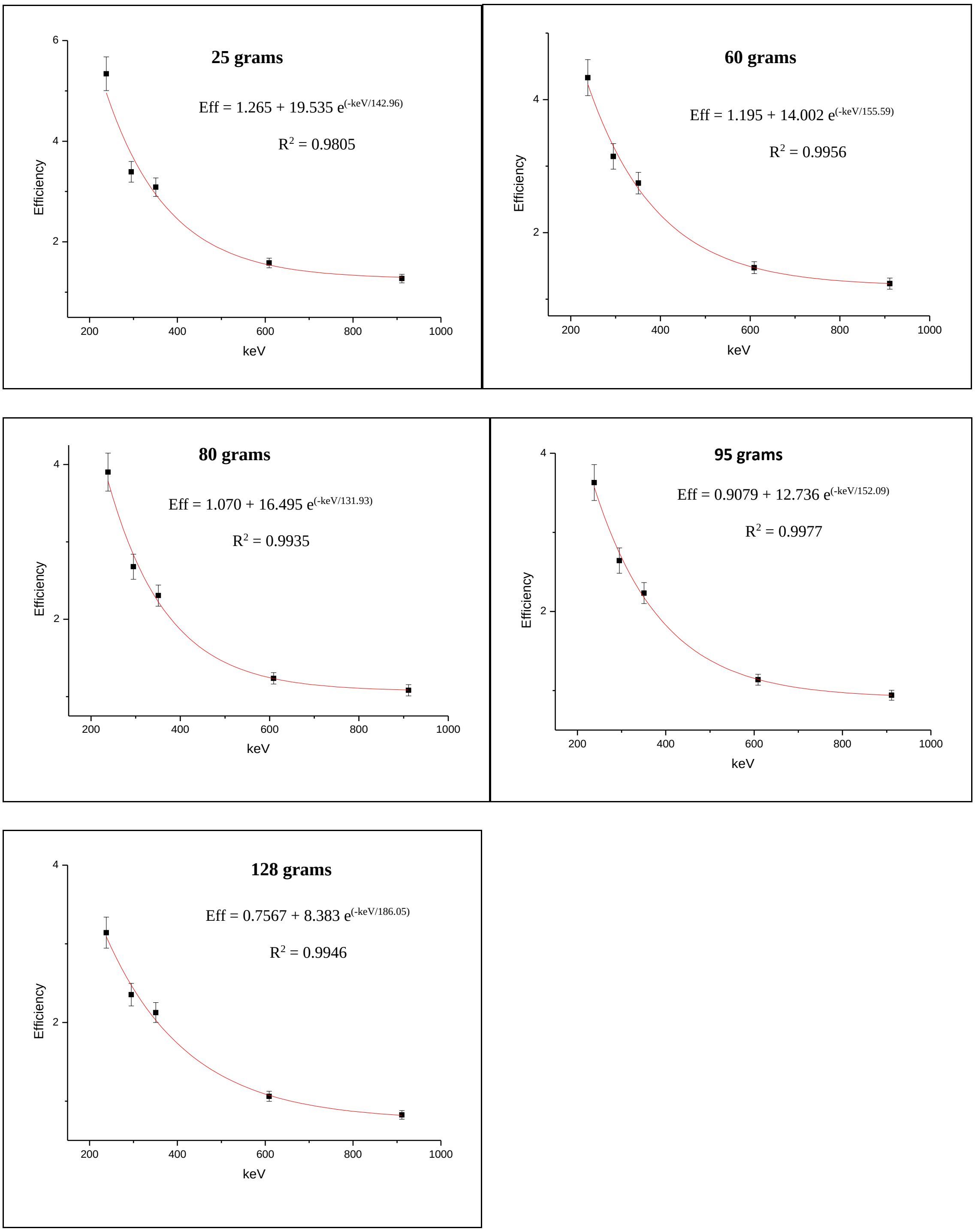

Fig. 2. Exponential decay (first order) equation with energy range from 238.6 to 911.2 keV.

The efficiency values obtained compared to the adjusted efficiency values were similar and the graphs showed a good correlation coefficient. The uncertainty of the adjusted efficiency was estimated using uncertainty propagation.

The blank sample was used to determine the minimum detectable activity (MDA) "*a priori*" and was calculated by the model proposed by Currie [11]. The counting time of 24 h was used for each sample mass and energies of interest are shown in Figure 3.

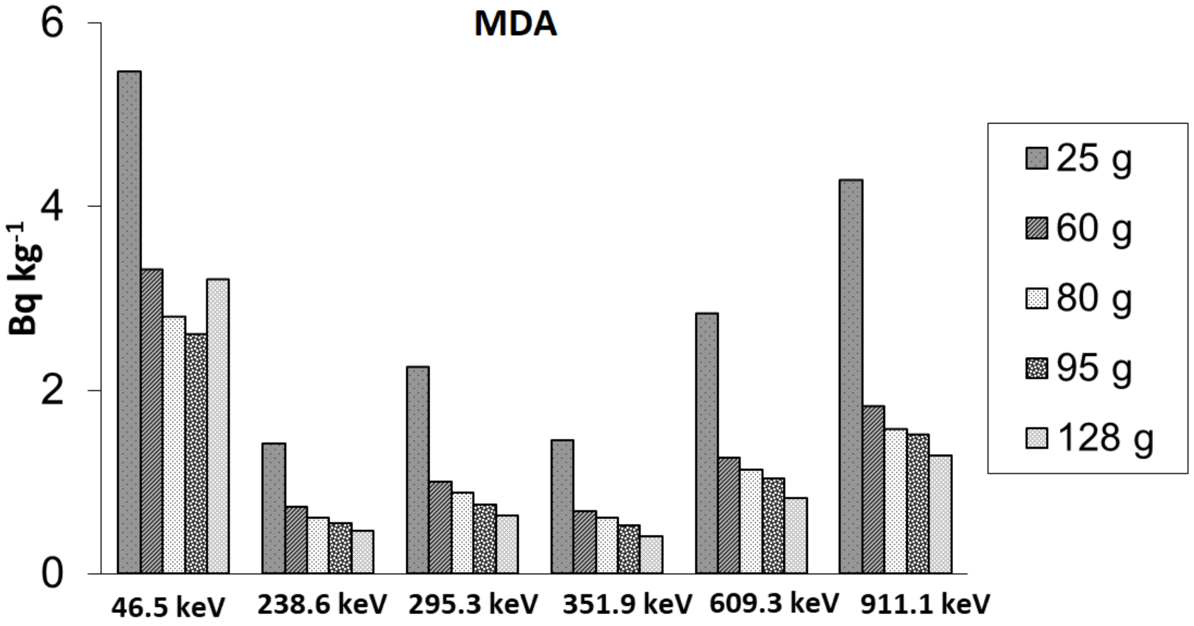

Fig. 3. MDA "a priori" for the energies of interest, calculated from the blank samples spectra

When the MDA is higher than desired, it is necessary to increase the counting time with time optimization properly. Counting time optimization is the fastest, economical alternative and with a good accuracy to solve the problem [3,12,13,14,15]. The technique to be chosen to measure the radionuclide it has to be appropriate for the type of radiation and sensitive to demonstrate compliance with the criteria.

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The performance (precision, accuracy and normalized deviation) of the method was verified using the ^{137}Cs (661.6 keV) [16] and comparing the presented value in the intercomparison report [17].

The precision was evaluated by the equation (1).

$$RSD = \frac{SD}{AV}.100 \qquad (1)$$

Where: *RSD* is the relative standard deviation,

SD is the standard deviation,

AV is the average value.

The accuracy was evaluated by the equation (2).

$$RE = \frac{|AV - RV|}{RV}.100 \qquad (2)$$

Where: *RE* is the relative error,

RV is the reference value,

AV is the average value.

The criteria used in the performance evaluation are the normalized deviation (E), when $|E| < 1$ the result is consistent [8]. The normalized deviation, E, was evaluated by the equation (3).

$$E = \left(\frac{(X - U)}{k\sqrt{Sx^2 + Su^2}} \right) \qquad (3)$$

Where: *E* is the normalized deviation,

X is the laboratory mean value,

Sx is the standard deviation of the laboratory,

U is the reference value,

Su is the standard deviation of the reference value,

k is the coverage factor (k=2).

Table 4 shows the performance of the method using Cs-137 for all analyzed masses. The accuracy and precision of the method was acceptable for all different studied masses. The values obtained for the normalized deviation (E) were always below one, indicating results consistent for the method.

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Table 4. Performance of the method: relative standard deviation (RSD), relative error (RE) and normalized deviation (E)

^{137}Cs (137.5 ± 18.6 Bq kg^{-1})				
Mass (g)	**Bq kg^{-1}**	**RE**	**RSD**	**E**
25	151±10	9.8	6.5	0.32
60	130±9	-5.8	6.6	-0.20
80	127±8	-7.8	6.5	-0.26
95	138±9	0.5	6.3	0.02
128	127±8	-7.3	6.5	-0.25

4. CONCLUSION

This paper aimed to propose an easy and fast method for determining efficiencies with different volumes for the same geometry; it becomes important when the samples have high density, sample activity, insufficient sample mass, among other factors.

The energies of gamma-rays used in this paper are recommended considering proper gamma intensity value, peak quality, spectral region without interference and gamma-ray energies of the radionuclides from the ^{238}U and ^{232}Th decay series, that are very important for determining natural radioactivity in environmental samples.

The efficiency values obtained, when compared to the adjusted efficiency values, were similar and the plots showed a good correlation coefficient. The performance was acceptable for all different masses studied, indicating consistent results for the method.

The proposed method could be useful as a tool for laboratories, dealing with samples on a routine basis, by reducing the costs on the purchase of additional counting geometry and optimizing the use of the detection system, thus improving their performance.

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