COMPTON SUPPRESSION GAMMA RAY SPECTROMETRY

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Abstract. In the past decade there have been many studies to use Compton suppression methods in routine neutron activation analysis as well as in the traditional role of low level gamma ray counting of environmental samples. On a separate path there have been many new PC based software packages that have been developed to enhance photopeak fitting. Although the newer PC based algorithms have had significant improvements, they still suffer from being effectively used in weak gamma ray lines in natural samples or in neutron activated samples that have very high Compton backgrounds. We have completed a series of experiments to show the usefulness of Compton suppression. As well we have shown the pitfalls when using Compton suppression methods for high counting deadtimes as in the case of neutron activated samples. We have also investigated if counting statistics are the same both suppressed and normal modes. Results are presented in four separate experiments.

Influence of dead time in Compton suppression NAA

Background

Compton suppression spectrometry has developed into an established position in low-level counting. It is attractive because of the reduction in the Compton continuum, and cosmic and natural background. Usually Compton suppression methods have been used in environmental studies with low-level activities and subsequent low dead times. In neutron activation analysis (NAA), Compton suppression can help to improve precision and accuracy for isotope identification and also substantially reduce spectral interferences. In general, NAA deals with a much broader range of dead times. The principle of the system is based on the Compton effect described as follows. When a gamma ray interacts with the main detector, the Compton effect may occur, in which a recoil electron and a scattering photon are created sharing the initial gamma ray energy. The recoil electron has a short range and deposits its energy in the main detector, while the scattered photon is more likely to escape the main detector. In a normal detection system, the signal from the recoil electron is recorded as a contribution to the baseline since the energy of the recoil electron is lower than the original gamma ray energy. In the Compton system, photons passing the main detector are detected by the surrounding NaI(T*l*) detector. If both the main and NaI(T*l*) detectors record the signals within a specific time interval, the signal is eliminated under the assumption that the signals result from Compton scattering. Through this procedure, the baseline of Compton continuum in the spectrum is reduced to a level much lower than in a normal spectrum, and the analytical sensitivity is drastically improved.

Experimental

The Compton suppression gamma ray detection system used in the study consists of a main germanium detector which has an 18% relative efficiency with 1.9 keV resolution for a 1332 keV photopeak of ^{60}Co, a large NaI(T*l*) crystal ring detector outside the main detector, and an ORTEC ADCAM PC based multi-channel analyzer. The diagram of the system is shown in Figure 1 and a detailed description of the system has been presented elsewhere [1]. The detection system can measure a sample in both normal and Compton modes simultaneously.

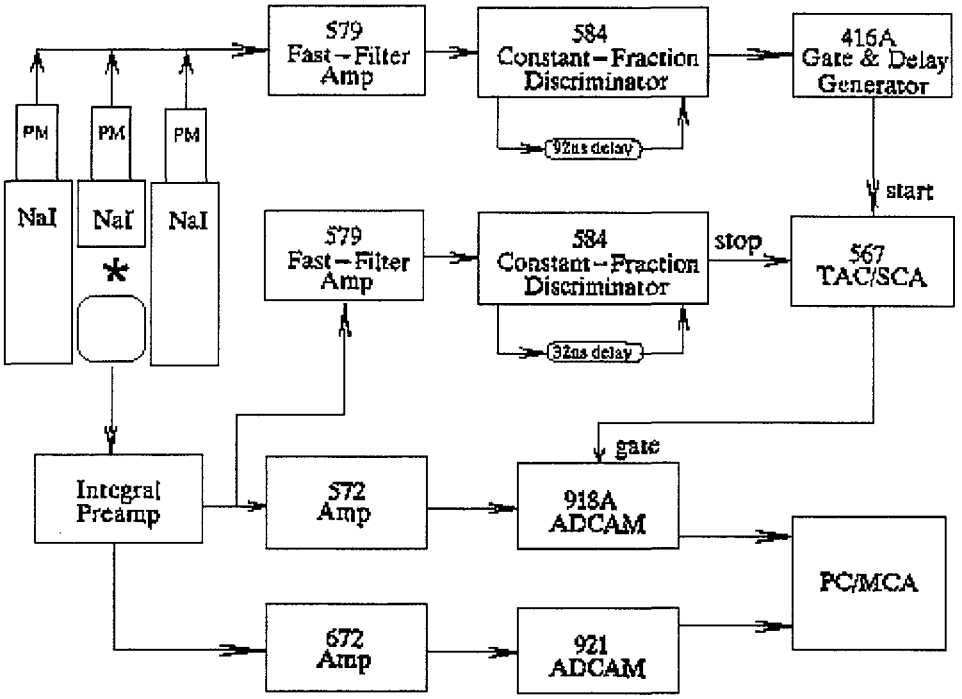

Fig. 1. Schematic diagram of the Compton suppression gamma ray counting system.

The Compton suppression system can reduce the continuum quite significantly, usually by a factor of 5 to 10. This is useful especially for low level elemental analysis. However, this advantage can only be taken if the radionuclide has a single or one major gamma ray, such as ^{65}Zn(1115keV), and ^{58}Co (810keV) for Ni analysis, and gamma rays which are not coincident with each other. The Compton suppression system can significantly improve the detection limit and minimize the statistical error of the peak detection. Some radionuclides with more than two coincident gamma rays can not be effectively determined by Compton suppression. For example, the radionuclide ^{46}Sc has two gamma rays with energies 889 keV and 1120 keV. These two photopeaks are coincident with each other in the Compton spectrum. The count rates of two peaks in this case are no longer proportional to amount of the scandium. The peak counting rates vary with deadtime that can not be adequately corrected using the usual procedures. An example of ^{46}Sc-1120 keV photopeak measured in the Compton mode is presented in Figure 2. This experiment was done by using a fixed ^{46}Sc source and another removable gamma ray source (without 1120 keV peak) to adjust the dead time. In normal mode, the peak rate decreases with dead time, which can be compensated by using pulser, two-source correction or internal ADC correction. However, the count rate in the Compton mode increases with dead time, and this phenomenon depends on the individual radioisotope. Ratio of the peak rates, also shown in Figure 3, decreases significantly with dead time. This means that detection of two coincident gamma rays from the same isotope depends on dead time of the Compton system. The higher the deadtime, the less effective the Compton suppression, due to higher random coincidences being detected. Millard [2] did a study on this subject for dead times up to 18%. It was concluded that the results obtained with Compton suppression counting depended not only on the physical arrangement of the detectors and the performance characteristics of the electronic equipment but also on the decay characteristics of the nuclides analyzed. The ratio of the count rate in a gamma line in anti-coincident and coincident modes was used to monitor the proper quantification of the activity measured. Therefore, for successful and reliable implementation of Compton suppression in NAA, a detailed investigation on the change in activity quantification is warranted. In this work a different approach was used. A 1% standard liquid solution of manganese was irradiated to produce ^{56}Mn with a half-life of 2.6 h. The same activated sample was counted repeatedly so that as the overall activity decreased,

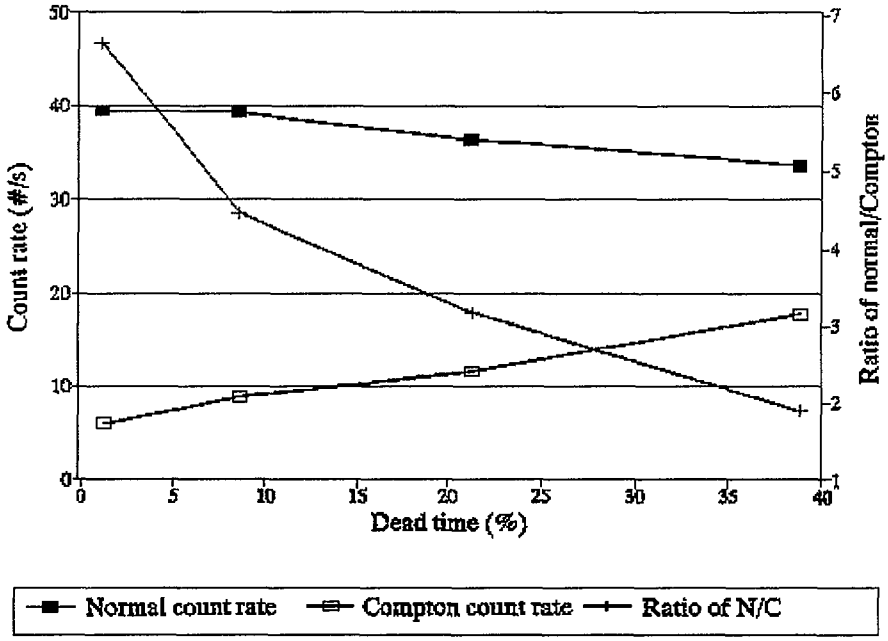

Fig. 2. Count rate of ^{46}Sc 1120 keV peak as a function of dead-time in normal and Compton counting modes.

there was a decrease in the dead time of the measurement. The analysis was done exactly like routine NAA. The irradiated solution had predominantly ^{56}Mn activity. The three gamma lines at 846.8, 1810.7, and 2113.2 keV were evaluated. The first one has a 50% coincident rate, while the other two are 100% coincident. Both Compton suppressed and unsuppressed spectra were collected and analyzed. The results for a 96% and 19 per cent deadtime are shown in Figure 3 and 4, respectively. Corrections for the dead time by means of a pulser were applied in both suppressed and unsuppressed modes. The figures presented can be interpreted as the departure of the efficiency of detection present because of an increase in the overall dead time of the measurement. As the dead time increases, the Compton suppression becomes less effective; therefore, photons that are suppressed because they are coincident become less suppressed as the dead time increases. The Compton suppressed mode, on the other hand presents an increase in the overall count rate for the gamma lines with 50 and 100% coincidence rate. The exact difference in the detector efficiency with variations in the dead time will depend on the coincident rates of the various gamma lines, among other factors. It is not feasible that such differences can be exactly calculated and corrected for individual isotopes; therefore, reliable analysis with Compton suppression is best applied when the dead times are low involving non-coincident or mostly non-coincident gamma lines. More recently Westpahl et al. [3] have introduced loss-free counting techniques to Compton suppression methods but it has seen only limited use. As well Westpahl [4] also reported on the real-time correction of chance coincidence losses in high count rate Compton spectrometry.

Influence of Compton suppression on counting statistics

Background

It has been pointed out by Pomme et al. (2000) that although nuclear emission does indeed often show Poisson statistical behavior, nuclear spectroscopy counting may not. This is primarily due to non-random counting losses due to deadtime and and/or pulse pileup, which can transform some of the Poisson statistical characteristics. Furthermore, Pomme et al. (1999) showed that pileup rejection causes an enhanced count scatter when applying loss free counting. We investigated if Compton suppression did alter any of the normal statistical fluctuations observed seen in routine gamma ray counting.

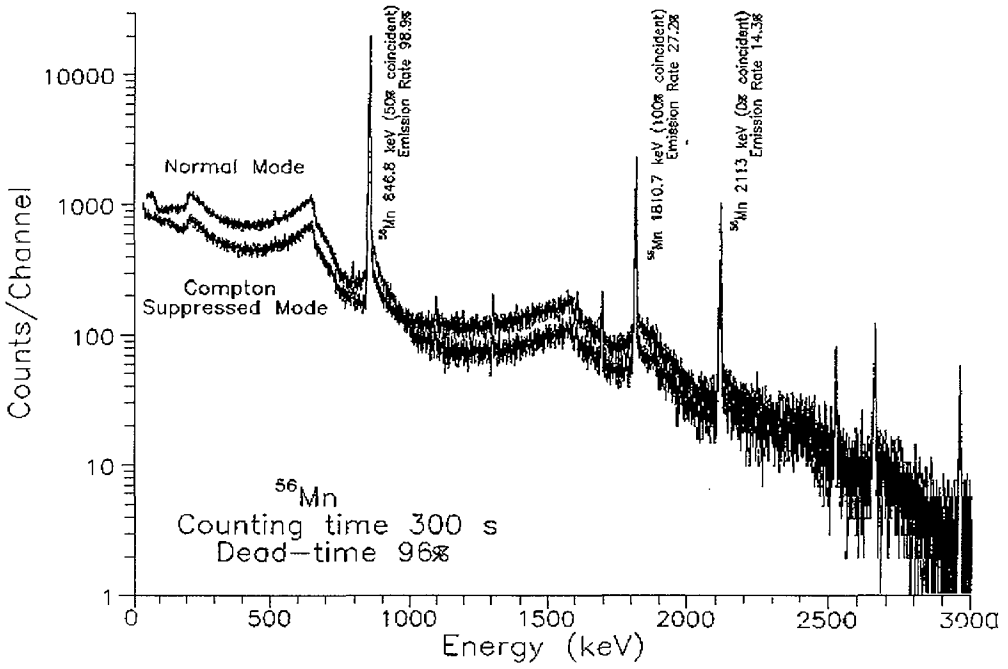

Fig. 3. Spectra of ^{56}Mn measured in normal and Compton counting modes with dead time 96%.

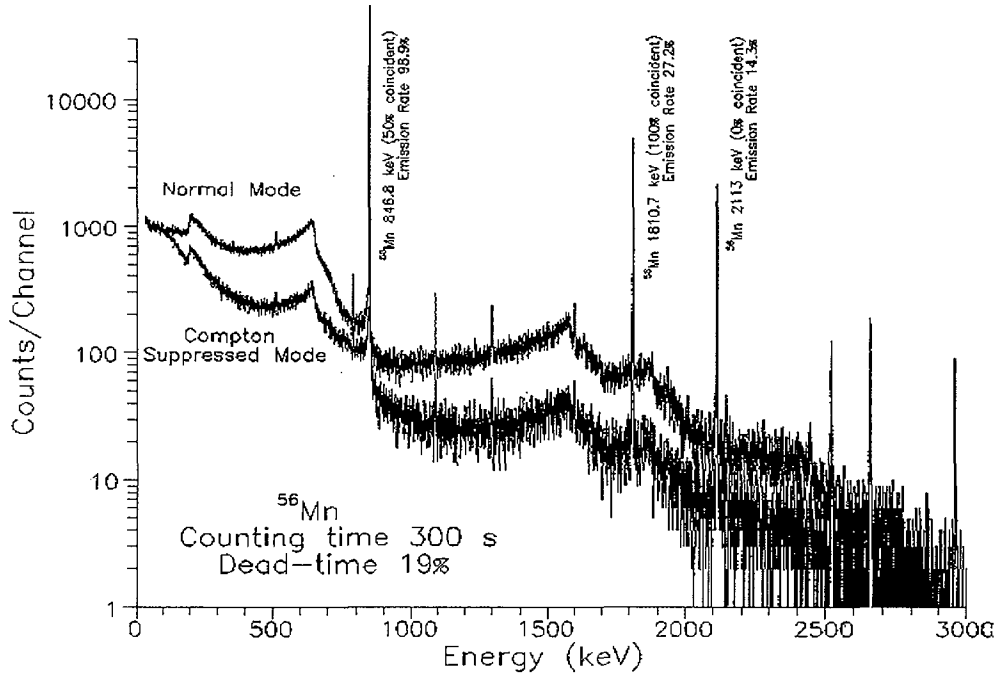

Fig. 4. Spectra of ^{56}Mn measured in normal and Compton counting modes with dead time 19%.

Experimental

We counted a ^{60}Co source in a normal and Compton mode. A counting position was chosen to give a deadtime of less than 2%. Both the 1173 and 1332 keV photopeaks are in 100% coincidence with each other. This results in a counting efficiency being reduced almost by factor of eight which ideal since this is using the Compton effect to its fullest. We acquired almost 250 spectra both in the normal and counting mode having average net peak areas of 59,640 and 8,356 counts.

Normal Acquisition (1332 keV photopeak):

mean	=	5.964 E+4 counts
std (exp)	=	230.2 (experimental result)
sqrt (mean)	=	244.2 (Poisson theoretical std)

Compton Suppression (1332 keV photopeak):

mean	=	8.356 E+3 counts
std (exp)	=	99.0 (experimental result)
sqrt (mean)	=	91.4 (Poisson theoretical std)

Results for the normal and Compton data were statistically treated and graphically represented in Figures 5 and 6. As can be seen there appears to be virtually no significant statistical differences between the normal and Compton modes.

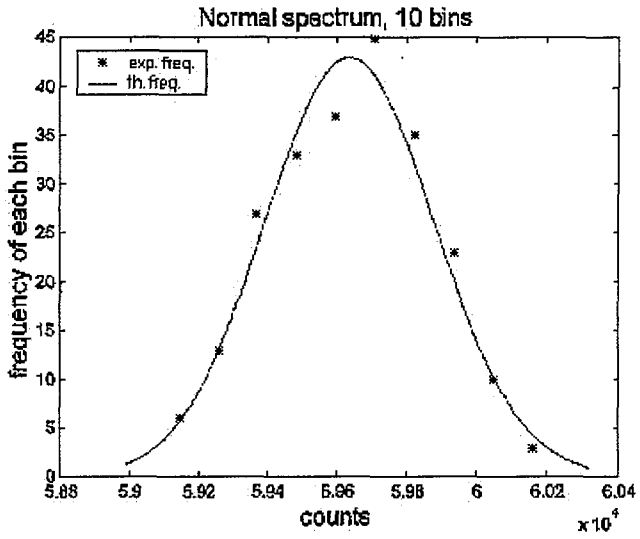

Fig. 5. Normal spectrum, 10 bins.

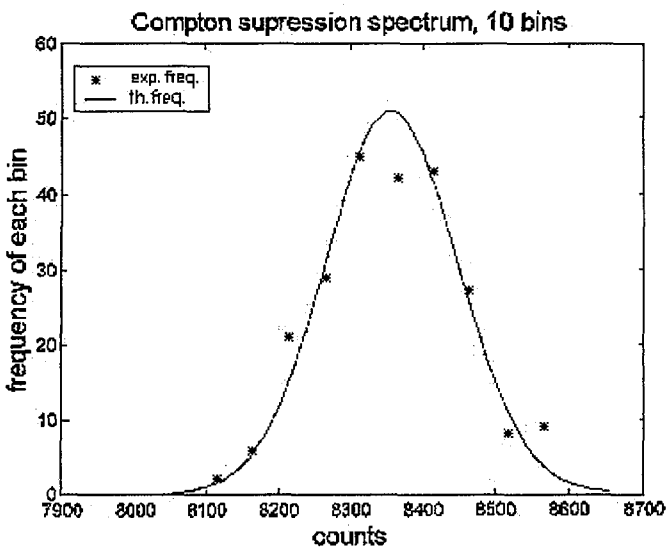

Fig. 6. Compton Suppression spectrum, 10 bins.

Determination of ^{137}Cs in soil samples by low-level Compton suppression gamma counting

Background

Ever since the advent of atmospheric nuclear testing, there have been intense studies into the environmental and medical consequences of ^{137}Cs, which is a major product of the fission process. ^{137}Cs has a half-life of 30 years and has become ubiquitous in the northern hemisphere due to atmospheric tests conducted primarily by the United States and the Former Soviet Union. Atmospheric tests were also conducted to a much lesser degree by the French and later on by the Chinese. The British mainly conducted their tests in the southern hemisphere. Although atmospheric testing was halted in 1963 with the advent of the partial test ban treaty, ^{137}Cs was again widely released as a direct result of the Chernobyl nuclear reactor accident in 1986. Large areas of the Ukraine, and other Former Soviet Union Republics were contaminated with ^{137}Cs and other radionuclides. Furthermore, areas in various parts of Europe received the radioactive fallout. Thus, one can expect that ^{137}Cs will remain in the environment at measurable quantities for the next two-three centuries. In fact ^{137}Cs is now considered to be part of the "natural background" in naturally occuring radioactive materials (NORM). ^{137}Cs is readily absorbed onto clay particles, for those size fractions less than 2 micrometers (< 2 μm). Because of this phenomenon one can derive a historical dating (1945 onwards) of soils by sampling up to one meter below the surface.

One area of continued research is in soil erosion, which can be modeled using the ^{137}Cs data collected in soil samples. One can also measure the on-site distribution of soil erosion, deposition within several watersheds, determine annual average soil loss, and ultimately evaluate the consequences for plant productivity and loss. The US Army conducts its training and various maneuver exercises on land. At present the Army Training and Testing Area Carrying Capacity (ATTACC) model is currently the only officially sanctioned tool to determine the capacity of military lands to support training and testing missions. Special consideration is given to land rehabilitation and maintenance including carrying capacity.

As a result of this need, the US Construction Engineering and Research Lab contracted the Nuclear Engineering Teaching Laboratory to measure ^{137}Cs in two soil environments. One in an undisturbed place for background information and the other in general areas of a US Army training facility. Forth Hood, Texas was chosen for sampling site.

Sampling and sample preparation

In the first expedition undisturbed samples were collected at two gravesites; Walker (W) and Spring Hill (S) Cemetery at Fort Hood. Two locations (1 and 2) several meters apart were chosen at each gravesite. Four separate pits about one meter apart in a circle were dug at each location. Very careful sampling was done to ensure accurate depth profile. From each pit 3 cm slabs sections down to 24 cm were collected, placed into polyethylene bags and labeled. Therefore for each specific location there were 4 sub-samples at every depth. These sub-samples were mixed together, to ensure maximum representation of the locations, making a total of eight samples per location. This procedure was repeated several meters away at the second location. Samples were identically collected from another two locations in the second gravesite. The total number of composite samples collected was 32. Once the samples delivered to the laboratory, they were sieved using a standard ASTM 1.18 mm to remove small stones and other types of debris. The sieving significantly helped in homogenizing the soil samples. The average final combined weight was about 0.4–0.6 kg. In the second expedition, 86 grab samples were taken from various sites at Fort Hood (these results will be reported in a future communication).

Experimental

From each bag 100 grams of soil was placed into a 250-mL glass beaker. The Compton suppression gamma ray spectrometer was used to detect the 662 keV photon decaying from ^{137}Cs. This system greatly reduces the background from naturally occuring radiation such as the multitude of radiaoctive decay products from the uranium series and ^{40}K. Ultimately, the signal to noise ratio significantly improves making the detection of small peaks easier. A complete technical description of the set-up and review of Compton suppression is given elsewhere (Landsberger and Peshev, 1996). A list of the major gamma rays found in our laboratory environment and their reduction factors are shown in Table 1. Samples were typically counted for 0.6–1 day. A partial spectrum between 600–700 keV with and without Compton suppression is seen in Figure 7. A comparison between Figs. 8 and 9 reveals that for high ^{137}Cs concentrations there is little advantage of using Compton suppression. However, for low ^{137}Cs concentrations the signal to noise ratio improves to reduce the error on the photopeak significantly. The advantage of using Compton suppression is the ability to achieve good sensitivity for ^{137}Cs using relatively small sample sizes; from 100–200 grams.

Accurate radioactive counting of low-levels of contaminated or environmental soils is one of the most difficult analytical procedures to undertake. Firstly, one needs to have enough material to count for a reasonable amount of time. One the other hand, one needs to limit the amount of material so that matrix effects in the form of gamma ray attenuation is reduced or minimized. In the second requirement matrix matching of samples and calibration material is crucial. Not all soils have the same major matrix composition and density. Thus, a soil that is denser and has higher amounts of potassium and calcium my attenuate the gamma ray signal more substantially than a soil that is less dense and may have a typical aluminum-silicate base.

TABLE 1. SUMMARY OF MAJOR NUCLIDES FOUND IN LABORATORY BACKGROUND AND THE REDUCTION FACTOR WHEN USING COMPTON SUPPRESSION (CPD = COUNTS PER DAY)

Energy Kev	Nuclide	Source	cpd without Compton	cpd with Compton	cpd without Comp/ cpd with Comp
186.2	^{226}Ra	^{226}Ra series	5604	1935	2.90
238.6	^{212}Pb	^{232}Th series	3609	2264	1.59
295.4	^{214}Pb	^{226}Ra series	2635	1065	2.47
352.0	^{214}Pb	^{226}Ra series	2250	1033	2.18
511.0		annihilation	2317	466	4.97
583.1	^{208}Tl	^{232}Th series	999	318	3.14
609.3	^{214}Bi	^{226}Ra series	1252	444	2.82
911.0	^{228}Ac	^{232}Th series	546	225	2.43
968.8	^{228}Ac	^{232}Th series	447	159	2.81
1120.0	^{214}Bi	^{226}Ra series	419	132	3.17
1460.8	^{40}K	primordial	1229	1042	1.18
2614.3	^{212}Pb	^{232}Th series	236	139	1.70

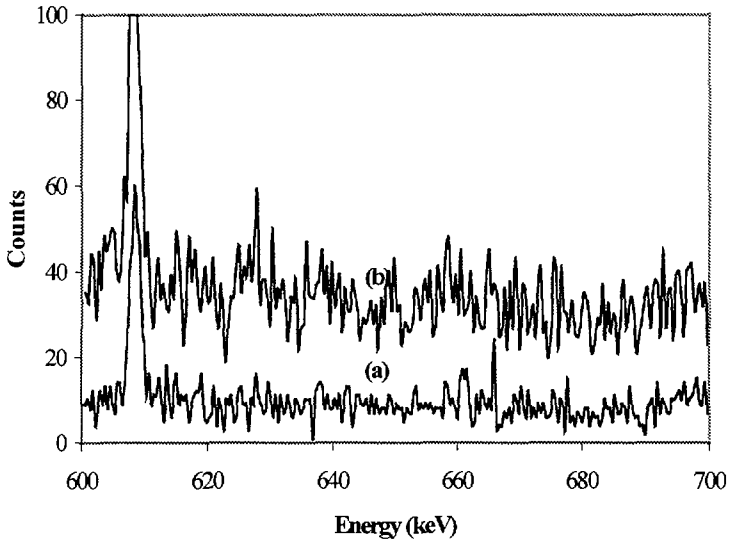

Fig. 7. Comparison of laboratory background. (a) with and (b) without Compton suppression.

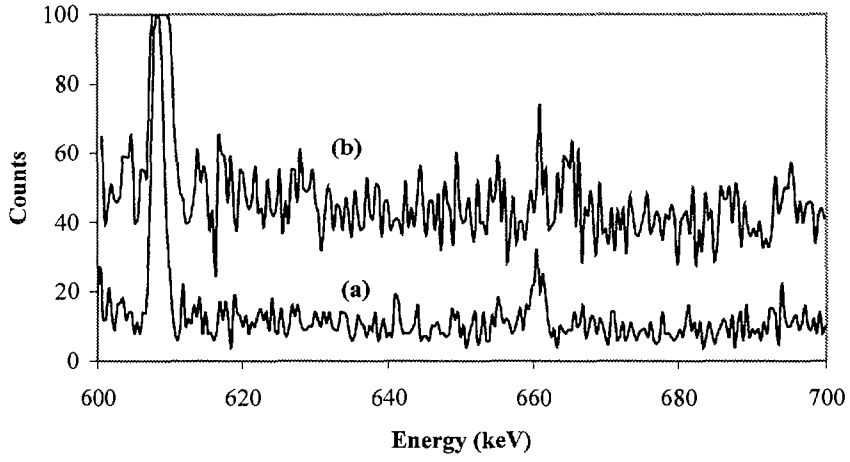

Fig. 8. Low concentration of ^{137}Cs (0.7 Bq/kg). (a) with and (b) without Compton suppression.

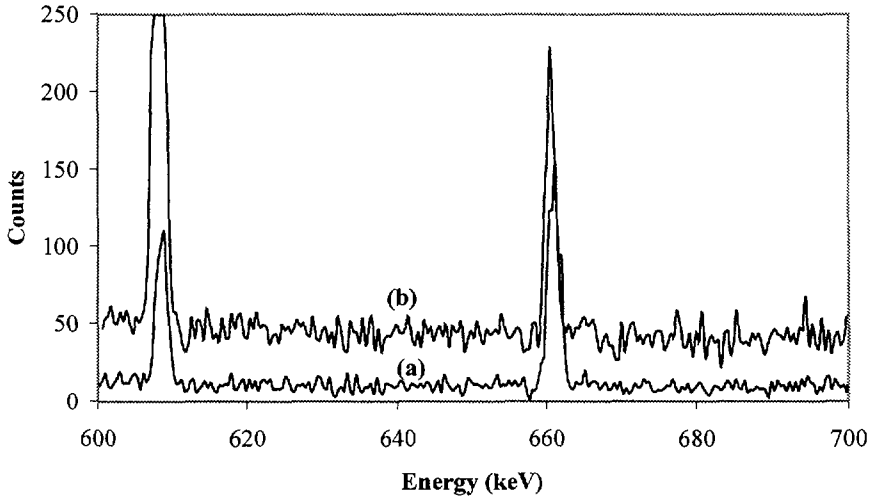

Fig. 9. High concentration of ^{137}Cs (12 Bq/kg). (a) with and (b) without Compton suppression.

In this work two International Atomic Energy Agency (IAEA) standard reference materials were used to calibrate the system and perform quality assurance. IAEA (375) sample with a concentration of 5280 Bq/kg (with a confidence interval of 5200–5360 Bq/kg) was used as the calibration source and IAEA Soil 6 with a concentration of 53.65 Bq/kg (with a confidence interval of 51.43–57.91 Bq/kg) was used as an unknown. The result was within 10% of the certified value. We suspected that the difference in soil density mainly attributed to this error. To test our hypothesis we used only five grams of IAEA Soil 6 as the standard and five grams of IAEA 375 as the unknown. Five separate measurements were performed with results all lying between ±3.5% of the mean value of 5280 Bq/kg. The results are shown in Table 2.

We performed a T-test [7] to evaluate statistical control with 4 degrees of freedom and $P \geq 0.20$ and found the results to be acceptable.

The next set of experiments was to obtain a soil profile of the four sites, two each from the same cemetery. These results are seen in Table 3 and are plotted in Figure 10. The reproducibility of locations S-1 and S-2 from the first cemetery are in very good agreement, while the W-1and W-2 locations do not agree as well. However, the general ^{137}Cs profile of all sites shows a exponential drop from top soil to a depth of 24 cm.

TABLE 2. QUALITY CONTROL REPORT FOR MEASURING ^{137}CS USING IAEA SOIL 6

Measured Activity (Bq/kg) ± Std. Dev.	% Agreement
5102 ± 86.8	96.5
5299 ± 89.9	100.3
5191 ± 80.0	98.3
5200 ± 88.2	98.5
5388 ± 93.8	102.0

TABLE 3. MEASUREMENT OF ^{137}CS IN SOIL SAMPLES AT VARIOUS DEPTHS

ID	Depth cm	S-1 Bq/kg	S2 Bq/kg	W1 Bq/kg	W2 Bq/kg
A	3	13.81 ± 0.77	16.58 ± 0.54	17.74 ± 0.76	24.57 ± 0.52
B	6	13.22 ± 0.67	16.66 ± 0.84	14.79 ± 0.65	19.84 ± 0.73
C	9	8.46 ± 0.58	9.79 ± 0.71	8.17 ± 0.75	11.44 ± 0.59
D	12	6.94 ± 0.36	5.64 ± 0.33	3.51 ± 0.45	5.66 ± 0.30
E	15	3.18 ± 0.31	3.52 ± 0.23	2.09 ± 0.21	4.42 ± 0.32
F	18	2.96 ± 0.27	2.82 ± 0.27	1.24 ± 0.20	3.03 ± 0.24
G	21	2.02 ± 0.20	1.69 ± 0.18	1.16 ± 0.20	2.04 ± 0.20
H	24	1.93 ± 0.20	1.02 ± 0.16	1.06 ± 0.18	1.66 ± 0.19

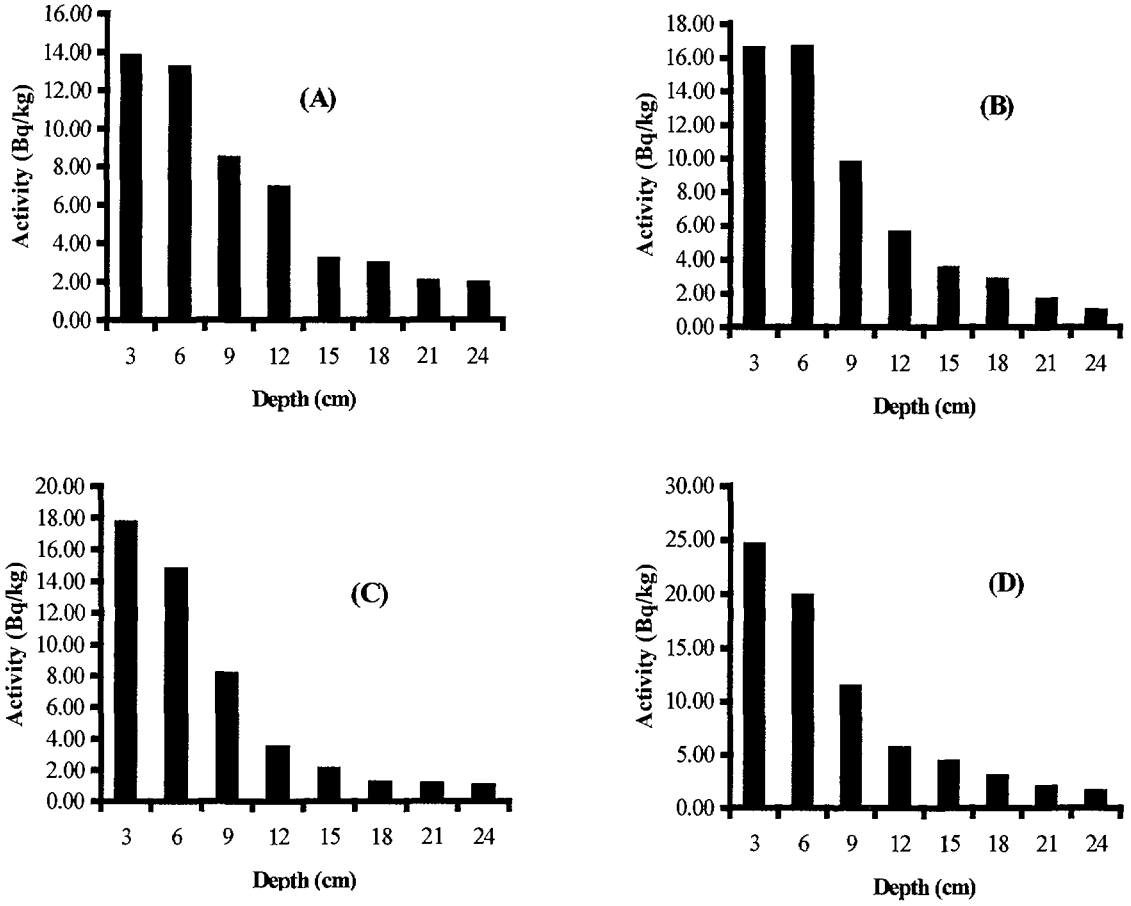

Fig. 10. Graphical interpretation of ^{137}Cs in soil samples at various depths from the four sampling locations; (A) Spring Hill-1; (B) Spring Hill-2; (C) Walker-1 and (D) Walker-2.

In the second set of experiments we investigated the effect of counting time on the counting statistical error. For samples with high ^{137}Cs concentrations greater than 10 Bq/kg, we found no statistical difference between the normal and Compton suppression gamma counting mode for counting periods of 0.5, 1, 1.5, 2, and 3 days (Table 4). However, for ^{137}Cs concentrations less than one Bq/kg, the error significantly drops when using the Compton mode after one day of counting (Table 5).

TABLE 4. COMPARISON BETWEEN COUNTING STATISTICS UNDER COMPTON VS NORMAL COUNTING MODE FOR A SAMPLE WITH ACTIVITY OF 12.59 Bq/Kg

Count Time (Days)	% Counting Statistics Normal Mode	% Counting Statistics Compton Mode
0.5	9.4	10.9
1	6.5	6.8
1.5	5.8	5.3
2	5.5	4.6
3	4.3	4.9

TABLE 5. COMPARISON BETWEEN COUNTING STATISTICS UNDER NORMAL VS COMPTON COUNTING MODE FOR A SAMPLE WITH ACTIVITY OF 0.73 Bq/Kg

Count Time (Days)	% Counting Statistics Normal Mode	% Counting Statistics Compton Mode
0.5	31.0	32.6
1	27.9	20.0
1.5	27.9	20.0
2	28.8	16.7
3	25.6	7.2

Conclusions

We have demonstrated that Compton suppression gamma ray spectrometry is ideal to determine ^{137}Cs in soil samples as low as 100 grams. The main advantage of the system is in determining ^{137}Cs concentration concentrations at levels between 1–3 Bq/kg. Typical counting statistical errors are 10% at 5 Bq/kg and 20% at 1 Bq/kg. Below 1 Bq/kg the errors rise to 30-35% for a one-day count. It is recommended that for a large sampling program the amount of sample be increased to 200 grams to reduce the counting time for lower ^{137}CS concentrations and acquire a much higher efficiency detector (> than 50%) which would further reduce the counting time. As well for such larger sampling programs a specific sample changer and dedicated Compton suppression system should be allocated.

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