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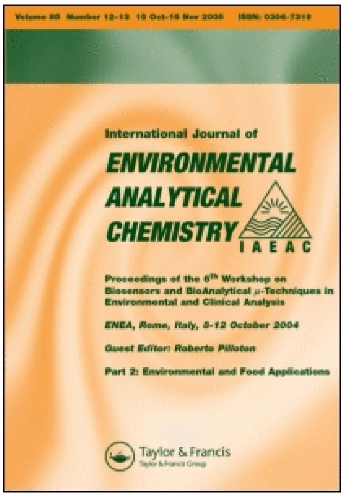

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J. J. Labrecque[a]
[a] Laboratory of Atomic and Nuclear Spectroscopy, Instituto Venezolano de Investigaciones Cientificas-IVIC, Caracas, Venezuela

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THE DETERMINATION OF ENVIRONMENTAL LEVELS OF 137Cs IN VENEZUELAN MILK POWDERS*

J. J. LABRECQUE

Laboratory of Atomic and Nuclear Spectroscopy, Instituto Venezolano de Investigaciones Científicas-IVIC, Apartado 21827, Caracas 1020A, Venezuela.

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The measurement of ^{137}Cs in six Venezuelan milk powder samples and four reference materials was performed by a standard calibration curve method and a standard reference material ratio method. This was accomplished employing both a high efficiency Hype pure Germanium semiconductor coupled to a multichannel analyzer and a simple NaI(Tl) scintillator with a single channel analyzer. The standard addition technique was also applied to two samples since they were used as a dilutants to prepare the standards for the calibration curves. The two samples of the government subsidized milk powder were not significantly different from the other five national milk powders, thus no indication that they were mixed with contaminated milk powder imported from Europe. Finally, these levels can be referred to in the future in case of a nuclear accident in the region or if the national product is suspected of being mixed with imported contaminated milk powder.

KEY WORDS: ^{137}Cs, Venezuelan, milk powder, method comparison

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INTRODUCTION

Since 1970, when the world treaty on the non-proliferation of nuclear weapons came into effect, the concern about the radioactive contamination of the environment in Venezuela had become very limited. The study of the beta activity in the air and ^{90}Sr in the rain at Instituto Venezolano de Investigaciones Científicas[1] that began in 1959 in conjunction with the U.S. Atomic Energy Commission was discontinued in 1963. Since then only a little interest has been given to radioisotope contamination, such as the marine environment[2]. But again, shortly after the Chernobyl nuclear power plant accident in the USSR, the concern was renewed. At first, studies on the direct deposition of the fallout were taken up by various groups and later on the screening of the fallout which found its way into the food chain of the imported foodstuffs from Europe, such as milk powder and meats.

The most dangerous long-lived radionuclides released from a nuclear accident are ^{137}Cs and ^{90}Sr[3]. The main source of these radionuclides in the diet are milk and dairy products, thus affecting children more who usually consume relatively more milk than adults. Thus, milk is the most studied component of the human diet for

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^{137}Cs and ^{90}Sr. Because milk powder is imported into Venezuela to make up for the national production shortages, it was decided to determine the environmental levels of ^{137}Cs in the different national brands of milk powders in order to have a reference value, in case a nuclear accident does occur in our region which results in deposition of ^{137}Cs and ^{90}Sr, as well as to be able to suspect if contaminated milk powders are being mixed with the national product.

In this work, we have compared a simple 5 cm × 5 cm NaI(Tl) detector with a single channel analyzer with an expensive high efficiency (about 25%) Hype-pure Germanium semiconductor coupled to a multichannel analyzer employing both a standard calibration curve method and a standard reference material ratio method for ^{137}Cs in milk powders.

MATERIAL AND METHODS

Six different brands of Venezuelan milk powder and four standard reference materials from the International Atomic Energy (IAEA) were analyzed for ^{137}Cs. One-half kilogram samples were directly transferred from their respective containers upon opening to 1-liter plastic bottles and closed. At the same time three sub-portions of about five grams each were taken for the humidity determination. The water content was determined by oven drying at 100°C ± 2°C for 4 hours. The values reported herein have been corrected for their respective water content, in other words on a dry-basis.

Two independent sets of calibration standards were prepared by serial dilution with the first dilution being only ten to one. The first set employed IAEA standard reference material, whey powder-154 as the base and commercial milk powder X as the dilutant. This resulted in standards with the following ^{137}Cs content after correcting for the amount of ^{137}Cs in the commercial milk powder X which was determined by standard additions from these same standards: 6, 11, 20 and 39 $BqKg^{-1}$. The second set employed IAEA standard reference material, milk powder-152 as the base and commercial milk powder Y as the dilutant. Again, after correction for the ^{137}Cs content of the commercial milk powder the calculated values were: 3, 4, 6 and 10 $BqKg^{-1}$. A detail description of the preparation of these synthetic standards has been previously reported[4]. It should be noted that the reference date for these standards was 31 August 1987. A composite calibration curve for ^{137}Cs in these standards is given in Figure 1. The values for the Venezuelan milk powder samples were corrected to the time of their respective measurements while that of the IAEA standard reference materials were corrected to their respective reference dates for their certification.

The measurements of ^{137}Cs were performed by placing the 1-liter bottles upright in a circular lead shield which is 5 cm in thickness with a 5 cm × 5 cm NaI(Tl) crystal surrounded by 7.5 cm of lead at the base of the shield. The detection system consisted of Eurocard components supplied by the International Atomic Energy Agency and assembled by the electronic group at IVIC's nuclear reactor. The following modules were assembled and employed in this work: 1) a NIM Bin power supply; 2) a 902

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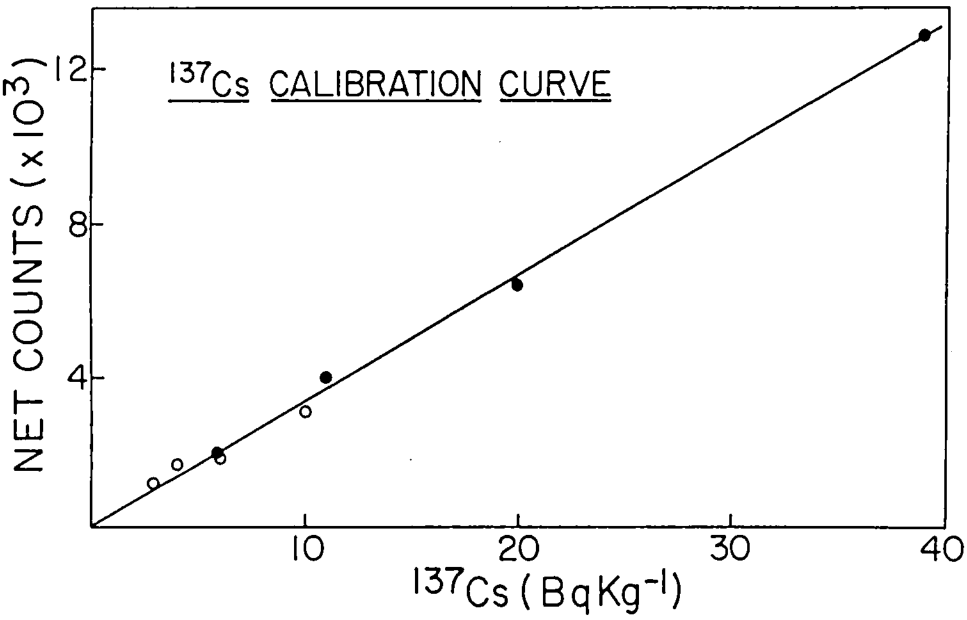

Figure 1 A composite calibration curve for ^{137}Cs from the two different sets of calibration standards employing the NaI(Tl) system with a single channel analyzer. The solid dots are from the standards prepared from IAEA-154 whey powder and the open circles are from those employing IAEA-152 milk powder.

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model preamp power supply; 3) a 201 model high voltage power supply; 4) a 102 amplifier; 5) a 301 timing single channel analyzer; and 6) a 402 scalear. All samples, standards and blanks were measured for 5×10^4 seconds preset. This long counting period was usually performed during the night and because of the frequent power outages, the whole system was protected with an IAEA drop-out relay. The content of ^{137}Cs was calculated from either the lower or higher part of the composite calibration curve in Figure 1 depending on the concentration involved and also by a standard reference material ratio employing A-14.

An attempt to quantify the ^{137}Cs content in the local milk powder with an Apple II+ microprocessor with a Nucleus ADC/interface card with 1024 channels as a multichannel analyzer instead of the single channel analyzer was not successful. A typical spectrum of one of the national milk powder, X and the 20 $BqKg^{-1}$ standard is shown in Figure 2 for this detection system. The same samples and standards in the same plastic container were measured by a 25% efficient Hype-pure Germanium detector with an Apple IIe and a Nucleus interface/ADC card as a multichannel analyzer in a similar lead shield with the appropriate bias supply and spectroscopy amplifier for 5×10^4 seconds. This system was also protected with an IAEA drop-out relay for power failures. A typical spectrum of a Venezuelan milk powder, X and the 20 $BqKg^{-1}$ standard is given in Figure 3. Again both the standard calibration curve method and the standard reference material ratio method were employed after the peaks areas were determined and corrected for background and decay time.

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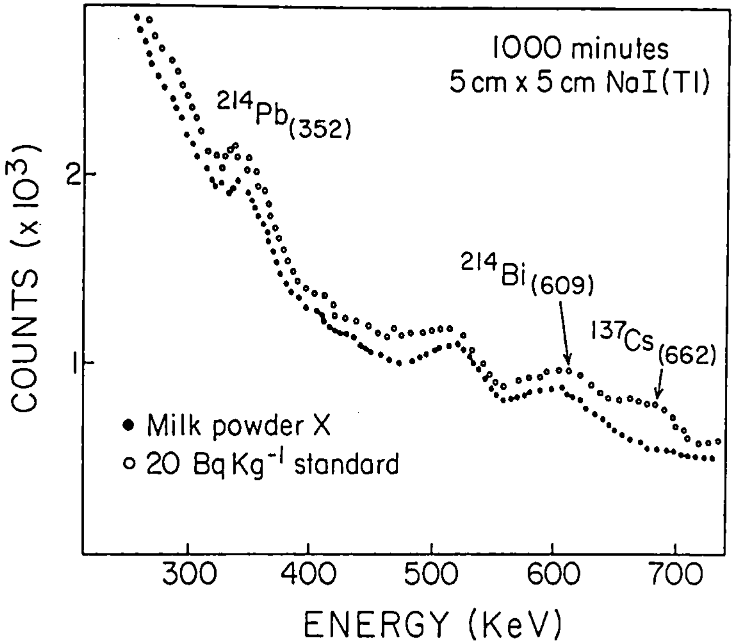

Figure 2 A gamma ray spectrum of milk powder X (solid dots) and the 20 $BqKg^{-1}$ standard of ^{137}Cs (open circles) with the NaI(Tl) system coupled to a multichannel analyzer for 1000 minutes each.

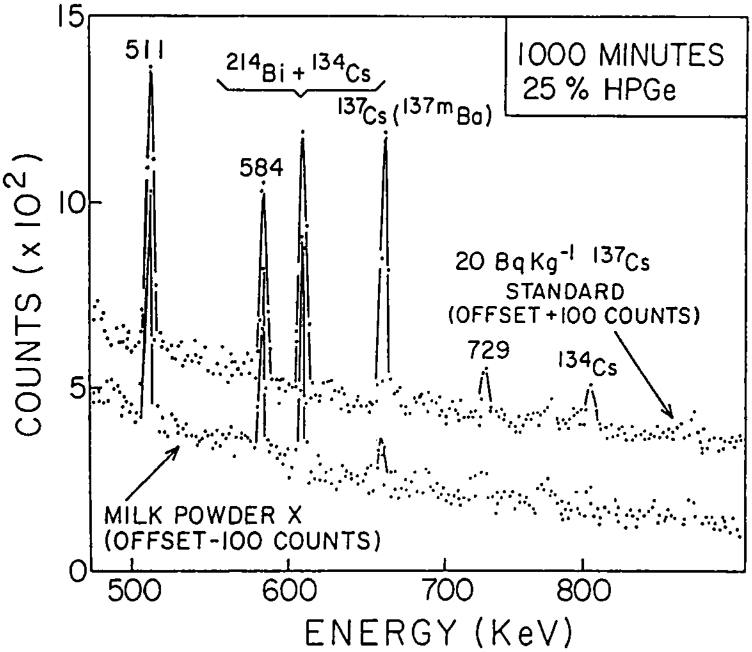

Figure 3 A gamma ray spectrum of milk powder X (bottom; offset − 100 counts) and the 20 $BqKg^{-1}$ of ^{137}Cs (top; offset + 100 counts) with the Hype-pure germanium system for 1000 minutes each.

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RESULTS AND DISCUSSION

The composite graph of the calibration curves as shown in Figure 1 for the NaI(Tl) system from the two sets of standards shows that the two curves coincide with each other even though their matrices were somewhat different. The values for the milk powder samples employing these two different calibration curves were slightly different but not significantly, thus we employed the lower concentration calibration curve since the values of these standards were closer to the values of our milk powder samples, as well as the measured value of A-14 was closer to its certified value. To calculate the exact values of the standards, it was necessary to correct the serial dilution for the amounts of ^{137}Cs in the dilutant milk powder. This was performed by employing the standard additions methods to the synthetic standards. The determined value for ^{137}Cs content of milk powder X was determined to be 1.6 $BqKg^{-1}$ when the standard additions were performed with IAEA-154 whey powder standard reference material. Similarly, the value of ^{137}Cs in milk powder Y was measured to be 2.4 $BqKg^{-1}$ using IAEA-152 milk powder standard reference material for the additions. These values are not significantly different than the direct measured values considering their standard deviation (1σ).

The mean values and their respective standard deviations (1σ) of ^{137}Cs content in the six commercial Venezuelan milk powders and the four standard reference materials from the IAEA are summarized in Table 1 from the four different methods and are also compared graphically in Figure 4. In general, there is no significant differences in the values from the different methods considering the 1σ standard

Table 1 Summary of results for the measurements of ^{137}Cs in six commercial Venezuelan milk powders* and four standard reference materials.

		CONCENTRATION OF ^{137}Cs (Bq/Kg) (Mean Standard Deviation)				
		HpGe Measurement (MCA)		*NaI(Tl) Measurement (SCA)*		
Sample	*Certified value***	*Standard curve*	*SRM ratio*	*Standard curve*	*SRM ratio*	*Standard additions*
A-14	1.79 ± 0.18	1.81 ± 0.21	1.66 ± 0.28	1.74 ± 0.34	1.68 ± 0.31	—
IAEA-321	72.7 ± 5.5	71 ± 2	71 ± 2	69 ± 3	67 ± 4	—
IAEA-152	2129 ± 78	2082 ± 31	2120 ± 21	1938 ± 88	2103 ± 60	—
IAEA-154	3749 ± 137	3711 ± 42	3752 ± 27	3671 ± 123	3725 ± 82	—
X	—	0.98 ± 0.10	0.57 ± 0.06	1.74 ± 0.36	1.43 ± 0.30	1.6 ± 0.6
Y	—	1.68 ± 0.26	1.83 ± 0.28	1.94 ± 0.51	1.53 ± 0.40	2.4 ± 0.8
T	—	2.20 ± 0.57	1.73 ± 0.46	1.98 ± 0.58	1.51 ± 0.44	—
W	—	1.10 ± 0.29	0.94 ± 0.25	1.67 ± 0.54	1.32 ± 0.42	—
V	—	2.00 ± 0.39	1.98 ± 0.39	2.40 ± 0.69	2.16 ± 0.76	—
P1	—	1.52 ± 0.10	0.89 ± 0.06	0.90 ± 0.29	0.59 ± 0.12	—
P2	—	1.66 ± 0.30	0.98 ± 0.18	1.50 ± 0.72	1.34 ± 0.25	—

* The following brand names were coded P1 to Y in order to be anonymous: Camprolac, Rika, Indosa, La Pradera, La Campiña and leche popular (subsidized by the government).

** Certified values are the values for the respective reference dates of the SRM and the measured values are corrected to these dates.

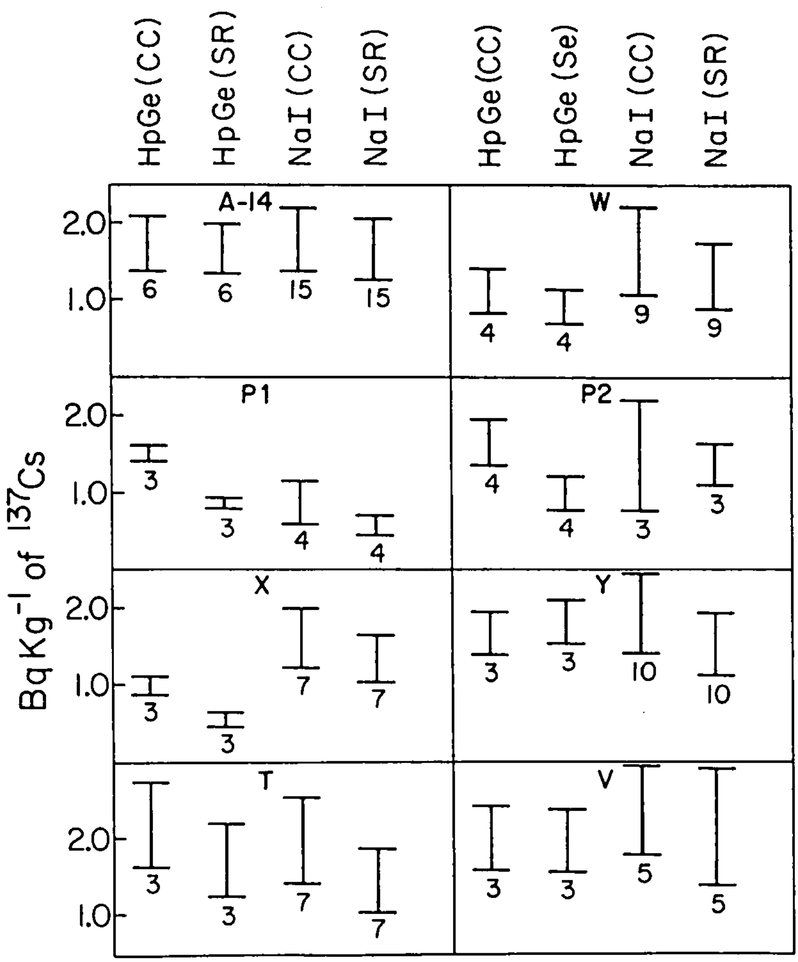

Figure 4 A graph of the mean values and their respective standard deviations (1σ) for A-14 standard reference material and six different Venezuelan milk powders from the different methods employed. The number at the bottom of the error bars is the number of the replicates (n); CC = standard calibration curve method; SR = standard reference material ratio method.

deviation. The values from the Hype-pure Germanium system were somewhat more precise, a smaller standard deviation than those of the NaI(Tl) system. The standard calibration curve method resulted with slightly higher mean values compared to the standard reference material ratio method and was even significantly different for two of the milk powder samples when employing the Hype-pure Germanium system.

Our measured mean values for the standard reference materials are slightly lower than the certified values but fall well within the 95% confidence limits, thus can be considered accurate. A higher concentration calibration curve in the range of 500–3500 $BqKg^{-1}$ of ^{137}Cs was employed for the proper analysis of IAEA-152 and

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IAEA-154. And IAEA-152 was the standard reference material employed for the standard reference material ratio method.

The milk powders coded P1 and P2 were different lots of "leche popular", the milk powder subsided and supplied by the government for the poorer population, this is the milk powder one would expect to be mixed with imported milk powder when there are national shortage. But as can be seen in Table 1 and Figure 4, the measured mean values of ^{137}Cs for the milk powders were actually lower than the other national milk powders. There is only a slight difference in the composition as reported on the respective containers.

In conclusion, it has been shown that a simple 5 cm × 5 cm NaI(Tl) detection system with a single channel analyzer can be employed for the measurement of environmental levels of ^{137}Cs in Venezuelan milk powders with long counting periods even though this system has a poorer energy resolution and precision than the Hype-pure Germanium system. But the mean values of the various measurements were shown to be accurate by the comparison of the measured and certified values for IAEA milk powder A-14 standard reference material which has a ^{137}Cs content in the middle of the range of values determined in the Venezuelan milk powders.

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