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Determination of Radioactive Contaminant ^{137}Cs (Fission Product) and Comparison with Natural ^{40}K Present in Mexican Strawberries

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Abstract

Several previous papers have been published about radioactive contamination in Mexican soil, marine sediments and foodstuffs. Ever since, results have been according to expectation, mainly consisting of natural isotopes in small concentration. Continuous research leads with the surprising finding of fission product ^{137}Cs ($t_{1/2}$ = 30 years) in strawberries produced in different states, about 2000 km away from each other. Never before, such contaminant has been observed in foodstuffs, produced in this country. The aim of this work is to report the presence of ^{137}Cs in Mexican strawberries and compare the activity of such contaminant with the natural radioactivity of ^{40}K ($t_{1/2} = 1.28 \times 10^9$ years), through gamma spectrometry. Eventually, the specific activity of ^{137}Cs observed in samples FM4, FM5 and FBC is respectively 0.7, 0.8 and 0.3 Bq/kg. The limits established in CODEX-STAN-193-1995 for ^{137}Cs are 1000 Bq/kg, so all samples meet the recommendation. It's presence now in vegetables result with enough interest to be known and considered in the near future to measure the possible effect that radioactive contamination could reach after nearly 80 years from second world war, where no nuclear test has been performed, nuclear accident occurred, neither nuclear weapon has ever been used.

Keywords

Radioactive Contamination

1. Introduction

Now at days, radiological contaminants, both natural and anthropogenic, have

been identified in different regions of the world, especially where radiological events such as Chernobyl or Fukushima have occurred, to name a few. In Mexico, no nuclear power plant has reported incidents, no nuclear weapons have been tested in this territory, nor is there any report of intentional detonations of any type of nuclear weapon. In other words, Mexico does not have records of nuclear tests or accidents where fission products have been released.

During the last 30 years, multiple research works have been carried out on soils, water and food, to know and determine their radiological composition. Powdered milk has been analysed (Navarrete et al., 2007), soluble and grain coffee (Espinosa et al., 2009), soft drinks (Espinosa et al., 2009), around 30 medicinal plants (Espinosa et al., 2016), marine sediments and sea water in areas of the Gulf of Mexico and Coasts of Baja California (Navarrete et al., 2014); among others. The foods and plants analysed had never before resulted with contamination from ^{137}Cs, among the natural contaminants found, the highest activity reported is given by the ^{40}K content. On the other hand, for marine sediments, different types of radionuclides have been found in relatively high concentrations, to the point of being considered reservoirs of these contaminants (Navarrete et al., 2014), which seems to match results from different research groups around the world.

In this regard, the objective of this work is to confirm the presence of the fission product ^{137}Cs in strawberries (Fragaria x ananassa) produced in Mexican territory. Likewise, the activity of the contaminant in the samples is determined to compare with international recommendations. Two spectrometry equipment are used, first to detect the contaminant, a spectrometer with a Hyperpure Germanium (HPGe) detector is used, while the activity determination is made by one with a NaI(Tl) scintillation detector.

2. Experimental Theory

In order to evaluate the radiologic content of strawberries, 4 samples were analysed using semiconductor HPGe spectrometer. The HPGe analysis system is calibrated using GammaVision with the radioactive sources ^{241}Am (60 keV), ^{137}Cs (662 keV), ^{60}Co (1173 and 1332 keV) and ^{40}K (1460 keV), following the Ortec-Maestro® protocols. All samples were analysed for 24 hours.

To obtain activity of the samples, they were analysed with NaI(Tl) scintillation spectrometer. Each sample was introduced in Marinelli container, mass used was the corresponding to the volume of the container about 500 ml. Time for the analysis was chosen as 4 days, or 345,600 seconds, to obtain a clear peak for ^{137}Cs.

Specific disintegration rate of the naturally occurring radionuclide ^{40}K is given by the activity expression

$$A(t) = \left|\frac{\mathrm{d}N}{\mathrm{d}t}\right| = \lambda N(t) = \lambda N_0 \mathrm{e}^{-\lambda t}$$

Thus, activities for ^{40}K and ^{137}Cs were obtained with the corresponding half-life of each radionuclide, giving:

$$A_{^{137}\mathrm{Cs}} = \lambda N = \frac{N \ln 2}{t_{1/2}} = \frac{4.157\times10^{11}\,\dfrac{^{137}\mathrm{Cs}}{\mathrm{g}\ ^{137}\mathrm{Cs}}(0.693)}{948287520\ \mathrm{s}} = 303.790\,\frac{\mathrm{Bq}}{\mathrm{g}\ ^{137}\mathrm{Cs}}$$

$$A_{^{40}\mathrm{K}} = \lambda N = \frac{N \ln 2}{t_{1/2}} = \frac{\left(1.81\times10^{18}\right)\left(\dfrac{^{40}\mathrm{K}}{\mathrm{g\ K}}\right)(0.693)}{3.97\times10^{16}\ \mathrm{s}} = 31.685\,\frac{\mathrm{Bq}}{\mathrm{g}\ ^{40}\mathrm{K}}$$

where number of nuclei for ^{40}K was calculated as

$$N = 1\ \mathrm{g\ K}\left|\frac{1\ \mathrm{mol\ K}}{39.1\ \mathrm{g\ K}}\right|\left(\frac{6.02\times10^{23}\ \mathrm{K}}{1\ \mathrm{mol\ K}}\right)\left(\frac{0.0118\ ^{40}\mathrm{K}}{100\ \mathrm{K}}\right) = 1.18\times10^{18}\,\frac{^{40}\mathrm{K}}{\mathrm{K}}$$

and for ^{137}Cs in similar way

$$N = 1\ \mathrm{g\ Cs}\left|\frac{1\ \mathrm{mol\ K}}{137\ \mathrm{g\ K}}\right|\left(\frac{6.02\times10^{137}\ \mathrm{Cs}}{1\ \mathrm{mol\ Cs}}\right)\left(\frac{0.946\ ^{137}\mathrm{Cs}}{100\ \mathrm{Cs}}\right) = 303.790\,\frac{^{137}\mathrm{Cs}}{\mathrm{Cs}}$$

Efficiencies of these procedures have been about 5.6% for ^{137}Cs and 2.9% for ^{40}K in NaI(Tl) scintillation detector used, and 0.47% for ^{137}Cs and 0.25% for ^{40}K in HPGe detector. Detector efficiency may be calculated by dividing the number of counts per second (cps) by the theoretical ^{40}K activity in disintegrations per second (dps), relative abundance dec_γ and the mass of standard. The detection efficiency for the detector used in this study as a function of gamma ray energy of 1460 keV was calculated using the equation given by (Navarrete et al., 2005):

$$E = \frac{cps_{std} - cps_b}{A_{^{40}\mathrm{K}}\, dec_\gamma\, m_{sta}\, t}\times100$$

3. Main Results

To visualise the spectrum, MAESTRO® software was used. The preliminary detection of radiologic content in strawberries from Michoacan state (FM1) is shown in **Figure 1**.

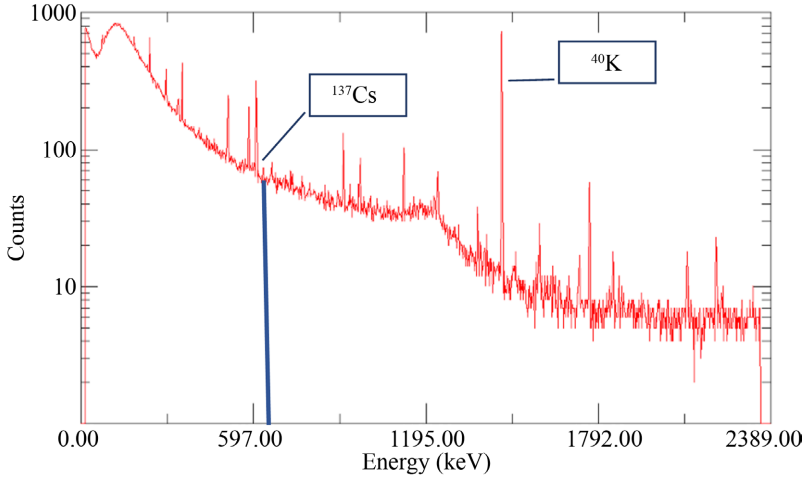

Figure 1. Spectrum of sample FM1 obtained with HPGe detector with marked ^{137}Cs photopeak.

The same way, spectrum of strawberries from State of Mexico (FSM) is shown in Figure 2.

Detection is confirmed through the spectrum data, since concentration is quite low there is a small photopeak that stands out from the baseline. Net area of the entire photopeak is given by the software and it was used instead of counts as the difference of sample energy contribution from the background energy contribution. Therefore, Net Area represents only the radionuclide contribution of the sample.

For better interpretation of the result, data from ^{137}Cs and ^{40}K is resumed in Table 1 for 3 samples from Michoacan (FM1, FM2 and FM3) and 1 from State of Mexico (FSM). Same data is shown in Figure 3.

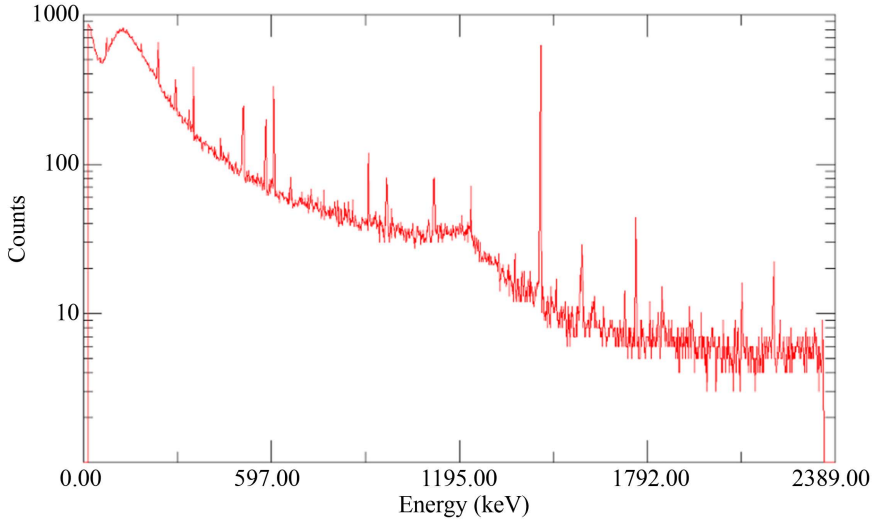

Figure 2. Spectrum of strawberries from State of Mexico obtained with HPGe detector and marked ^{137}Cs photopeak.

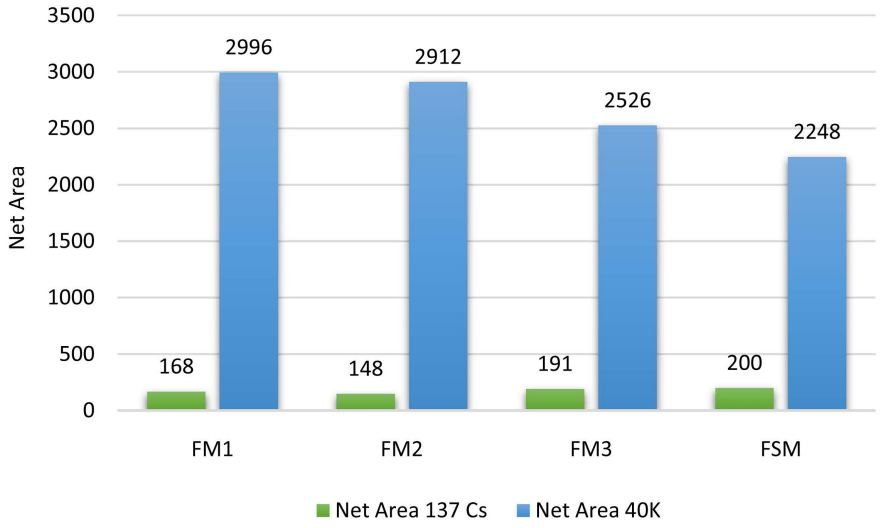

Figure 3. Data form samples analysed with semiconductor HPGe spectrometer for ^{137}Cs and ^{40}K.

Table 1. Photopeak area for samples analysed with semiconductor HPGe spectrometer.

Sample	Net Area ^{137}Cs	$A_{^{137}Cs}$ (Bq/kg)	Net Area ^{40}K	$A_{^{40}K}$ (Bq/kg)
FM1	168 ± 42	0.62	2996 ± 116	39.23
FM2	148 ± 42	0.55	2912 ± 115	33.63
FM3	191 ± 41	0.71	2526 ± 112	28.02
FSM	200 ± 74	0.74	2248 ± 125	22.42

Once the presence of ^{137}Cs was confirmed, activity determination of ^{137}Cs in the sample was made counting the number of disintegrations through gamma spectrometer with scintillation detector. As can be seen in **Figure 4**, spectrum from sample FM4 there is a clearly distinguishable photopeak for ^{40}K about 1461 keV. And about half of that distance, photopeak from ^{137}Cs at 662 keV can be seen. Scintillator detector allows a better efficiency for the determination of sample radioactive components, reducing noise from background and giving a visually smooth baseline. Therefore, the presence of both radionuclides is confirmed by the spectrum of each sample (Navarrete et al., 2005).

The rest of samples also showed presence of ^{40}K and ^{137}Cs. Spectrum from FM4, FM5 and FBC are shown in **Figure 5**. This view allows the selection of a range of energy reduced about 400 - 1600 keV and enhance complete photopeaks with red color.

Form spectrum data the number of disintegrations per second was obtained for ^{40}K and ^{137}Cs, once excluding the background contribution, counts were used to calculate activity and radionuclide content (Navarrete et al., 2005). In first instance, specific activity in the sample was calculated with number of disintegrations (cts) over time of analysis about 395,000 seconds, which were measure as counts per second (cps). Considering efficiency (E) for each radionuclide and its relative abundance (dec_γ). For total mass (W_M) amount of sample was used, calculation for ^{137}Cs in FM4 sample is shown as example

$$A_{^{137}Cs} = \frac{C_m - C_f}{E dec_\gamma W_M} = \frac{\frac{5204 \text{ cts}}{317642.62 \text{ s}}}{(0.056)(0.946)(0.3699 \text{ kg})} = \frac{0.016 \text{ cps}}{19.596 \text{ g}} = 0.0008 \frac{\text{Bq}}{\text{g}}$$

Same calculation was made with each sample to obtain activity of both radionuclides.

As can be seen in the results on **Table 2**, the amount of ^{137}Cs found in the samples is different for those produced in Baja California, about 1500 km away from Michoacan, where other samples came from. Nonetheless, some amount of the radionuclide was found for the first time in a food product. Since nuclear tests have been performed closer to Baja California, probability of finding contamination could be higher, but results showed no relation with distance from the testing place. On the other hand, the expectation was not finding any ^{137}Cs since other samples from Sonora, Baja California, and other close places have been analysed without reports of contamination (Lizarraga, 2006; Martinez, 2009).

Table 2. Results for activity from ^{40}K and ^{137}Cs in samples.

Sample	Mass (g)	Time of analysis (s)	Radioisotope	Counts	A_m (Bq/kg)
FM4	369.9	317642.62	^{137}Cs	5204	0.83
			^{40}K	1816	45.17
FM5	390.5	236401.64	^{137}Cs	3759	0.81
			^{40}K	1596	50.52
FBC	410.0	325263.56	^{137}Cs	2207	0.35
			^{40}K	1194	26.16

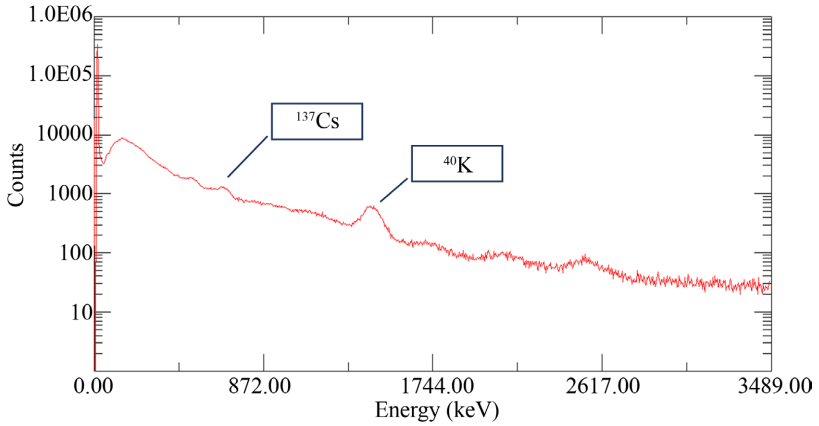

Figure 4. Spectrum from sample FM4 obtained with scintillation spectrometer.

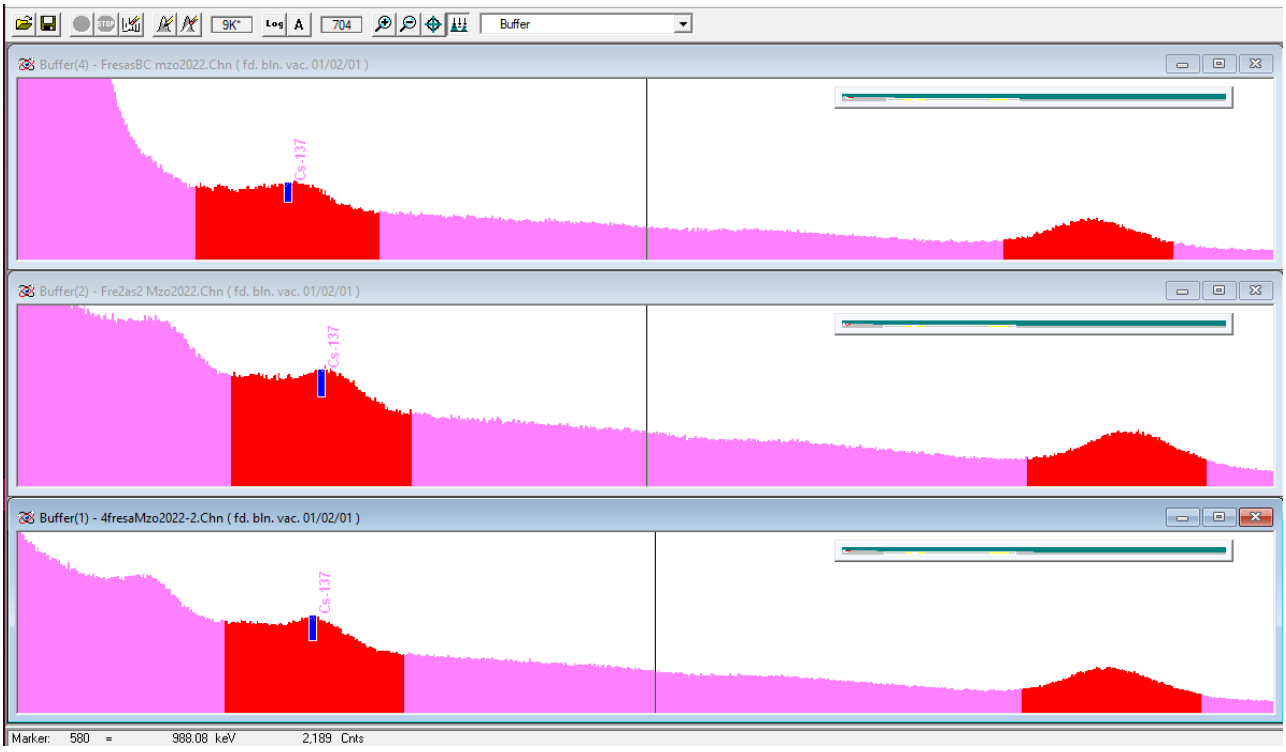

Figure 5. Spectrum from simples analysed with scintillation detector FBC (top), FM5 (middle) and FM4 (bottom), enhancing photopeak region of ^{137}Cs and ^{40}K in red and ^{137}Cs channel labelled.

An important consideration for this finding is the nature of the sample. Plants and vegetables have their own metabolism, and the distribution or bioaccumulation of nutrients depends on the type of food that is being analysed, even within the same plant it can depend on the tissue, as shown by other studies done on wild berries and mushrooms (Gwynn et al., 2013; Canbazoğlu & Doğru, 2013; Changizi et al., 2010; Tadaaki Ban-Nai et al., 2004). That is why this contamination had not been found in other foods from Mexico. It shows that some physiologic attribute of strawberry helps to better retain ^{137}Cs and may allow bioaccumulation in this fruit's tissue (Guillaume et al., 2012). There is certain relation in pathways of $^{+}$K and $^{+}$Cs absorption observed in other plants (Kumar, 2008; Dragović et al., 2004; Panchal et al., 2011). Mainly since both ions have same charge and similar properties because they belong group 1 on the periodic table (Butkus & Konstantinova, 2005), certain inverse relation between the ^{40}K and ^{137}Cs could be seen in **Figure 3**. A transfer factor could be obtained in future research for confirmation.

It is possible to know the activity of ^{137}Cs at any time, using the activity of ^{137}Cs in FM4 sample and assuming the contamination occurred around 1945 during second world war, we obtain:

$$A_0 = \frac{A_e}{e^{-\lambda t}} = \frac{0.0008 \frac{\text{Bq}}{\text{g}}}{e^{-\frac{0.693}{30}(77)}} = 0.0047 \text{ Bq/g}$$

Which is about 4.7 Bq/kg, still a concentration well below the recommendations but above natural content of ^{40}K found, for example. Still let us know the importance behind continuous research in radioactive contamination represent since those days.

The significance of the study is that this is the first study to detect and determine the presence of ^{137}Cs in fruits produced in Mexico. This is the most important fission product released to the environment result of nuclear activities, because this radionuclide rapidly passes to foodstuffs (Korobova et al., 2007). Radiation levels in food products from this region will provide data for future studies to determine level of contamination.

4. Conclusion

As consequence of multiple testing sites and several tests performed last century in different places around Mexico, after years of research and surveillance on possible radioactive contamination, today we accomplished the detection of a product of fission ^{137}Cs, released by nuclear weapons. The specific activity of ^{137}Cs present is 0.83, 0.81 and 0.35 Bq/kg in FM4, FM5 and FBC respectively. The limits established in CODEX-STAN-193-1995 for ^{137}Cs are 1000 Bq/kg (FAO, 1995), so all samples meet the recommendation. The presence of ^{137}Cs in foods produced in Mexico does not represent a food safety problem and the risk to the exposed population is minimal. Results showed the contamination with

^{137}Cs found in strawberries is well below CODEX recommendation limits and means no harm for consumer's health either affects quality of the product.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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