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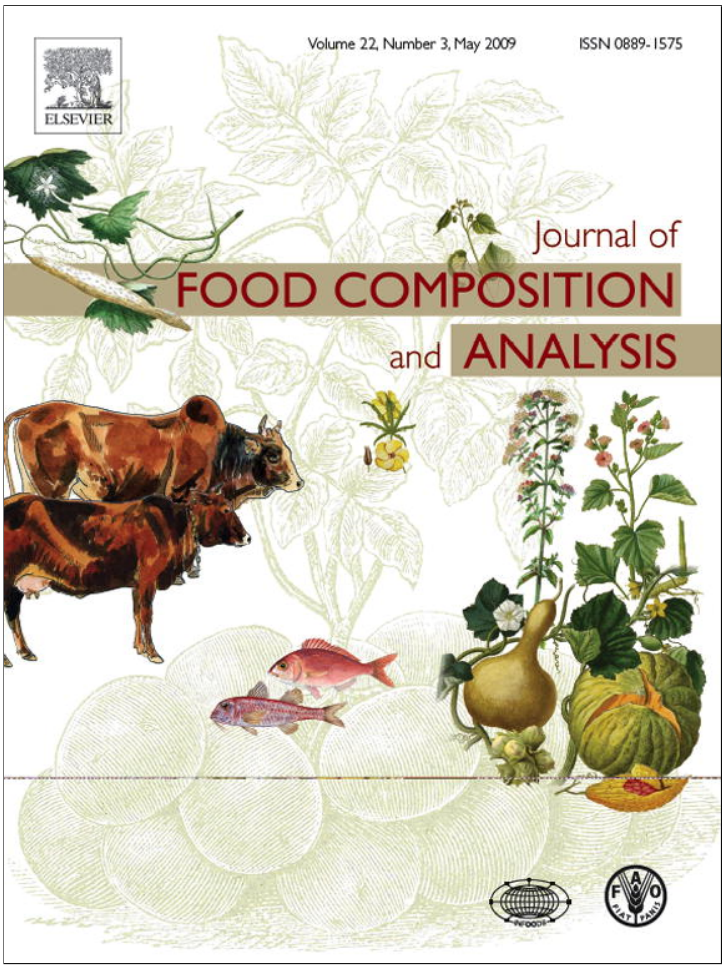

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Short Communication

Activity levels of gamma-emitters in Argentinean cow milk☆

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ABSTRACT

With the aim of contributing to the establishment of baselines for natural and man-made radioisotopes, the concentration of gamma-emitter radionuclides was determined in a set of commercial and dairy farm liquid cow milk produced in the most important production regions of Argentina using a gamma-ray spectrometer with a high-resolution HPGe detector. For comparison, samples from Chile and Uruguay were also analyzed. The main detected activity corresponded to ^{40}K, while the activities of elements of the natural chains were below the detection limit. The determined ^{40}K average activity for the Argentinean milk was 60 Bq/L. Traces of the anthropogenic radionuclide ^{137}Cs were observed only in the Chilean milk sample.

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1. Introduction

There are many sources of radiation and radioactivity in the environment. The earth and the atmosphere present different activity levels of natural radionuclides such as those of the ^{238}U and ^{232}Th decay chains as well as the ^{40}K. To begin with, during the 1960s and 1970s, the South American environment was polluted by the radioactive fallout originating from the nuclear weapons tests performed in the South Pacific area (Schuller et al., 1993, 2002, 2004; Arnaud et al., 2006; UNSCEAR, 1982). Additionally, nuclear industry, mining, crude oil and gas manufacturing also contributed to the radionuclide inventory of the environment. All these nuclides, natural and man-made, have radiobiological significance because they can be incorporated into the human body from the food chain and also through inhalation of suspended dust in the air, constituting a direct route for internal exposure. Therefore, the verification of the natural and artificial concentration levels in food supplies is an important parameter for evaluating abnormalities. Consequently, international organizations have recognized the need of the assessment of radiation doses received by the population (UNSCEAR, 2000). In this sense, the build-up of baselines of natural and anthropogenic concentration of radioisotopes in food represents an important issue. However, in Argentina very few surveys of radioactivity in food have been carried out (Ciallella et al., 2003), and no baselines of concentration of natural and anthropogenic radioisotopes have been reported.

Since milk and milk products are important components of human diet, especially in Argentina (INDEC, 2005), the establishment of radioisotope concentrations will provide meaningful information that can contribute to the knowledge of population exposure and to the setting up of a regional baseline (IAEA, 1989). In Argentina, milk is produced over large areas (approximately 1,000,000 km^2) and collected daily at dairy farms.

A first systematic study of the gamma-emitter radionuclide content (natural and anthropogenic) in widely consumed Argentinean milk produced in the principal productive regions is presented. Thirty-one commercial farm cow and ranch cattle liquid milk samples coming from different productive regions of

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Argentina and neighboring countries were analyzed. The samples were collected over a long time period (from 2000 to 2007). During this period, no climatic anomalies were registered. Also natural phenomena (such as volcanic activity) or nuclear accidents that may have an impact on redistribution of radionuclides on the grass eaten by the cows producing milk did not occur. Due the long time period monitored and the extended region studied, the present results could contribute to a baseline for gamma-emitter radionuclide content in Argentinean milk. Results are compared with those of different geographic locations worldwide, and committed doses are evaluated.

2. Material and methods

Samples were collected from 2000 to 2007. To ensure a representative sampling, several samples of the same commercial milk were collected during different years and seasons. One liter of each milk sample was dried at 80 °C to obtain a solid residue and then milled in a ceramic mortar. The solid residue was properly accommodated in 200 mL Petri dishes (9 cm diameter × 3 cm height), then sealed and kept for nearly three weeks before analysis to achieve secular equilibrium. For measurements, similar amounts of dry residue were used and geometry was preserved. The main characteristics of the samples are displayed in Table 1.

A high resolution HPGe gamma EG&G Ortec spectrometer in the range of 30–1800 keV with a standard electronic chain and 8192 channels multichannel, and an EG&G Ortec low background chamber were used. Energy calibration was performed with ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu sources. The efficiency calibration was carried out using an admixture of known amounts of naturally occurring ^{176}Lu and ^{138}La long-lived isotope, dispersed in a milk substrate (Perillo Isaac et al., 1997). The absolute efficiency and the energy resolution at 1332 keV ^{60}Co line were 0.5% and 1.9 keV, respectively. Room background was determined and peak background corrections were done to the corresponding milk spectrum. In these corrections, only gamma-emitter nuclides belonging to the ^{235}U, ^{238}U and ^{232}Th chains, ^{7}Be and ^{40}K were taken into account. The spectra were recorded during seven days and analyzed with a commercial program.

Table 1
Main characteristics of the samples ranked by province of collection, country and type (C: commercial milk, NC: non-commercial). The resulting solid residua after drying 1 L are also given.

Sample	Province	Country	Type	Solid residue (g/L)
1	Buenos Aires	Argentina	C	106
2	Buenos Aires	Argentina	C	112
3	Buenos Aires	Argentina	C	106
4	Buenos Aires	Argentina	C	111
5	Buenos Aires	Argentina	C	119
6	Buenos Aires	Argentina	C	95
7	Buenos Aires	Argentina	C	100
8	Buenos Aires	Argentina	C	102
9	Buenos Aires	Argentina	C	77
10	Santa Fe	Argentina	C	106
11	Santa Fe	Argentina	C	113
12	Santa Fe	Argentina	C	102
13	Santa Fe	Argentina	C	109
14	Santa Fe	Argentina	C	112
15	Santa Fe	Argentina	C	102
16	La Pampa	Argentina	NC	85
17	La Pampa	Argentina	NC	122
18	La Pampa	Argentina	NC	126
19	La Pampa	Argentina	NC	135
20	La Pampa	Argentina	NC	117
21	La Pampa	Argentina	NC	124
22	Córdoba	Argentina	C	86
23	Córdoba	Argentina	C	105
24	Córdoba	Argentina	NC	102
25	Córdoba	Argentina	C	83
26	Córdoba	Argentina	C	96
27	Córdoba	Argentina	C	100
28	Tucumán	Argentina	C	127
29	Mendoza	Argentina	NC	83
30	Montevideo	Uruguay	C	102
31	X° Región	Chile	C	108

3. Results and discussion

The principal productive regions are located at Santa Fe Province (37% of the total production), Córdoba Province (36%), and Buenos Aires Province (23%), and more than 90% of the production is used for internal consumption (Fig. 1a). One liter of commercial milk resulted, in some cases, from an admixture of the production of dairy farms located at widely separated areas. Liquid commercial and cattle ranch milk samples, widely consumed in Argentina and collected from the main production regions, as indicated in Fig. 1b, were analyzed.

Selected recorded gamma spectra are shown in Fig. 2, where the characteristic lines associated with the nuclides of the U and Th chains are observed together with that corresponding to ^{40}K. No anthropogenic isotopes were detected. The activity of ^{137}Cs resulted below the detection limit (0.06 Bq/L), except for the Chilean sample, as observed in the inset of Fig. 2. In this sample, the only anthropogenic detected radionuclide was ^{137}Cs with an activity of 0.11 ± 0.05 Bq/L, well below the guideline level for this radionuclide in food, milk and infant food (Codex Alimentarius Commision, 2004). In the Southern Hemisphere, ^{137}Cs activities due to fallout were previously reported, ranging from 0.05 to 0.11 Bq/L and 0.013 to 0.09 Bq/L, for milk from New Zealand (Hermanspahn, 2007) and Chile (Schuller et al., 1993), respectively. Regarding this observed radioactivity, the absence of ^{137}Cs in Argentinean and Uruguayan samples could be related to the Andes Mountains, which act as a barrier to rains and winds from the West (Schuller et al., 2004; Arnaud et al., 2006). In effect, according to the data published by UNSCEAR, the ^{137}Cs activity determined in milk samples recorded between 1976 and 1979 in Argentina was many times lower than that determined in Chilean milk samples for the same period (UNSCEAR, 1982).

Concerning the natural nuclides, the measured activity corresponds mainly to ^{40}K, the activities of the constituents of the U and Th chains below the detection limits (2 Bq/L and 1 Bq/L for the U and Th chain, respectively) after peak background correction. The corresponding ^{40}K activity values are given in Fig. 3, varying from 36 Bq/L to 76 Bq/L for the Argentinean milk, with a mean value of 60 Bq/L. Similar ^{40}K activity levels were found in the Chilean and Uruguayan samples (see Fig. 3). The average activity value of the milk collected in Buenos Aires Province (54 Bq/L) is the lowest compared with the average values of the other provinces.

To go further with the discussion, a compilation of ^{40}K activity values in milk samples around the world is presented in Table 2. It is important to remark that the ^{40}K activity levels determined in the present study are similar to those of milk consumed in other countries, with the exception of milk imported by Nigeria, which presents higher activity levels (Osibote et al., 1999).

Internal exposures arise from the intake of terrestrial radionuclides by inhalation and ingestion. Doses by inhalation result from the presence of dust particles in air containing radionuclides of the ^{238}U and ^{232}Th decay chains, which are the dominant components of inhalation exposure, the short-lived decay products of radon. Doses by ingestion are due mainly to ^{40}K and to the ^{238}U and ^{232}Th series radionuclides present in food and drinking water. The dose rate from ^{40}K can be determined directly and accurately from external measurements of its concentration in the body. However, the analysis of the radionuclide contents of food and water, along with bioassay data and the knowledge of the

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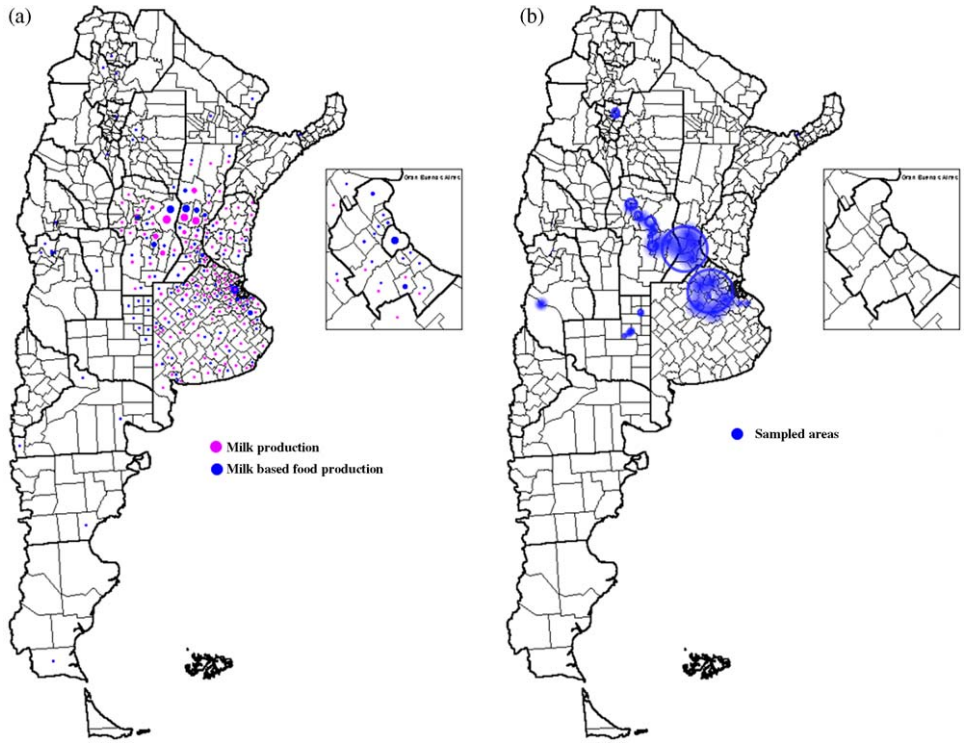

Fig. 1. (a) Location of the main productive Argentine regions and (b) sampling sites.

metabolic behavior of the radionuclides, provides an alternative basis for dose estimation. Ingestion intake of natural radionuclides depends on the consumption rates of food and water and on the radionuclide concentrations. With these data, doses for children as well as adults can be derived.

The consumption of milk varies widely around the world, depending on climate, food availability and cultural dietary preferences. In the present article, in order to determine the annual committed effective dose of ^{40}K intake, the accepted dose coefficients (UNSCEAR, 2000) and the annual milk consumption by individuals in Argentina (SAGYPA, 2007) were taken into account. An overall consumption rate of 219 L per inhabitant per year has been reported without discrimination of inhabitant age (SAGYPA, 2007). After that, the average of the dose coefficient was calculated weighting the dose coefficients with the population pyramid

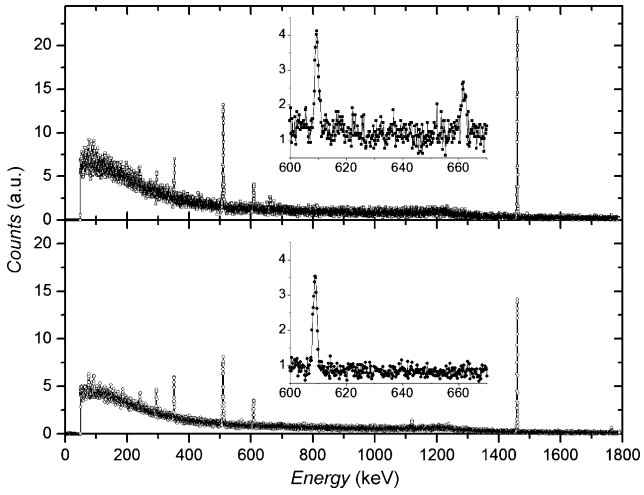

Fig. 2. Typical gamma-ray spectra recorded from the Chilean sample (top) and a selected Argentine sample (bottom). The inset corresponds to the 662 keV energy region (^{137}Cs energy).

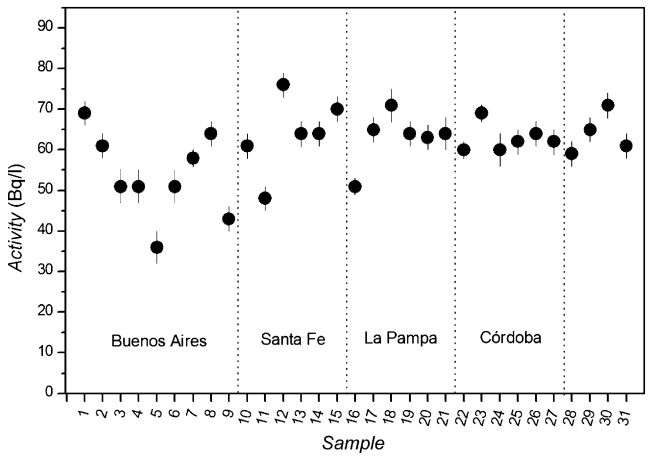

Fig. 3. ^{40}K determined activities labeled with the Argentinean province and foreign country. Samples 28–31 correspond to Tucumán and Mendoza Provinces (Argentina), Montevideo (Uruguay), and X° Región (Chile), respectively. The error bars are the total uncertainties of the determined activities.

Table 2
^{40}K activities reported for milk consumed in different countries.

Country	Activity (Bq/L)	Reference
Argentina	50 to 76	Present results
Brazil	53 to 55*	Melquíades and Appoloni (2001)
Costa Rica	37 to 56	Loría et al. (2007)
France	48*	Hosseini et al. (2006)
Germany	68*	Hosseini et al. (2006)
Hong Kong	48	Yu and Mao (1994)
Israel	49 to 52*	Lavi et al. (2006)
New Zealand	61 to 67*	Hosseini et al. (2006)
Nigeria	85 to 105*	Osibote et al. (1999)
Spain	32 to 64	Baeza et al. (2004)
Syria	54*	Al-Masri et al. (2004)
Syria for infants	14 to 48*	Al-Masri et al. (2004)
Ukraine	44	Shiaishi et al. (1997)
Venezuela	45*	La Becqque et al. (1992)

* Equivalent to 1 L of liquid milk prepared from powder milk.

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(INDEC, 2005), resulting in a value equal to 10.8×10^{-9} Sv/Bq. After this, the annual committed effective dose per unit intake of ^{40}K has been estimated ranging from 80 μSv to 180 μSv, with the average equal to 143 μSv. The net radiological impact due to milk intake is close to levels reported for worldwide average exposure (170 μSv) due to ^{40}K (UNSCEAR, 2000) and 24% lower than the ICRP recommended annual limit dose (222 μSv) (ICRP, 1991).

4. Conclusions

A systematic study of radioactivity in Argentinean milk is presented for the first time. The extended region and the long period of time investigated will help to establish a baseline of the natural and anthropogenic gamma-emitter radioisotopes present in this important component of the human diet of South America. For comparison, milk samples from Uruguay and Chile were also measured. The main gamma detected activity arises from ^{40}K, which is the determined value closer to the values recommended by UNSCEAR and ICRP and similar to that observed in other regions around the world. The activities of the natural radionuclides belonging to the U and Th chains were below the detection limits. Anthropogenic ^{137}Cs traces were detected only in the Chilean milk sample, possibly as a result of South Pacific nuclear weapon tests performed in the 1960s. The ^{137}Cs activity was close to the one determined for New Zealand milk.

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