



# Radioactivity in milk consumed in Nigeria 10 years after Chernobyl reactor accident

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## Abstract

The average concentrations of the radioactivity in milk imported into and consumed in Nigeria, 10 years after Chernobyl-4 nuclear power plant accident, have been measured by means of a well-calibrated high-purity germanium detector. The photopeaks observed with reliable regularity belong to the naturally occurring series-decay radionuclides headed by  $^{238}\text{U}$  and  $^{232}\text{Th}$ , as well as the non-series decay type,  $^{40}\text{K}$ . Ten years after the nuclear accident,  $^{137}\text{Cs}$  was not detected in any of the milk samples in view of the possible transfer through the soil–grass–cow–milk route, the time interval being one third the  $^{137}\text{Cs}$  half-life (30.2 yr). The average total specific activity values of  $23.07 \pm 7.75$ ,  $4.35 \pm 2.06$  and  $831.66 \pm 54.83 \text{ Bq kg}^{-1}$  for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , respectively, were obtained. © 1999 Elsevier Science B.V. All rights reserved.

*Keywords:* Radionuclides; Specific activity concentrations; Gamma spectroscopy analysis

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## 1. Introduction

The most severe civil nuclear reactor accident at Chernobyl power station [1–3] occurred on 26 April 1986. The post-impact assessment programme for the estimation of ingestion dose as a component of radiation burden of the populace in Nigeria was put in place because of the worldwide distribution of its radioactive effluents. As part of this assessment, there is a need to measure the radioactivity in foods, particularly in milk which

forms a major component in the human food chain because it is consumed by people of various ages and sexes. After the decay of the short-lived radionuclides which formed the earlier phase of population radiation exposure, the later phase was the radioactive fallout consisting of caesium isotopes ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ) as well as isotopes of strontium and plutonium which accounted for the radiobiological significance of contamination as they were released into the environment, thereby finding their way into the terrestrial food chain. These radionuclides, when accumulated in foods and water, constitute a direct route of exposure to human population when the contaminated foods and water are consumed [4–6]. Thus, in the longer term, the concern for human health, ingestion doses

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from caesium isotopes being long-lived radionuclides ( $^{137}\text{Cs}$  with half-life of 30.2 yr,  $^{134}\text{Cs}$  with half-life of 2.1 yr) pose a greater problem since these isotopes will persist in the environment for many years. The resulting consequence of the ingestion doses is an increase in the incidence of cancers. The regularities revealed in  $^{137}\text{Cs}$  redistribution in natural and agricultural systems as well as the migration through the soil–grass–cow–milk food chain in many countries after the Chernobyl-4 reactor accident made it essential to take stock of radioactivity content of milk after a decade. Milk and milk products are important components of the diet of both infants and adults in many countries including Nigeria. Milk is one of the few foods produced over large areas and collected on a daily basis from dairy cattle. Contamination of milk will be greatest when cows are grazing during the fall-out periods, but when cows are kept indoors contamination of milk may also occur by inhalation of radionuclides or ingestion of radionuclides in drinking water and contaminated feed.

In the United States of America, Broadway and his co-workers in 1988 [7] reported the estimates of both individual and collective doses received by the United States inhabitants following the Chernobyl nuclear power plant accident. They estimated 20 excess fatalities from the ingestion of  $^{137}\text{Cs}$  in milk during all subsequent years with six of these due to lung cancer and the majority of the remaining distributed approximately equally among cancers of the thyroid, breast, liver, and leukemia. To date, concerning thyroid effects, only three children in the cohort of diagnosed cases have been reported to have died of thyroid cancer [8].

The decline in the caesium radioisotope levels has been studied in tissues from lambs grazing lowland pastures in United Kingdom by Martin et al. [9]. They reported that the  $^{137}\text{Cs}$  concentration in grass from the field grazed by the lambs decreased with a half-time of 22 d over the period of 11–100 d after contamination. The amount of caesium radionuclides removed from the pasture by the grazing animals amounted to only 0.01% of the total, the rest remaining in the soil, with over 40% in the upper 10 mm and small amounts of  $^{110m}\text{Ag}$ , found in grass, declined with a half-time of 8.9 d, and the radionuclide was found to accumulate in liver tissue.

In a cooperative research project initiated in June 1986 between Colorado State University, USA and National committee for technical development, Ward et al. [10] reported the transfer coefficient for  $^{137}\text{Cs}$  from the Chernobyl accident to milk of cows and sheep in Hungary both fed on forage harvested within one month of the accident. They observed that the transfer coefficient for milk and for meat were lower than results reported for worldwide fallout from weapons tests. Furthermore, Ward and his co-workers [10] observed that forage harvested 60 d or later after the accident produced a transfer coefficient for  $^{137}\text{Cs}$  for milk similar to results from feeding soluble  $^{134}\text{Cs}$ . Tracy and his group [11] reported their work on transfer to milk of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  released during the Chernobyl reactor accident. They observed that dry disposition was more significant for  $^{131}\text{I}$  and wet disposition for  $^{137}\text{Cs}$  and that the concentration of iodine in milk measured in  $\text{Bq l}^{-1}$  were 1000–2000 times the concentration of particulate  $^{131}\text{I}$  in air measured in  $\text{Bq m}^{-3}$ . The transfer of iodine from grass to milk according to them was consistent with the predictions of conventional models. Furthermore, they reported that about 10% of reported caesium was intercepted by the edible portion of the grass while transfer of caesium from grass to milk, following a major pathway of exposure to fallout of air–grass–cow–milk food chain was an order of magnitude lower than that predicted by conventional models.

In Nigeria, Farai [17] reported  $^{137}\text{Cs}$  activity concentrations ranging from 0.5–5.1  $\text{Bq kg}^{-1}$  in eleven widely consumed imported milk products and infant cereals manufactured in European countries at least 6 months after the reactor accident. In a country of over 100 million people, most of the cows reared are not used for dairy purposes but for their meat consumed by the Nigerian population. Thus, the milk consumed by the vast majority of Nigerians is imported from countries with functional commercial dairies.

The purpose of this study is to identify the type of radionuclide still present and to determine the  $\gamma$ -activity levels in the widely consumed milk imported from some countries in Europe which were once contaminated with radioactive effluents from the damaged Chernobyl nuclear power reactor.

## 2. Materials and methods

Large samples of various instant powdered milk imported from United Kingdom, Ireland, Denmark, Switzerland, France and Netherlands were collected between 26 April and 10 May 1996 from departmental stores located in many metropolitan cities: Lagos, Ibadan, Benin City, Ilorin, Minna, Abuja, Kaduna, Jos, Enugu, Sokoto, Maiduguri, Ile-Ife, Akure and Port Harcourt (as shown in Fig. 1 and listed in Table 1). The milk samples came from the above-mentioned, one-time radionuclide-polluted countries in Europe (as shown in Fig. 2). A total of nine different makes of instant powdered milk produced in the aforesaid six countries was used for this study. The composi-

tion of milk is almost identical the world over and it is easy to collect a representative sample that can be analyzed in liquid or powdered form. Ten 1-liter Marinelli beakers, i.e. one for each milk sample and one for recording the background counts, were washed, rinsed with diluted  $H_2SO_4$  acid to prevent the samples from being contaminated, and then dried. These clean 1-liter Marinelli beakers are then filled with known masses of milk samples which have been dried to constant weight and then sealed for 28 d to allow a sufficient time to attain a state of secular radioactive equilibrium prior to  $\gamma$ -spectroscopy. The  $\gamma$ -counting equipment used consists of a Canberra vertical high-purity coaxial germanium (HPGe) crystal (model GC2018-7500 serial number b 87063) enclosed in a 100 mm thick lead shield.

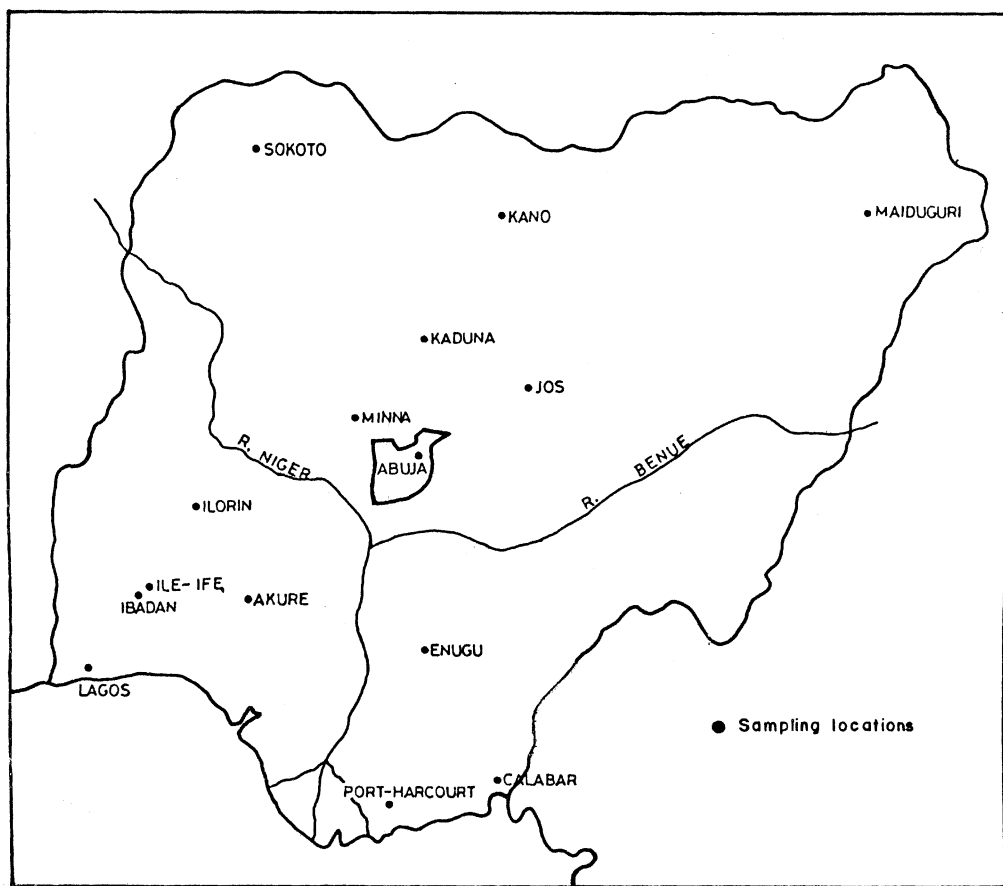


Fig. 1. Map of Nigeria showing the sampling locations.

The HPGe detector was connected to a Canberra computer-assisted Multichannel Analyzer (MCA). Accurate energy and efficiency calibrations of the gamma-spectrometry system were made using

Table 1  
Milk samples, their countries of origin and the sampling locations in Nigeria

Powdered milk samples (trade name)	Country of origin	Sampling locations
Peak	Netherlands	Ibadan, Jos
Coast	Netherlands	Kaduna, Lagos
Carnation	Netherlands	Sokoto, Port Harcourt
Nido	Netherlands	Ile-Ife, Kano
Holt	Denmark	Ilorin, Enugu
Cowbell	Switzerland	Abuja, Maiduguri
France Lait	France	Ibadan
Klim	Ireland	Minna, Calabar
Mix Me	United Kingdom	Lagos, Enugu

a standard source of radionuclides present in a sample since the accuracy of all quantitative results depends on the attainable accuracy of the system's calibration [12]. The description of the gamma-spectrometry system has been well documented in earlier publications [4,12]. The sealed milk samples were placed over the HPGe detector for counting, and counting times for accumulating spectral for the milk samples and their backgrounds were set at 36000s while the same geometry was employed for the counting of both milk samples and background. Each milk sample was counted twice in order to check the stability of the counting system.

The  $\gamma$ -spectroscopy analysis employed in this work was based on a computer program SAMPO 90 which matched  $\gamma$ -energies at various energy levels to a library of possible isotopes. This data analysis routine subtracted a linear background distribution from pulse-height spectra of both

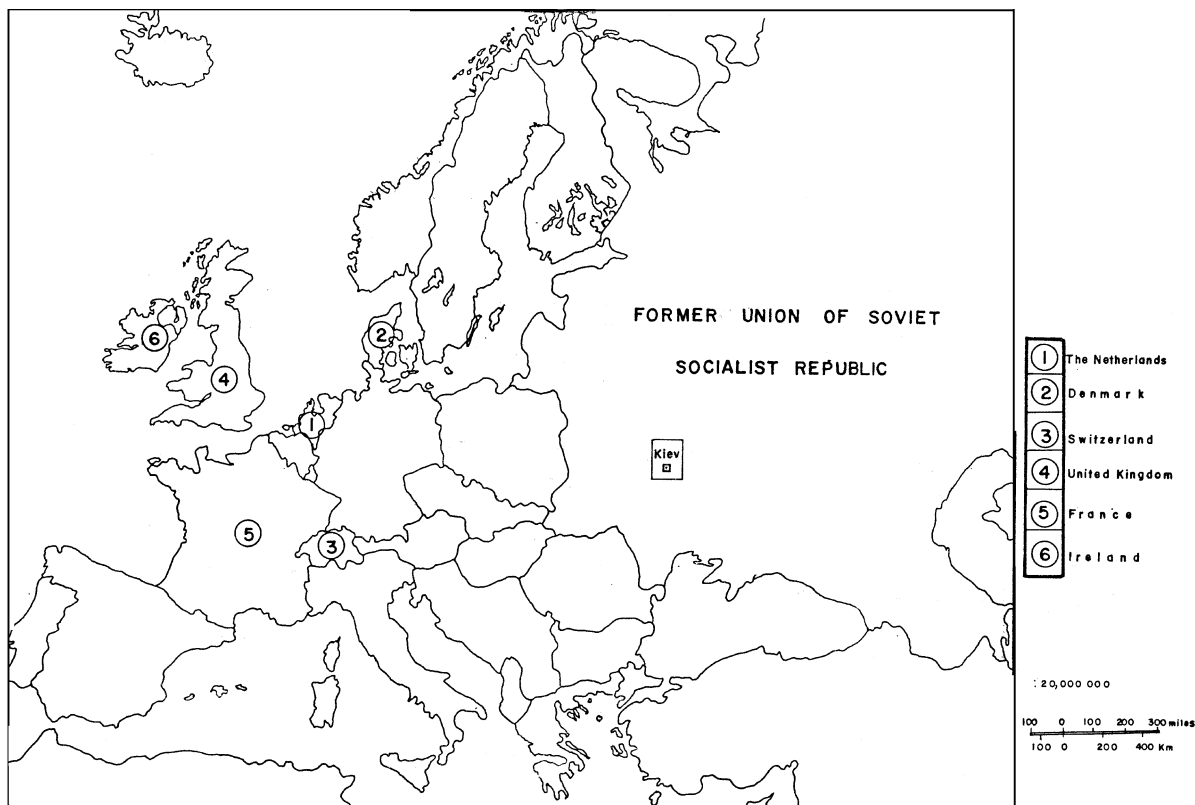


Fig. 2. Map of Europe showing the origin of the imported milk samples.

sample and background in addition to the net background peak area being subtracted from the corresponding net peak area for a particular radionuclide. The characteristics of  $\gamma$ -emitting isotopes used as monitor nuclides are as listed in an earlier work [12].

### 3. Results and discussion

The average specific activity concentrations of the radionuclides identified with reliable regularity in all the milk samples analyzed by gamma-spectrometry are given in Table 2. The photopeaks observed with regularity in the milk samples were identified to belong to the naturally occurring series-decay radionuclides headed by  $^{238}\text{U}$  and  $^{232}\text{Th}$ . The third natural radionuclide is a non-series one,  $^{40}\text{K}$ . Of these two series-decay radionuclides, it is well established that radium isotopes are of major concern as a source of dietary contamination and consequently of serious internal radiation to man [13]. For the uranium series, most of the  $\gamma$ -rays are emitted during the decay of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  but the latter is the principal contributor to internal dose rate as documented by the NCRP [14]. Similarly,  $^{208}\text{Tl}$  in the thorium series contributes most to the dose rate. Thus, the activity concentration of  $^{214}\text{Bi}$ , determined from its 609.3 keV photopeak, was chosen to provide an estimate of the parent  $^{226}\text{Ra}$  nuclide while the daughter radionuclide  $^{208}\text{Tl}$  whose activity concentration was determined through its 583.0 keV

photopeak, was selected as an indicator of  $^{228}\text{Ra}$ . The emission line at 1460.8 keV was used for the estimation of  $^{40}\text{K}$  activity concentration. The total specific activity from each radionuclide determined at two-standard-deviation analytical error was calculated on the basis of a mean value obtained from the two measurements made for each sample. The concentration of  $^{226}\text{Ra}$  ranged from 17.56 to 41.42  $\text{Bq kg}^{-1}$  with a mean of  $23.07 \pm 7.75 \text{ Bq kg}^{-1}$ , the concentration of  $^{228}\text{Ra}$  recorded a range of 0.96–7.77  $\text{Bq kg}^{-1}$  with a mean of  $4.35 \pm 2.06 \text{ Bq kg}^{-1}$  and the concentration of  $^{40}\text{K}$  ranged from 772.42 to 949.21  $\text{Bq kg}^{-1}$  with a mean concentration of  $831.66 \pm 54.83 \text{ Bq kg}^{-1}$ .  $^{40}\text{K}$  recorded the highest average total specific activity in all the milk samples and, hence, will have the highest contribution to ingestion doses of the consumers. However,  $^{40}\text{K}$  is an essential biological element and its concentration in human tissue is under close metabolic (homeostatic) control [15]. It may appear strange that none of the long-lived fission products, e.g.  $^{137}\text{Cs}$ , was detected in any of the milk samples judging by the initial environmental radionuclide pollution. However, the answer may lie in the fact that radiocaesium has short biological half-life of the order of 2–4 weeks as well as possible countermeasures which were applied since the first few weeks after deposition of the long-lived radioactive effluents. Richards and Hance in 1996 [16] highlighted such possible countermeasures, namely, applications of high levels of potassium fertilizer can reduce radiocaesium uptake because mineral uptake by

Table 2  
Mean radionuclide activity concentrations ( $\text{Bq l}^{-1}$ ) in imported powdered milk

S/N	Milk sample	Radionuclides		
		$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{40}\text{K}$
1	Peak	$23.57 \pm 1.90$	$4.21 \pm 0.55$	$815.10 \pm 80.49$
2	Coast	$17.97 \pm 1.59$	$0.96 \pm 0.39$	$793.47 \pm 78.39$
3	Carnation	$18.34 \pm 1.58$	$3.35 \pm 0.49$	$802.48 \pm 79.08$
4	Holt	$18.97 \pm 1.67$	$4.92 \pm 0.66$	$775.94 \pm 76.79$
5	Cowbell	$18.91 \pm 1.66$	$4.85 \pm 0.60$	$772.42 \pm 76.47$
6	France lait	$17.56 \pm 1.56$	$4.86 \pm 0.58$	$843.06 \pm 83.13$
7	Nido	$18.36 \pm 1.58$	$1.54 \pm 0.40$	$839.54 \pm 82.67$
8	Klim	$32.57 \pm 3.14$	$7.77 \pm 1.14$	$949.21 \pm 97.18$
9	Mix Me	$41.32 \pm 3.55$	$6.68 \pm 1.03$	$893.72 \pm 91.38$

plants is related to total availability and relative abundance of their different ions, limiting by increasing calcium levels can reduce radiostrontium uptake, cultivating crops such as sugar-beet or oilseed rape where the edible products are processed and contamination reduced, burying the contaminated surface of the land by deep ploughing could be an effective procedure for large farms provided that proper ploughs are available because  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are widely distributed in near-surface soils with their concentrations usually decreasing roughly exponentially with depth, feeding uncontaminated stored feedstuffs and, more effectively, using Prussian blue.

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