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Uranium, polonium and thorium in infant formulas (powder milk) and assessment of a cumulative ingestion dose

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ABSTRACT

It is well known that all food and foodstuffs contain naturally occurring radionuclides originating from uranium and thorium decay chains. Since a dose coefficient is always related to a specific radionuclide, it is therefore necessary to determine the activity concentrations of particular radionuclides when completing a radiological risk assessment. Dose coefficients, however, are age dependent, with the highest values being prescribed for infants. Due to the fact that the data on particular radionuclide content in infant formula are scarce, the aim of our research was their determination in infant formulas available on the Slovenian market. ^{238}U, ^{234}U, ^{230}Th and ^{210}Po activity concentrations were determined in five samples and dose assessment was carried out with dose coefficients listed in the IAEA International Basic Safety Standards (2014). The results obtained show that the main contributors to the estimated cumulative radiation dose (230 to 350 μSv y^{-1}) is ^{210}Po.

1. Introduction

According to the data available online (UNICEF, 2014; CDC, 2014; WHO, 2015) less than 40% of infants worldwide under six months are exclusively breastfed. The reasons for this low percentage vary, with the most common being a lack of mother's milk and socio-economic conditions (e.g. working mothers).

The diets of infants that are not exclusively breastfed are supplemented or completely comprised of powdered milk (baby formula) which is a special synthetic supplement designed to provide nutrients necessary for the normal development of infants. Depending on the infant's needs several different types of infant formulas are available on the market. Among the most common are those based on cow's milk (most infants do not have a problem ingesting cow's milk). However, for infants with strong sensitivity to cow's milk and for infants with other formula related medical or digestive conditions there are many special products available on the (Slovenian) market. (Novalac, 2016; Aptamil, 2016; Hipp, 2016)

The research on alpha-emitting radionuclides in infant formulas are scarce. Prabhath et al. (2015) reported ^{210}Po activity concentrations and the committed effective dose associated with it in Mumbai, India. The results showed that activity concentrations of ^{210}Po vary from 0.08 to 0.23 Bq/kg and that the average annual effective dose by ingestion of infant formulas is 150 μSv. Uwatse et al. (2015) determined ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in 14 brands of powdered milk for infants from various regions around the world. The estimated annual effective doses for infants under 1 year was 635.13 μSv y^{-1}. Additionally, Štrok and Smodiš (2011) reported activity concentrations of ^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb and ^{210}Po in infant formulas available on the Slovenian market. Their results showed that the highest combined annual effective ingestion dose for infants is 648 ± 98 μSv with the main contribution originating from ^{210}Po and ^{210}Pb.

For infants (< 1 years old), who form one of the most sensitive segments of the population, it is important to consider their exposure to different food contaminants, including various naturally occurring radionuclides. Infants in particular have a greater intestinal absorption and lower threshold for adverse effects than adults (Tripathi et al., 2001; Fergusson, 1990). In light of this information, we have decided to analyze activity concentrations of the natural alpha-emitting radionuclides (^{238}U, ^{234}U, ^{230}Th and ^{210}Po) in the most commonly used infant formulas available on the Slovenian market.

2. Materials and methods

All reagents used in the analysis were of analytical grade. The tracer solutions ^{232}U (SRS 82712-482), ^{209}Po (SRS 82710-482) and ^{229}Th (SRS 82711-482) and used in the study were prepared from calibrated solutions purchased from Analytics, Inc. (Analytics, Inc., Atlanta, GA, USA). The producer maintains traceability to the NIST (NIST, Gaithersburg, MD, USA). Uranium (U) standard solution (SRM 3164) was obtained from NIST. The extraction resins employed in this work was UTEVA®, available from Triskem International (Triskem

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International, Brus, France).

Five samples (S1–S5) of infant formulas for infants aged 0–12 months from two different producers (Nutricia Zakłady Produkcyjne sp. z o.o., Poland (Aptamil) and UP Medi-Europa SA, Swiss (Novalac)) were analyzed.

Tap water from Ljubljana, Slovenia was also analyzed.

An alpha spectrometer (CANBERRA's Alpha Analyst™; Canberra Industries, Meriden, CT, USA) with passivated implanted planar silicon (PIPS) semiconductor detectors with an active area of 450 mm^2 and 28% efficiency for 25-mm diameter discs was used for alpha-particle spectrometry measurements. The measured source was placed in a parallel plane, centered at the symmetry axis of the detector at a distance (varying a bit amongst chambers) of about 5.0 ± 0.5 mm. The calibration of the detectors was made with a standard radionuclide source containing ^{238}U, ^{234}U, ^{239}Pu and ^{241}Am (code 67978-121), obtained from Analytics, Inc.

Water samples (~3 mL) were irradiated in the Institute's TRIGA MK II reactor in the pneumatic tube (rabbit system) at a neutron fluence rate of $4{\cdot}10^{12}$ $ncm^{-2}s^{-1}$ for up to 5 min with a uranium standard (~100 ng, NIST, Gaithersburg, MD, USA).

Infant formula samples (~ 0,5 g) were irradiated in the Institute's TRIGA MK II reactor in the pneumatic tube (rabbit system) at a neutron fluence rate of $4{\cdot}10^{12}$ $ncm^{-2}s^{-1}$ for 90 s with a uranium standard (~100 ng).

When ^{238}U is irradiated in a reactor the following capture reaction takes place:

$$^{238}U(n,\gamma)^{239}U(t_{1/2} = 23.5\ min) \rightarrow {}^{239}Np\ (t_{1/2} = 2.35\ d) \rightarrow$$

Gamma-ray measurements of isolated uranium fraction after irradiation and added U-235 for determination of recovery s were conducted by well-type HPGe detector with an active volume of 277 cm^3 (well diameter 26 mm, well depth 45 mm), having an absolute efficiency of 5.6% at 122 keV. Direct gamma-ray measurements of samples (approx. 30 g) were conducted with a coaxial HPGe detector (ORTEC GEM-30, 37% relative efficiency and 1.8 keV resolution for ^{60}Co at 1332 keV line). The detector was calibrated with a certified reference material "simulated vegetation" 85344-443 obtained from Eckert & Ziegler (Eckert & Ziegler, Berlin, Germany). The reference material and samples were measured in identical cylindrical containers. The density corrections were applied using EFFTRAN software (Vidmar et al., 2011; Vidmar, 2005). All spectra were evaluated using Genie-2000® software (Canberra Industries, Meriden, CT, USA).

For thermal fusion dissolution, a Cleise LeNEO furnace (Corporation Scientifique Claisse, Ville de Québec, QC, Canada) in combination with a platinum crucible and a Teflon beaker was used.

2.1. Radiochemical procedures

2.1.1. Determination of uranium mass concentration by radiochemical neutron activation analysis (RNAA)

Each sample (0.4–0.6 g) was sealed in a clean polypropylene container and irradiated simultaneously with a uranium standard (100 ng U/g) for 90 s at the Jožef Stefan institute's TRIGA Mark II reactor (Ljubljana, Slovenia).

Immediately after irradiation, the irradiated sample was rapidly wet-ashed over a glass flame in a 100 mL long-necked silica Kjeldahl flask already containing 3 mL of 9 mol L^{-1} sulphuric acid (Sigma-Aldrich, Steinheim, Germany) and 50 mg of natural uranium (prepared from $(UO_2)(NO_3)_2.6H_2O$, Merck, Darmstadt, Germany) by heating with repeated additions of concentrated nitric acid (Sigma-Aldrich, Steinheim, Germany)until a pale yellow-green colour was obtained which did not darken on heating. The flask was then cooled by plunging into water, 1–2 mL of concentrated perchloric acid (Sigma-Aldrich, Steinheim, Germany) was added, and the flask reheated to evaporate the perchloric acid as dense white fumes. After dissolution the contents were transferred to a 50 mL separatory funnel with 20 mL of 5 mol L^{-1} nitric acid, split into two rinses. Uranium was extracted by vigorous shaking for 30 s with 50% tri-*n*-butylphosphate (TBP) (Sigma-Aldrich, Steinheim, Germany)in toluene (Sigma-Aldrich, Steinheim, Germany). The organic phase was briefly cleaned up with two washes of 5 mol L^{-1} nitric acid containing 0.2% hydrofluoric acid (Sigma-Aldrich, Steinheim, Germany). This washing helps to strip daughters decay products from natural uranium. The organic phase was drawn off by pipette, run into a measuring vial and measured directly in a Ge well-type detector (Byrne and Benedik, 1988; Repinc and Benedik, 2008). To avoid bias in the measurements, sample and standard were prepared in the same matrix and the measuring geometries.

2.1.2. Determination of ^{234}U, ^{238}U and ^{230}Th activity concentrations

For determination of thorium and uranium radioisotopes by alpha-particle spectrometry the samples (15–30 g) were ignited at 650 °C for 4 h and the remaining material was decomposed by lithium borates thermal fusion. The decomposed samples were loaded directly on the UTEVA® resin (Eichrom Technologies Inc., 2001) preconditioned in 3 mol L^{-1} HNO_3. The beakers were then washed twice with 3 mol L^{-1} HNO_3 (5 mL). After the sample was loaded the column was consecutively washed with 3 mol L^{-1} HNO_3 (20 mL) and 9 mol L^{-1} HCl (5 mL). In the next step thorium radioisotopes were stripped with 5 mol L^{-1} HCl with 0.5 mol L^{-1} oxalate (25 mL). In the final step the uranium radioisotopes were stripped with 1 mol L^{-1} HCl (15 mL). The sources for alpha-particle spectrometric measurement were prepared by microcoprecipitation with NdF_3 (Neodymium (III) Oxide) (Merck, Darmstadt, Germany) (Hindman, 1983; Sill and Williams, 1981). The neodymium fluoride suspension was filtered through a 0.1-μm polypropylene filter with a 25-mm diameter Resolve® filter (Eichrom Technologies, Lisle, IL, USA). The microcoprecipitate was dried under an infrared lamp, mounted on an aluminum disc, and measured on an alpha spectrometer.

2.1.3. Determination of ^{210}Po activity concentration

Determination of ^{210}Po was conducted according to the procedure described by Benedik and Vreček (2001). Each sample (5–6 g) to which Po-209 tracer was added was digested in a glass beaker at temperatures lower than 160 °C by mineral acids (HNO_3, $HClO_4$ and HF). The Po radioisotopes were deposited on a silver disc (Thessco B.V., Amsterdam-Zuidoost, Netherland). The measurements were conducted by an alpha spectrometer.

3. Results and discussion

In this study, 5 different types of infant formula for infants under 1 year of age from two of Slovenia's most popular producers of infant formulas were analyzed. It is worth mentioning that the analyzed infant formulas are widely available in pharmaceutical shops and supermarkets all over Slovenia. Samples 1–4 represent "normal" milk based infant formula with no modifications while sample 5 is a special formula that reduces the colonic fermentation which is responsible for bloating and excessive gas. Table 1 summarizes the details of the analyzed infant formulas and gives the date of ^{210}Po measurement. For the cumulative consumption calculation, the fact that infant diets should be supplemented with additional foods after the infant's 4th month was taken into account, as recommended by the producers. The cumulative consumption for each infant formula was calculated using the data available on the product's declaration.

To determine the activity concentrations of gamma-emitting radionuclides, the samples (approx. 30 g) were sealed in 100 mL plastic containers and after one month measured on a coaxial HPGE detector for 300 000 s. Inspection of the obtained spectra revealed that with the exception of ^{40}K and ^{210}Pb, no other gamma-emitting radionuclides had a high enough activity concentration to be accurately determined by direct gamma-ray spectrometry. Due to the low activity concentration

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