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Intake of ^{210}Po, ^{234}U and ^{238}U radionuclides with wine in Italy

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ABSTRACT

^{238}U, ^{234}U and ^{210}Po activity concentration was determined in 70 samples of red and white wine coming from 16 Italian regions. The radionuclides were determined by alpha spectrometry after separation. The results show that the mean concentrations of ^{238}U, ^{234}U and ^{210}Po were $3.63 \times 10^{-3} \pm 2.19 \times 10^{-3}$, $4.41 \times 10^{-3} \pm 3.10 \times 10^{-3}$ and $6.85 \times 10^{-2} \pm 3.79 \times 10^{-2}$ Bq L^{-1} for red wine and $5.08 \times 10^{-3} \pm 4.20 \times 10^{-3}$, $5.59 \times 10^{-3} \pm 8.49 \times 10^{-3}$ and $3.92 \times 10^{-2} \pm 2.63 \times 10^{-2}$ Bq L^{-1} for white wine, respectively. The effective radiation dose due to the uranium and polonium ingestions by wine (0.5 l per day) ranges from 2.68×10^{-4} to 4.91×10^{-2} mSv year^{-1}.

1. Introduction

Natural environmental radioactivity arises mainly from primordial radionuclides, such as ^{40}K, and the radionuclides from the ^{232}Th and ^{238}U series. The major contribution to the radiation exposure received by mankind comes from natural sources. These include external sources such as cosmic rays and radiation from primordial radionuclides (^{238}U and ^{232}Th) and their decay products in the environment. Information on the levels of naturally occurring radionuclides is important as they also contribute a substantial fraction of the radiation dose to the natural ecosystems (Holtzam, 1966).

Among natural radionuclides, alpha emitters are considered the most important with respect to potential internal radiation exposure as they are responsible for a significant proportion of the radiation exposure of humans, particularly through food consumption. ^{210}Po is estimated to contribute about 7% of the effective dose equivalent to man from ingested natural internal radiation (UNSCEAR, 1988). UNSCEAR (2000) quotes a worldwide average annual intake of 58 Bq of ^{210}Po in the diet. The value for annual ^{210}Po intake in the typical European diet is 40 Bq; however, this is based on data from only five countries (Italy, Poland, Romania, Russia and the UK) (UNSCEAR, 2000).

Wine is a widely consumed beverage in the world. Natural radioactivity has been measured in drinking and mineral water and soft drinks (Desideri et al., 2007a,b; Forte et al., 2007; Skwarzec et al., 2003, 2004; Pietrzak-Flis et al., 2001; Vesterbacka, 2007; Repinc and Benedik, 2002) but there are not data for wine. For this beverage, data are reported only on the artificial radioactivity (particularly ^{137}Cs) after Chernobyl (Renaud et al., 2003; Wagner Diehl, 1991).

The composition of wine is influenced by many factors related to the specific production as area, grape varieties, soil and climate and viticultural practices. There are several sources contributing to wine contamination, and it is known that radionuclides occur in wine from fruit contamination which can result from direct deposition on fruit surfaces, absorption by the fruit skin and transport to the pulp, deposition to soil, root uptake and transfer to fruit (Siveira et al., 2007).

The aim of the present study was to provide information on the levels of natural radionuclides ^{238}U, ^{234}U isotopes and ^{210}Po in samples of red and white wine product in the different Italian regions. The purpose of this work is to calculate the committed effective dose from ^{210}Po, ^{234}U and ^{238}U for the individual local public through wine ingestion.

2. Materials and methods

2.1. Samples

The samples of wine coming from 16 Italian regions (Fig. 1) were purchased from the traditional consumer markets. Red wine (36 samples) and white wine (34 samples) were analyzed.

2.2. Sample pretreatment

To determine ^{238}U and ^{210}Po by alpha spectrometry, it necessary to separate these radionuclides (by extraction chromatography, precipitation, electrodeposition etc.) from the matrix. For this aim, after addition of a known activity of ^{236}U or ^{209}Po as the yield internal standards, the sample of wine was first evaporated, dried and mineralised and the residue treated with conc. HNO_3 and H_2O_2 for three times. Finally the solution was evaporated to dryness and the residue was dissolved in 1 M HCl or in 2 M HNO_3 for polonium or uranium analysis, respectively and the solution was filtered (Meli et al., 2008).

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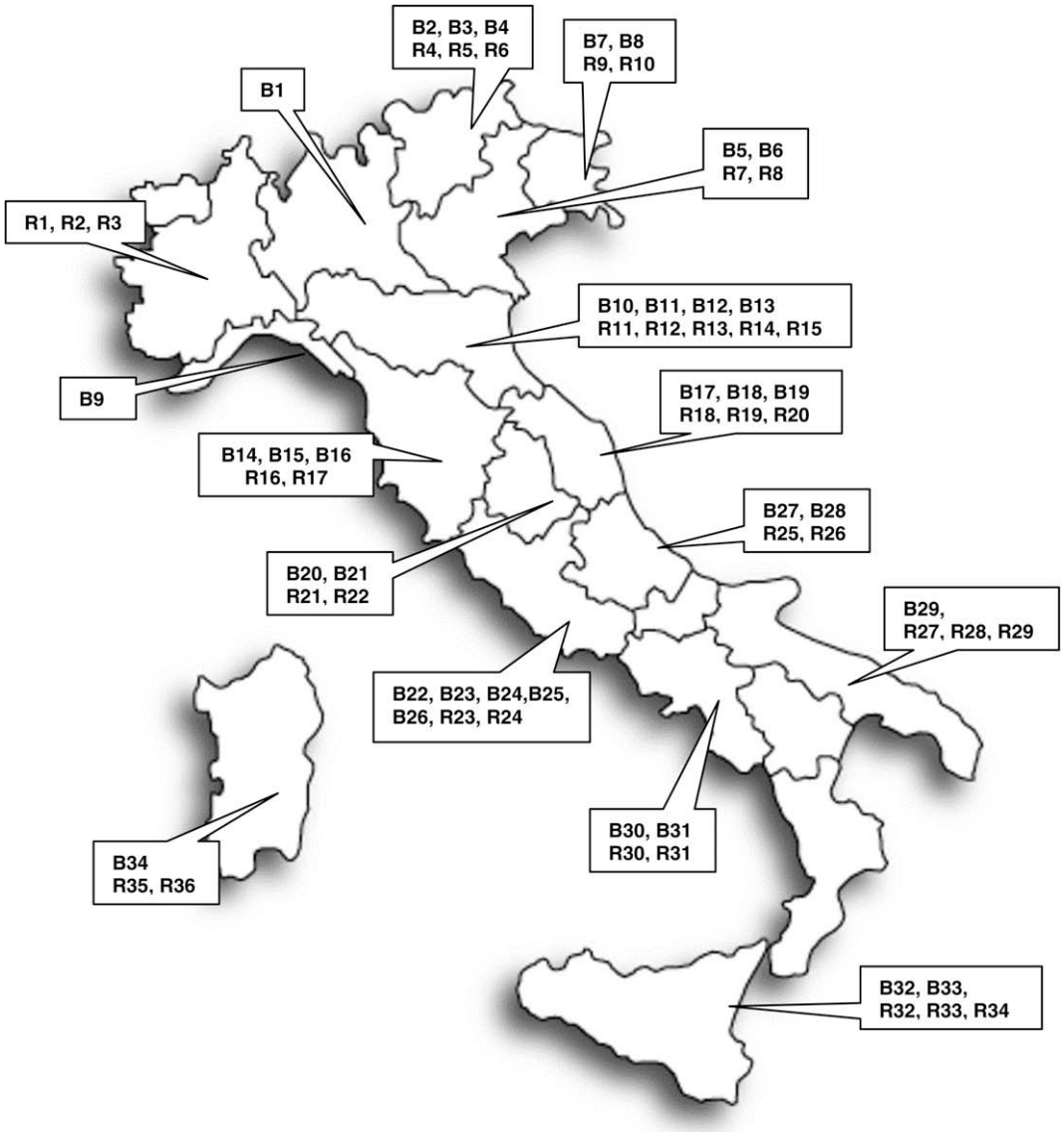

Fig. 1. White (B) and red (R) wines analyzed and their region of origin.

2.3. Radioanalytical methods

The radioanalytical methods used in this study are the following.

2.3.1. ^{238}Uranium

Radiochemical separation of uranium was performed by extraction chromatography. The nitric solution of uranium, coming from the sample pretreatment and containing ^{236}U as the yield internal standard, was passed through a column of microporous polyethylene supporting tri-n-octylphosphine oxide (TOPO) and conditioned with 2 M HNO_3. TOPO is a selective extractant in nitric medium for tetravalent and hexavalent actinides (Th, U, Pu, Np) and particularly for UO_2^{++}.

The alpha isotopes of natural thorium, particularly ^{230}Th (αEn = 4.68 MeV) which could interfere in the ^{234}U determination (αEn = 4.77 MeV), were washed out by 1 M HCl, then uranium was eluted by 1 M $(NH_4)_2CO_3$ which forms strong uranilcarbonate complexes (Desideri et al., 2007b).

First the elution solution was evaporated, dried and mineralised; then the residue was dissolved in conc. H_2SO_4 and transferred into an electrolytic cell. Uranium was electroplated from ammonium sulphate solution at pH 4; the measurements of ^{234}U, ^{235}U and ^{238}U were carried out using an alpha spectrometry system with silicon detectors (Canberra, USA) counting the source for 86,000s. The mean counting efficiency was 31.7 ± 3.1% and the background was approximately 2×10^{-6} s^{-1} in the energy region of interest. The mean chemical yield resulted 67.5 ± 7.4%.

2.3.2. ^{210}Polonium

^{210}Po cannot be determined by gamma spectrometry because it is not a gamma emitter, it emits only alpha particles at 5.407 MeV. It is not possible to use the gamma emission of other members of ^{238}U series because a secular equilibrium for all members of the series cannot be assumed.

Polonium was deposited, from the hydrochloric solution coming from the sample pretreatment, at 85–90 °C and pH 1.5–2.0 in continuous for 4 h in a silver disk, placed in a syringe and immersed into a 200 ml of 1 M HCl solution containing 10 ml of 20% hydroxylamine hydrochloride and 10 ml of 25% sodium citrate. The silver disk was measured by α-spectrometry. No preliminary separation was required and essentially quantitative recoveries (85 ± 8%) were obtained by using standard ^{209}Po tracer (Meli et al., 2008).

The uncertainties associated with the uranium and polonium activity concentrations are 15–25%; these values result from the propagation of all random counting uncertainties occurred anywhere in the entire measurement process.

2.4. Analytical quality control

Analytical quality control of these radioanalytical methods was carried out by different ways: (a) addition of ^{209}Po (En. Alpha = 4.976 MeV) and ^{236}U (En. Alpha = 4.572 MeV) as the yield tracers: this fact and the analysis of the relevant alpha spectrum permitted to obtain a precise figure for the recovery of any measure and (b) participation to a international intercomparison organized by Chemistry Unit of the International Atomic Energy Agency (IAEA) of Seibersdorf (Vienna) on the determination of ^{210}Po at different activity levels in five samples of acidified water.