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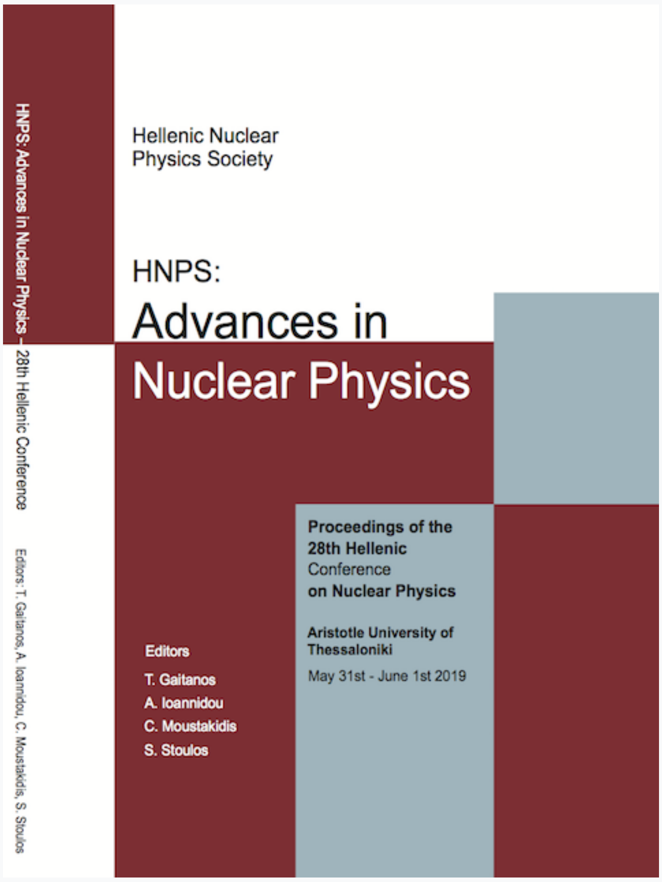

Natural radioactivity content in Italian bottled mineral waters

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Natural radioactivity content in Italian bottled mineral waters

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Abstract *Bottled water consumption has a long history but nowdays bottled water industry is a fast-growing sector of the world industry. Most of the water in bottles comes from springs or underground aquifers. The mineral content of the aquifer is diluted in small amounts into the water and although consuming mineral water is thought to be a healthy practice, there are several cases where radioactivity is also present. Monitoring the bottled water natural radioactivity is very important especially for the cases people consume mainly this kind of water. Radioactivity accumulation in the human body if exists in excess, can pose a threat for the health. In the present work, thirty brands of Italian bottled water were measured for gross alpha and beta radioactivity, uranium, radium and polonium radioisotopes. Radioactivity content was determined using the LSC method and sorbtion on polyamide pieces covered with thin film of MnO_2. The analysis using the MnO_2 thin films showed that the radium activity in waters varied from 4.7-69.3 $mBqL^{-1}$ and the polonium activity varied from 5.9-26.8 $mBqL^{-1}$. The measurements with the LSC method showed uranium concentrations varying from 0.7-93.1 $mBqL^{-1}$, while the radium activities exhibited variations from 1.6-34.1 $mBqL^{-1}$. Finally the gross beta activity values varied from 13.1-1584.9 $mBqL^{-1}$ and the gross alpha from 2.4-305.2 $mBqL^{-1}$.*

Keywords *LSC, uranium, radium, bottled water*

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INTRODUCTION

The bottled water industry is fast-growing worldwide. Worldwide consumption has been almost doubled the last decade (2007-2017) from about 210 to 390 billion liters [1]. With a total of ~440 billion liter consumption and ~$240 billion value of the global bottled water market, many issues arise concerning the energy needed for the production and transportation, the environmental footprint, the increase of the plastic pollution, the privatization and without any public control exploitation of the water resources etc. Although the consumption of bottled water is generally accepted as a healthy practice, in some cases the existence of natural radionuclides and thus of radioactivity in bottled water could pose a risk for the public health. especially in the case of increased consumption. The most common radioactive isotopes found in bottled waters are uranium, radium, polonium and in less extent thorium, due to its low solubility in water. These isotopes if exist in excess, may pose a threat for the public health and the monitoring for the radioactivity content is needed.

This work deals with gross alpha and beta measurements as well as uranium, radium and polonium radioactivity in 30 brands of bottled water produced in Italy. There are more than 240 brands and 140 bottling water factories through over Italy. This study aims to confront the lack of data for radioactivity in Italian bottled waters. Water samples were measured using the LSC method to measure the gross alpha and beta content as well as the activity concentration of uranium, radium and polonium isotopes. Also applying another method, in which the radium and polonium isotopes were first, sorbed onto small polyamide pieces of 2.3 cm × 2.3 cm^2 surface, coated with a thin film of MnO_2 and then measured by α-spectrometry. The details of the measurement methods were presented elsewhere [2, 3].

MATERIALS AND METHODS

Thirty (30) Italian bottled water brands (Fig. 1) were investigated for gross alpha and beta radioactivity as well as for uranium, radium and polonium radioisotopes. Five hundred mL of each sample, were evaporated into beakers and when the volume was below 20 mL, the remainder was transferred into polyethylene vials, suitable for LSC counting, of 20 mL capacity. After complete evaporation of the water samples, 4 mL of 2M H_3PO_4 and 16 mL of Ultima Gold AB scintillation cocktail were added and the vials were closed tightly. Afterwards, vials were placed into the LSC analyzer (Tricarb, 3170TR/SL), and were measured for alpha and beta radioactivity. The combined a-b spectrum was collected and analysed for α-emitters while beta background was also calculated (Fig. 2 and Fig. 3). Typical count time was 1000 min, and blank samples were used for background estimations. Beta background was calculated fitting blank spectra with double exponential decay fit and taking into account data from 120-2000 keV. Typical background was 0.141 ± 0.002 cps.

A quantity of 500 mL of each water sample were placed into a beaker, a pre-coated with MnO_2 thin film, small polyamide piece (23x23 mm^2), was immersed into the sample, pH was adjusted ~7 and radium and polonium isotopes were sorbed onto the polyamide pieces after 48 hours of stirring. Each of these pieces of MnO_2 coated polyamide was placed into an α-spectrometer (Canberra 7401VR) with vacuum chamber, equipped with a A900-29AM Canberra PIPS detector, working at 40 V, and standard electronic. Spectra were collected with an ORTEC acquisition card and Maestro software (Fig. 4).

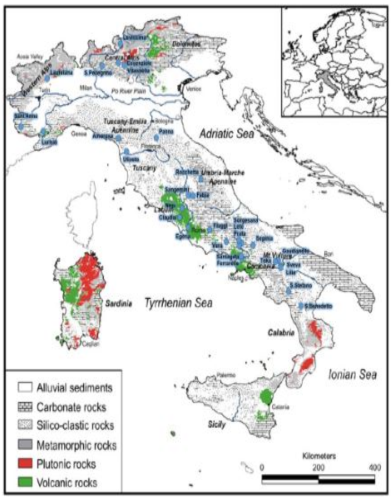

Figure 1. *Production locations of the Italian bottled water brands measured in the present study.*

RESULTS AND DISCUSSION

The results are presented in detail in Table 1. Gross beta and alpha radioactivity are presented in the 1st and 2nd columns of the Table. Concentrations varied from 13.1–1584.9 $mBqL^{-1}$ for gross beta radioactivity while for gross alpha values varied from 2.4–305.2 $mBqL^{-1}$. The concentrations of gross alpha and beta radioactivity were in the majority of the cases well below action levels which are 0.5 BqL^{-1} for gross alpha and 1 BqL^{-1} for gross beta radioactivity which would be sufficient to induce an annual dose of 0.1 mSv [4]. Only in five water brands the gross beta radioactivity exceeded 1 BqL^{-1} up to a concentration of ~1.6 Bq L^{-1}. Considering individual isotopes of uranium, concentrations of ^{238}U varied from 0.7 - 88.7 $mBqL^{-1}$ and for ^{234}U from 1.6 - 29.8 $mBqL^{-1}$.

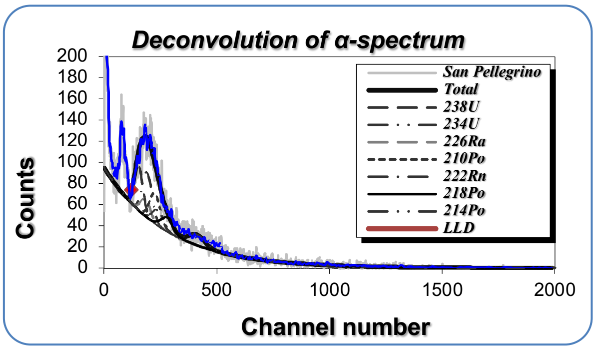

Figure 2. *Typical deconvolution of an α-spectrum of a water sample with uranium and radium isotopes.*

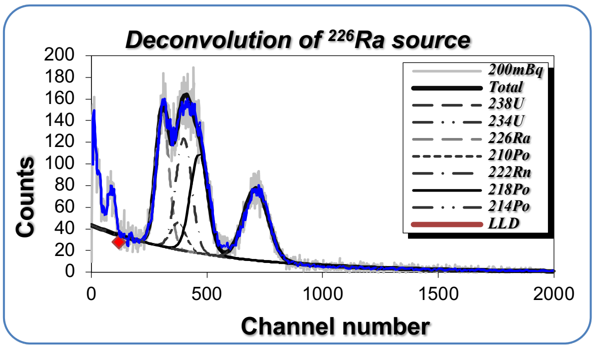

Figure 3. *Typical deconvolution of an α-spectrum with ^{226}Ra and decay daughter nucleus in equilibrium.*

Concentrations for ^{226}Ra were found to vary from 1.6–29.8 $mBqL^{-1}$ measured with the LSC method and from 4.7– 69.3 $mBqL^{-1}$ measured with the MnO_2 method. In the case of ^{224}Ra concentrations varied from 2.8– 31.1 $mBqL^{-1}$.

Finally, considering ^{210}Po isotope concentrations were found to vary from 1.0– 21.0 $mBqL^{-1}$ measured with the LSC method and from 2.8– 34.1 $mBqL^{-1}$ measured with MnO_2 method.

Prolonged consumption of bottled mineral water of elevated radioactivity, although does not exceed the action levels of the WHO guidelines for drinking water [4], should be reconsidered by the consumers due to the possibility of increasing the total absorbed dose. Thus the consumers should be well informed about the radiological aspects of the commercial water.

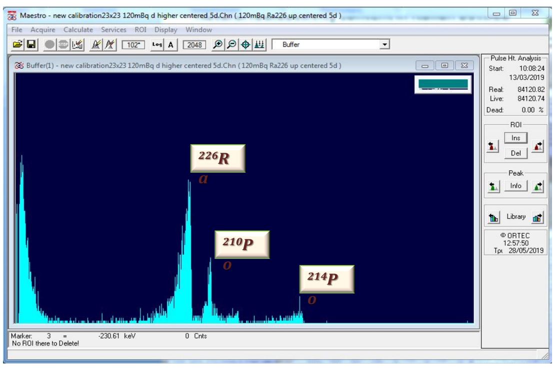

Figure 4. Typical α-spectrum of a polyamide piece coated with MnO_2 thin film. Radium and polonium isotopes are sorbed onto the coated polyamide, providing a very good resolution.

Table 1. Gross beta and alpha radioactivity measurements of Italian bottled waters are presented in the 1st and 2nd col. of the table. Uranium, radium and polonium isotopes radioactivity concentrations are presented in the 3rd-9th column. Labels LSC and MnO_2 refer to the method implied for measurements (see text). All concentrations are expressed in mBq L^{-1} with the associated error (1σ). ND = Not Detected, "-"= Not measured.

Water Brand	Gross beta (mBq L^{-1} ± 1σ)	Gross alpha (mBq L^{-1} ± 1σ)	^{238}U (mBq L^{-1} ± 1σ)	^{234}U (mBq L^{-1} ± 1σ)	^{226}Ra (mBq L^{-1} ± 1σ)		^{210}Po (mBq L^{-1} ± 1σ)		^{224}Ra (mBq L^{-1} ± 1σ)
	LSC	LSC	LSC	LSC	LSC	MnO_2	LSC	MnO_2	*LSC*
Amorosa	*27.1 ± 11.9*	*2.7 ± 7.4*	*2.7 ± 2*	*ND*	*ND*	*4.7 ± 2.7*	*ND*	*7.5 ± 3.1*	*ND*
Claudia	*1584.9 ± 19.6*	*121.7 ± 15.6*	*ND*	*ND*	*10.6 ± 4.1*	*11.4 ± 2.7*	*ND*	*18.4 ± 3.3*	*19.9 ± 4.1*
Egeria	*1398.8 ± 90.6*	*305.2 ± 34.3*	*30.1 ± 7.1*	*19.3 ± 8.6*	*29.8 ± 5*	*8.8 ± 2.7*	*ND*	*11.4 ± 3.2*	*34.1 ± 3.2*
Essentiale	*90.7 ± 14*	*18.6 ± 0.6*	*7.3 ± 0.7*	*11.3 ± 0*	*ND*	*5.9 ± 2.7*	*ND*	*7.7 ± 3.1*	*ND*
Fabia	*367.1 ± 24.1*	*191.5 ± 33.4*	*93.1 ± 7.2*	*52.7 ± 10*	*10.1 ± 6.4*	*-*	*5.2 ± 5.2*	*-*	*ND*
Ferrarelle	*788.8 ± 11.8*	*6.7 ± 0.3*	*5.9 ± 0.8*	*0.8 ± 1.1*	*ND*	*20.9 ± 2.8*	*ND*	*12.9 ± 3.2*	*ND*
Fiuggi	*368.9 ± 13.4*	*31.1 ± 9*	*7.9 ± 2.4*	*3.6 ± 2.4*	*1.7 ± 2.4*	*5.6 ± 2.7*	*ND*	*5.9 ± 3.1*	*3.2 ± 2.4*
Gaudianello	*1507.6 ± 16*	*12.7 ± 10.9*	*12.7 ± 3.1*	*ND*	*ND*	*-*	*ND*	*-*	*ND*
Lauretana	*91.7 ± 11.9*	*ND*	*0.6 ± 1.9*	*ND*	*ND*	*-*	*ND*	*-*	*ND*
Lete	*16.1 ± 9.1*	*12.7 ± 5.3*	*8.5 ± 0.1*	*4.2 ± 5.2*	*ND*	*6.4 ± 2.7*	*ND*	*7.9 ± 3.1*	*ND*
Levissima	*194 ± 103*	*85 ± 24*	*56.7 ± 11.2*	*27.3 ± 33.7*	*ND*	*13.9 ± 2.8*	*1 ± 1.5*	*11.9 ± 3.2*	*ND*
Lilia	*713.4 ± 16.2*	*74.4 ± 12.2*	*ND*	*ND*	*9.4 ± 3.2*	*-*	*ND*	*-*	*9.2 ± 3.2*
Lurisia	*151.1 ± 49.3*	*33.6 ± 13.8*	*7.1 ± 4*	*11.6 ± 1*	*2 ± 2.2*	*13.7 ± 2.8*	*6.9 ± 0.1*	*8 ± 3.1*	*ND*
Nepi	*1381.5 ± 19*	*119.7 ± 15*	*1 ± 3.9*	*ND*	*15.4 ± 4*	*9.8 ± 2.7*	*ND*	*20.4 ± 3.4*	*14.3 ± 4*
Panna	*81.2 ± 7.6*	*22 ± 3.1*	*12.3 ± 4.7*	*9.8 ± 1.6*	*ND*	*8.1 ± 2.8*	*ND*	*6.2 ± 3.2*	*ND*
Prata	*87.9 ± 4.2*	*18 ± 1*	*13.1 ± 0.2*	*4.9 ± 0.8*	*ND*	*-*	*ND*	*-*	*ND*
Rocchetta	*26.1 ± 21.1*	*10.3 ± 5.8*	*6.2 ± 2.4*	*4.1 ± 3.5*	*ND*	*6.7 ± 2.7*	*ND*	*6.4 ± 3.1*	*ND*
S. Anna	*13.1 ± 11.8*	*7.3 ± 7.2*	*6.2 ± 2*	*1.1 ± 1.9*	*ND*	*-*	*ND*	*-*	*ND*
San Agata	*770.3 ± 14.3*	*ND*	*ND*	*ND*	*ND*	*-*	*ND*	*-*	*ND*
San Benedetto	*129.1 ± 2.6*	*33.2 ± 7.1*	*16 ± 4*	*14.4 ± 7.3*	*ND*	*7.8 ± 2.7*	*2.7 ± 3.8*	*7.3 ± 3.2*	*ND*
San Pellegrino	*487.9 ± 165.2*	*239.5 ± 78.3*	*92.1 ± 27.6*	*88.7 ± 2.1*	*12.3 ± 8.9*	*69.3 ± 3.5*	*21 ± 0.4*	*26.8 ± 3.6*	*3.1 ± 4.4*
San Stefano	*112.1 ± 17*	*16.5 ± 1.8*	*9.7 ± 1.5*	*6.8 ± 0.3*	*ND*	*-*	*ND*	*-*	*ND*
Sangemini	*49.1 ± 14.6*	*6 ± 0.6*	*5.4 ± 0.1*	*0.7 ± 0.5*	*ND*	*11.2 ± 2.7*	*ND*	*9.2 ± 3.1*	*ND*
Sepinia	*31.5 ± 12*	*7.7 ± 7.5*	*7.5 ± 2.1*	*0.3 ± 2*	*ND*	*-*	*ND*	*-*	*ND*
Sorgesana	*99.2 ± 12.3*	*14.4 ± 7.8*	*7.9 ± 2.2*	*6.5 ± 2.2*	*ND*	*5.5 ± 2.7*	*ND*	*9.3 ± 3.1*	*ND*
Sveva	*978.6 ± 15.1*	*4 ± 10.5*	*4 ± 2.9*	*ND*	*ND*	*-*	*ND*	*-*	*ND*
Toka	*1299 ± 15.2*	*2.4 ± 10*	*2.4 ± 2.7*	*ND*	*ND*	*-*	*ND*	*-*	*ND*
Uliveto	*203.7 ± 8.7*	*11.8 ± 6.2*	*0.8 ± 1.1*	*ND*	*ND*	*16.6 ± 2.8*	*ND*	*11.4 ± 3.1*	*2.8 ± 1.3*
Vera	*156.8 ± 0.1*	*39.1 ± 9.8*	*16.8 ± 2.8*	*12.8 ± 6.4*	*1.6 ± 2.3*	*11.9 ± 2.8*	*3.1 ± 4.3*	*8.2 ± 3.1*	*ND*
Vitasnella	*253.4 ± 28.3*	*125.8 ± 1.5*	*56 ± 1.6*	*55.6 ± 1.8*	*ND*	*18.6 ± 2.9*	*14.2 ± 1.2*	*13.2 ± 3.4*	*ND*

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