



Radiological Investigation of a Portuguese Mineral Water

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Summary

The investigation of the Portuguese mineral water PEDRAS SALGADAS show clearly that the regular consumption of it results in a relatively high internal dose, for every age group. For parents we discourage from giving it to their babies or using it for the preparation of infant formulas. Even for children and adult persons the long lasting consumption of this mineral water will lead to increased internal doses. The real problem is that it no more exists a legal basis in Switzerland for the assessment of mineral waters concerning their radiological composition. Therefore, this mineral water remains on the Swiss market in spite of its high activities in radium (^{226}Ra and ^{228}Ra) and lead (^{210}Pb).

Introduction

Pedras Salgadas is a Portuguese mineral water from the springs of the same name origin from the Romans. It is exploited since 1871 commercially. It is carbonated naturally and shall not contain any pollutants...¹ The name *Pedras Salgadas* means "salty stones". In fact, it contains about 30 mg/L of chloride giving it a salty taste.

Even in 1996, Heinz Surbeck investigated this mineral water and found a surprisingly high activity of ^{226}Ra and its daughters ^{222}Rn , ^{218}Po and ^{214}Po . He could also detect ^{224}Ra and its daughters ^{220}Rn and ^{216}Po .² The limit value for natural radionuclides of group 2 (sum of ^{226}Ra , ^{228}Ra , ^{230}Th , ^{232}Th , ^{210}Pb , ^{210}Po and ^{231}Pa) of 1 Bq/L³ was overridden. Therefore, this mineral water was banned from the Swiss market. Since 2018, the mentioned *ordinance of foreign substances and ingredients* is no more valid. In consequence, there are no more regulations for mineral water concerning radionuclides.

¹ <http://www.finewaters.com/bottled-waters-of-the-world/portugal/pedras>

² Surbeck Heinz: alpha spectrometry sample preparation using selectively adsorbing thin films.

http://www.nucfilm.ch/nucfilm_discs.html

³ Ordinance of foreign substances and ingredients from 26.06.1995 (version 1.10.2015)

Legal basis

Since 2018, in Switzerland there exists no legal basis for radionuclides in mineral waters. However, since 2017, a new Ordinance for drinking water was put into force, the *Ordinance on Drinking Water and Water from Public Baths and Shower Facilities (TBDV)*⁴. It includes a limit value for uranium and guide values for tritium, radon and the parameter *indicative dose*. This ordinance is based on the *European Council Directive 2013/51/EURATOM*⁵. Fortunately, this council directive prescribes some more guidance limits for artificial and natural radionuclides, called “derived activity limits”. They are also applicable in Switzerland.

These *derived activity limits* (abgeleitete Konzentrationen, AK-values) are based on a yearly consumption of 730 Litres of drinking water resulting in an annual dose by ingestion less than 100 μSv .

$$\text{AK [Bq/L]} = 100 [\mu\text{Sv}] / (730 [\text{kg}] * e_{\text{ing}} [\text{Sv/kg}]) \quad (1)$$

With the conversion factor for ingestion, e_{ing} for adult persons taken from⁶.

For the calculation of individual doses for infants (1-2 years), children (< 17 years) and adults consumption of drinking water was taken from⁷. For infants a consumption of yearly 170 L of drinking water, included the water for the preparation of infant formula, children of 200L and adult persons 730 L.

Materials and Methods

The Pedras Salgadas mineral water was collected on the Swiss market.

Gamma spectrometry

The mineral water was filled into a 1 L Marinelli-beaker and counted on a high-resolution germanium detector. All emission lines and emission probabilities were taken from LNHB⁸.

Alpha and beta spectrometry

Uranium, radium, radon and polonium species were investigated with specific alpha spectrometric methods. ²¹⁰Pb was analysed by means of β -spectrometry.

Uranium (²³⁴U and ²³⁸U)

The U-species, acidified with 10 mL of conc. sulfuric acid. 1.5 mL of the extract was counted with a PERALS- alpha spectrometric counter during 24 hours. ²³²U was used as tracer.

Radium (²²⁴Ra and ²²⁶Ra)

The nuclides ²²⁴Ra and ²²⁶Ra were extracted from 200mL of water onto a MnO₂-surface.

⁴ Department of Inner Affairs: Ordinance on drinking water and water for public baths and shower facilities (TBDV); 2016. Status: May 1, 2018

⁵ Council Directive 2013/51/EURATOM of October 2nd, 2013 laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption

⁶ Swiss Federal Council: Radiological protection ordinance (RPO); April 26, 2017. Status: February 1, 2019

⁷ Fachbereich Strahlenschutz und Umwelt des BfS: Strahlenexposition durch natürliche Radionuklide im Trinkwasser in der Bundesrepublik Deutschland.2009.

⁸ LNHB: Laboratoire National Henri Becquerel. Available from: http://www.nucleide.org/DDEP_WG/DDEPdata.htm [Accessed: November 07, 2019]

The loaded disk was counted with a silicon barrier detector for 24 hours. The following a-lines were used for identification and quantification: ^{226}Ra : 4.6-4.8 MeV (100%), ^{224}Ra : 5.7 MeV (95%).

Polonium (^{209}Po und ^{210}Po)

These nuclides were extracted onto a silver disc (auto-deposition) from 100 mL of water (3 hours at 80°C). The loaded silver-disk was counted with a silicon barrier detector for 24 hours. ^{209}Po was used as a tracer. The following alpha emission lines were used for identification and quantification: ^{209}Po : 5.2 MeV, ^{210}Po : 5.4 MeV.

Lead-210 (^{210}Pb)

This radionuclide was analysed via its daughter ^{210}Bi , which is in secular equilibrium (after 35 days). The beta-species were extracted from 200 mL of water onto a steel disk (18 hours at 60 °C). After a short cooling time (other beta emitters, such as ^{214}Pb and ^{214}Bi) the beta was counted with a a/b-gas proportional counter (LB 4100 from Canberra) for one hour.

Radon (^{222}Rn)

For the analysis of ^{222}Rn , 10 mL of water were mixed with 10 mL of MaxiLight cocktail. After an equilibrium time of three hours, radon and its two polonium daughters were counted with means of a α/β liquid scintillation counter (Hidex 300SL).

Results

In Swiss legislation there are no limit values defined for mineral water. Therefore, there is no legal assessment possible for the Pedras Salgadas mineral water based on the present investigations. When considering the mineral water like drinking water, there are exceedances of the AK-values for ^{226}Ra , ^{228}Ra and ^{210}Pb (table 1).

Radionuclide	Activity \pm SD	AK-value	remark
^{223}Ra	100 \pm 80	1,400*	
^{224}Ra	110 \pm 25	2,100*	
^{226}Ra	1,360 \pm 350	500	exceedance
^{226}Ra via ^{214}Pb , ^{214}Bi	1,200 \pm 65	500	
^{228}Ra via ^{228}Ac	1,600 \pm 145	200	exceedance
^{234}U	12 \pm 4	2,800	
^{238}U	23 \pm 7	3,000	
^{222}Rn	2,800 \pm 700	100,000	
^{210}Po	48 \pm 10	100	
^{210}Pb	270 \pm 30	200	exceedance
^{227}Th	200 \pm 80	15,600*	
^{40}K	1,200 \pm 500	22,000*	
^{134}Cs	<10	7,200	
^{137}Cs	<10	11,000	

Table 1: Results of the radiological investigation of Pedras Salgadas. All values in mBq/L. *calculated AK-values according to EURATOM⁴ and equation (1).

Internal dose estimation

The indicative dose gives information on the dose intake by consumption of this mineral water and is a criterion for the decision if a drinking water has to be monitored extensively. It is not to be mixed with the term "total internal dose".

In 2013/51/EURATOM and TBDV the indicative dose is defined as the total dose, excluding the doses from tritium, ^{40}K and ^{222}Rn and its daughters. The indicative dose is almost equal to a total ingestion dose as the contributions of ^{40}K and ^{222}Rn are negligible.

Radionuclide	Activity mBq/L	Dose coefficients for ingestion				Annual Doses (m Sv/a)			
		Infant 0 – 1 a	Infant 1 – 2 a	Child 12 – 17 a	Adult >17 a	Infant 0 – 1 a	Infant 1 – 2a	Child 12 – 17 a	Adult > 17 a
^{223}Ra	100	5.3*	1.1*	0.37*	0.10*	0.002	0.002	0.002	0.01
^{224}Ra	110	2.7	0.66	0.20	0.07	0.05	0.012	0.006	0.01
^{226}Ra	1,360	4.7	0.96	1.5	0.28	1.1	0.22	0.22	0.28
^{228}Ra	1,600	30	5.7	5.3	0.69	8.2	1.55	0.54	0.81
^{234}U	12	0.37	0.13	0.074	0.049	0.001	0.0003	0.001	0.001
^{238}U	23	0.34	0.12	0.067	0.045	0.001	0.0005	0.001	0.001
^{210}Po	48	26	8.8	1.6	1.2	0.21	0.072	0.025	0.04
^{210}Pb	270	8.4	3.6	1.9	0.69	0.39	0.165	0.103	0.14
^{227}Th	200	0.03*	0.07*	0.015*	0.009*	0.002	0.002	0.001	0.001
Indicative Dose						9.9	2.0	0.9	1.3
^{40}K	1,200	0.068	0.042	0.0076	0.0062	0.01	0.04	0.008	0.006
^{222}Rn	2,800	0.04	0.004	0.004	0.004	0.01	0.002	0.04	0.02
Total dose by ingestion						9.9	2.0	0.9	1.3

Table 2 individual ingested, yearly doses by consumption of Pedras mineral water. All dose coefficients from 6. * From⁹. The year's consumption of drinking water for adult persons was set to 730 L per year.

The indicative dose in Swiss legislation for drinking water (0.1 mSv/a) is clearly overridden for every age group. Main Contributions come from the radionuclides ^{226}Ra , ^{228}Ra and ^{210}Pb . For babies of 0 - 1 year, the ingestion-dose reaches the 100fold of the indicative dose. Therefore, it is recommended not

⁹ Bundesamt für Sicherheit der nuklearen Entsorgung: Dosiskoeffizienten bei äusserer und innerer Strahlenexposition.

<https://www.base.bund.de/DE/base/gesetze-regelungen/dosiskoeffizienten/dosiskoeffizienten.html>

to let babies drink Pedras Mineral Water or to use it for the preparation of infant formulas. Fortunately, it seems unlikely that Pedras mineral water is given to babies. However, even children and adult persons get relatively high ingestion-doses when this mineral water is regularly consumed.

Conclusions

The investigation of the Portuguese mineral water PEDRAS SALGADAS show clearly that the regular consumption of it results in a relatively high internal dose, for every age group. For parents we discourage from giving it to their babies or using it for the preparation of infant formulas. Even for children and adult persons the long lasting consumption of this mineral water will lead to increased internal doses.