Natural radioactivity in mineral and spa waters: 
the current regulatory approach in Italy

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Abstract. Mineral and thermal waters can contain radioactivity of natural origin which, in some cases, can lead to radiation-protection problems for both workers and consumers. In Italy, as in many other countries, the consumption of bottled mineral water is rather high and the practice of spending short stays in spas is also popular. Consumer protection against natural radioactivity in mineral water is not regulated at all and exposure from thermal waters is not treated in detail, in either the European Union or Italy. For this reason, the Italian Ministry of Health - which has the duty to authorize spa activities and the sale of mineral water on the basis of water characteristics – asked the Istituto Superiore di Sanità (the Italian National Institute of Health) to provide ad-hoc reports containing both dosimetric calculations and analysis of the radiation-protection regulations to be applied in these situations. On the basis of these reports, the Ministry of Health decided, in two statements for spa and mineral waters respectively, to indicate upper levels for natural radioactivity content. Since 2002, spa water parameters have been in force for authorization of their use and sale, and are to be promulgated by specific and more comprehensive regulations. For mineral waters used for infant feeding and drinking, levels lower than those for general public uses have been proposed, in order to take into account the higher ingestion dose coefficients for infants. At the moment, mineral water levels are not enforced as law. This paper presents the dosimetric calculation results and the Ministry of Health statements.

1. Introduction

In Italy, as in many other countries, the consumption of bottled mineral water is high and continually increasing. The habit of spending short periods (1–2 weeks per year) in spas, for holidays or medical/cosmetic treatment, is also widespread. Both bottled mineral water and spa waters may contain significant amounts of natural radionuclides [1,2], so that, in some circumstances, a radiation-protection issue can arise for workers and/or the population. In the EU, the protection of workers from natural radioactivity is regulated by European Directive 96/29/Euratom [3], which was implemented into the Italian legislation in 2000 [4]. On the other hand, consumer protection against natural radioactivity in mineral water is not regulated at all and exposure from thermal waters is not treated in detail, either in the European Union or in Italy. For this reason, the Italian Ministry of Health - which has the duty to authorize spa activities and mineral water sales on the basis of water characteristics - asked the Consiglio Superiore di Sanità (CSS-National Health Council) to study these issues, in particular radon and its decay products in spa water and the natural radioactivity in mineral water. Consequently, the Istituto Superiore di Sanità (the Italian National Institute of Health) was requested by the CSS to carry out both dosimetric calculations and analyses of the radiation-protection regulations to be applied to these situations; two separate reports were produced concerning spa and mineral waters. On the basis of these reports, the CSS provided two distinct opinions, as requested by the Ministry of Health. It is worthy of note that since 2002 the Ministry of Health has based spa water authorizations on levels included in the relevant CSS opinion. This paper presents the results of dosimetric calculations and radiation-protection evaluations included in the Istituto Superiore di Sanità reports for spa water and mineral water. Moreover, summaries of CSS opinions are also presented.

2. Radioactivity in spa waters

2.1. Summary of the Istituto Superiore di Sanità report

The problem as regards spa waters is the radon and its decay product content. The radiation-protection problems arising from the use of radon-rich spa waters can be complex requiring careful analysis of exposure situations. Specific Italian or European regulations are missing. Therefore, the existing regulations concerning similar issues, in particular drinking waters, were adapted, taking into account
the general principles of radiation protection. The approach adopted was aimed at limiting and, when necessary, reducing the effective doses to users of spa waters. Typical exposures were considered and relevant doses evaluated with some conservative assumptions. The availability of simple and effective methods for removing radon from water [5,6] made limiting and reducing doses a feasible goal.

2.1.1. Dosimetric calculations for the different uses of spa waters

The following uses of radioactive spa waters were analyzed: 1) ingestion of bottled spa waters outside spas; 2) ingestion of spa waters in spas; 3) aerosol treatment; 4) thermal-bath treatment; 5) mud-bath treatment. The following natural radionuclides were considered: $^{222}\text{Rn}$ and, only for ingestion, its two long-life decay products $^{210}\text{Po}$ and $^{210}\text{Pb}$. For these last two radionuclides the committed effective dose per unit intake via ingestion of the European (and Italian) Basic Safety Standards were utilized. Since there are no $^{222}\text{Rn}$ coefficients in the European (and Italian) Basic Safety Standards, the values reported in the NRC report [7] were considered. In calculating the effective dose to the general population, the following concentrations of $^{222}\text{Rn}$ in water were assumed: 100 Bq·l$^{-1}$ for drinking of bottled spa waters and 500 Bq·l$^{-1}$ for all other uses.

2.1.1.1. Ingestion of bottled spa waters

Two consumption patterns were considered:

A) water consumption/ingestion on the same day it was bottled (this scenario is extremely rare, and was considered only to evaluate the highest possible dose);

B) water consumption/ingestion within two years after bottling, as recommended by Italian regulations on bottled water. In this scenario, the main contribution to the effective dose comes from the $^{210}\text{Pb}$ and $^{210}\text{Po}$ produced only by the decay of the $^{222}\text{Rn}$ contained in the water when bottled (see assumption n. 3)

Assumptions:
1) ingestion of 2 liters of bottled spa water per day, 365 days per year;
2) $^{222}\text{Rn}$ activity concentration of 100 Bq·l$^{-1}$ in the water when bottled;
3) $^{210}\text{Po}$ and $^{210}\text{Pb}$ not present in the water when bottled;
4) dose coefficient for ingestion of $^{222}\text{Rn}$: 3.5 $\times$ 10$^{-9}$ Sv·Bq$^{-1}$ (average value for adults from NRC [7]);
5) dose coefficient for ingestion of $^{210}\text{Pb}$ and $^{210}\text{Po}$: 6.9 $\times$ 10$^{-7}$ Sv·Bq$^{-1}$ and 1.2 $\times$ 10$^{-6}$ Sv·Bq$^{-1}$, respectively (average values for adults from European BSS [3]).

Results:
Ingestion pattern A: the effective dose is 0.256 mSv·year$^{-1}$, due to $^{222}\text{Rn}$ only.

Ingestion pattern B: the effective dose due to $^{210}\text{Pb}$ is 0.024 mSv·year$^{-1}$ (maximum value) and 0.023 mSv·year$^{-1}$ (average value for a constant rate in the two considered years). The $^{210}\text{Pb}$ activity concentration reaches its maximum value (0.047 Bq·l$^{-1}$) after about 40 days and then remains nearly constant for the following two years, with an average in this period of 0.045 Bq·l$^{-1}$. The effective dose due to $^{210}\text{Po}$ is 0.038 mSv·year$^{-1}$ (maximum value) and 0.029 mSv·year$^{-1}$ (average value for a constant rate in the two considered years). The $^{210}\text{Po}$ activity concentration reaches its maximum value (0.044 Bq·l$^{-1}$) after about 2 years, with an average in this period of 0.033 Bq·l$^{-1}$. The effective dose due to $^{222}\text{Rn}$ decreases rapidly and becomes negligible after about 20 days.

2.1.1.2. Ingestion of spa waters in spas

For this use, the consumption (ingestion) of spa water was assumed to be restricted to a limited period of time (typically two stays per year at spas), without any delay that could reduce the $^{222}\text{Rn}$ concentration due to its decay. In this scenario, the effective dose is due to the ingestion of $^{222}\text{Rn}$ only, whereas the contribution of its long-life decay products $^{210}\text{Pb}$ and $^{210}\text{Po}$ is negligible.
Assumptions:
1) ingestion of 2 liters of spa water per day, for 24 days per year (2 stays per year, 12 days per stay);
2) $^{222}$Rn activity concentration of 500 Bq·l$^{-1}$ in the water during ingestion;
3) $^{210}$Po and $^{210}$Pb not present in the water during ingestion;
4) dose coefficient for ingestion of $^{222}$Rn: $3.5 \times 10^{-9}$ Sv·Bq$^{-1}$ (average value for adults from NRC [7]).

Result: the effective dose is 0.085 mSv·year$^{-1}$, due to $^{222}$Rn only.

2.1.1.3. Aerosol treatment

For this use of spa waters, all the $^{222}$Rn contained in the water used to produce aerosols (typically 1 liter per treatment) was assumed, for protection purposes, to be inhaled. The most recent dose coefficient, for the inhalation of $^{222}$Rn and its decay products, evaluated by UNSCEAR [8] was applied. Another dose coefficient widely used – evaluated by ICRP in the ICRP-65 publication [9] and adopted in the European Directive 96/29/Euratom – is about 2/3 of the UNSCEAR dose coefficient value. Both dose coefficients were evaluated for exposure conditions - such as aerosol concentration, aerosol size distribution and the equilibrium factor between radon and its decay products - representative of the indoor air of dwellings or similar indoor environments, yet different from those in aerosol treatment for which no specific dose coefficient was available. No extreme differences were expected on the basis of available dosimetric calculations [10,11].

Assumptions:
1) consumption (through inhalation) of 1 liter of water for each treatment, 12 treatments per stay, 2 stays per year;
2) $^{222}$Rn activity concentration of 500 Bq·l$^{-1}$ in the water;
3) dose coefficient for inhalation of $^{222}$Rn: $3.0 \times 10^{-6}$ mSv·Bq$^{-1}$ of inhaled $^{222}$Rn. This value was derived from the dose coefficient of 0.025 mSv per Bq·m$^{-3}$ of $^{222}$Rn in air, which refers to an exposure in normal indoor environments of 7000 hours·year$^{-1}$ [7], with a breathing rate of 1.2 m$^3$·h$^{-1}$;
4) all the $^{222}$Rn contained in the water used for the treatment is inhaled (conservative hypothesis).

Result: the effective dose is 0.035 mSv·year$^{-1}$.

2.1.1.4. Thermal-bath treatment

In the thermal-bath treatment, both the effective dose due to external radiation coming from decay products of radon present in water and the effective dose due to direct radon inhalation in water were calculated. In order to assess the dose due to external radiation for immersion in water containing $^{222}$Rn, a model was used in which the absorbed dose rate in air (expressed in Gy) is estimated in a hole in the middle of a medium (in this case water). The effective dose is obtained from the resulting value by means of a conversion coefficient equal to 0.7 Sv·Gy$^{-1}$ [8], which represents a good approximation in this case. In order to estimate the dose due to inhalation of radon emanated from the water in a swimming pool, in the absence of experimental data, the radon concentration in the air immediately overhanging the water had to be estimated. To account for the large area of the emitting surface and the high temperature of the water in a thermal swimming pool, the air-water transfer factor was increased by an order of magnitude with respect to the air-water transfer factor of $10^{-4}$ estimated by NRC for usual water uses in dwellings [7].

Assumptions:
1) pool size: 50 m x 25 m x 150 cm;
2) 20 min for each treatment, 12 treatments per stay, 2 stays per year;
3) $^{222}$Rn activity concentration of 500 Bq·l$^{-1}$ in the water;
4) a transfer factor of $10^{-3}$ between the $^{222}$Rn activity concentration in the water and the $^{222}$Rn activity concentration in the air at 20–30 cm from the surface of the water;
5) breathing rate of 1.2 m$^3$·h$^{-1}$.

Results: the effective dose is 0.017 mSv·year$^{-1}$ (0.014 mSv·year$^{-1}$ due to inhalation of $^{222}$Rn and its decay products and 0.0025 mSv·year$^{-1}$ due to external gamma irradiation).
2.1.1.5. Mud-bath treatment

The dose due to external gamma irradiation was calculated in the same way as for the immersion in water, taking into account in this case the different values for the size of the pool, as well as the density and composition of mud compared to water. The inhalation of the $^{222}\text{Rn}$ given off by the mud was excluded because it was assumed that the transfer from mud to air is at least 10 times lower than from water to air in thermal-swimming pools.

Assumptions:
1) pool size: 1.5 m x 1.5 m x 3 m;
2) 20 min for each treatment, 12 treatments per stay, 2 stays per year;
3) $^{222}\text{Rn}$ activity concentration of 500 Bq l$^{-1}$ in the water used to produce the mud;
4) no $^{222}\text{Rn}$ activity concentration in the clay used to produce the mud;
5) mud composition: 50% (in weight) of water and 50% (in weight) of clay (the decay of $^{222}\text{Rn}$ in the water before its use for the production of mud was conservatively neglected).

Result: the effective dose is 0.002 mSv·year$^{-1}$ (in the middle of the pool, due to external gamma irradiation).

2.1.2. Discussion on the calculated effective doses and proposed upper values for activity concentrations of $^{222}\text{Rn}$, $^{210}\text{Pb}$ and $^{210}\text{Po}$ in spa waters

2.1.2.1. Ingestion of bottled spa waters outside spas

The European Recommendation [12] proposes an action level in the range of 100–1000 Bq l$^{-1}$ for $^{222}\text{Rn}$ activity concentration in drinking water. The corresponding effective doses would be 0.26–2.56 mSv·year$^{-1}$. Such rather high doses are “accepted” in this recommendation considering the primary value of drinking water. The situation is different for bottled water because many different bottled waters are available with very low or negligible radioactive content. The above dose values could be applied to bottled spa waters in the non-realistic case of consumption on the same day as bottling. For consumption within two years after bottling, the $^{222}\text{Rn}$ activity concentration decreases by an average factor of 100. In this period, the maximum and average effective dose are 0.062 mSv·year$^{-1}$ and 0.052 mSv·year$^{-1}$, respectively, due to the $^{210}\text{Pb}$ and $^{210}\text{Po}$ produced by the 100 Bq l$^{-1}$ of $^{222}\text{Rn}$ initially present in the water. In this two-year period, the activity concentrations of $^{210}\text{Pb}$ and $^{210}\text{Po}$ are always below 0.2 Bq l$^{-1}$ and 0.1 Bq l$^{-1}$, respectively, the upper values proposed by the relevant European Recommendation [12].

On the basis of the above considerations and due to the present lack of specific Italian or European regulations, upper levels of 100 Bq l$^{-1}$, 0.2 Bq l$^{-1}$ and 0.1 Bq l$^{-1}$ are here proposed for $^{222}\text{Rn}$, $^{210}\text{Pb}$ and $^{210}\text{Po}$ activity concentration, respectively, in bottled spa waters used for drinking outside spas. However, effective methods to remove $^{222}\text{Rn}$ are available, so it is recommended that such methods be applied to reduce $^{222}\text{Rn}$ concentration well below the proposed upper value.

2.1.2.2. Other uses of waters in spas

The total effective dose for the uses of spa waters calculated in sections 2.1.1.2 to 2.1.1.5 is 0.140 mSv·year$^{-1}$, if the $^{222}\text{Rn}$ activity concentration in water is 500 Bq l$^{-1}$, which is within the range of 100-1000 proposed by the European Recommendation [12]. The main contribution comes from ingestion (0.085 mSv·year$^{-1}$), which is below the value of 0.1 mSv·year$^{-1}$ of effective dose reported in the European Directive 98/83/EC [9] as a parameter for the evaluation of drinking water, although that value does not include the doses due to $^4\text{H}$, $^{40}\text{K}$, $^{222}\text{Rn}$ and its decay products. The indicative dose of 0.1 mSv·year$^{-1}$ was used in the European Recommendation [12] to derive the proposed upper levels of 0.2 Bq l$^{-1}$ and 0.1 Bq l$^{-1}$ for the activity concentration of $^{210}\text{Pb}$ and $^{210}\text{Po}$, respectively.

On the basis of the above considerations, a upper level of 500 Bq l$^{-1}$ is here proposed for $^{222}\text{Rn}$ activity concentration in spa waters used in spas. Effective methods to remove $^{222}\text{Rn}$ are however available, so
it is recommended that such methods be applied to reduce $^{222}$Rn concentration well below the proposed upper value, especially if the water is used for ingestion.

2.1.3. Comments on the advertisement of some spas

The advertisements of some spas claim that some positive health effects are related to the radioactive component of spa water, in particular radon. In the framework of the present analysis of radiation-protection problems of spa waters, this situation cannot be considered correct for the following reasons. No national or international health authority has so far recognized any positive health effect of radon and of any radioactive component of spa waters. Moreover, on the basis of European Directive 97/43 [14], patients are not to be exposed to ionizing radiations without a justification process concluding that there are positive effects of such exposure and that they are greater than the negative effects or risks. Therefore, this situation is here highlighted for a possible intervention.

2.2. Summary of the Consiglio Superiore di Sanità opinion

The Consiglio Superiore di Sanità, on the basis of the report prepared by the Istituto Superiore di Sanità, proposed the following actions to the Minister of Health, until such time as specific and more comprehensive Italian or European regulations are issued:
- to introduce, for bottled spa water, the following upper values for activity concentration: $100 \text{ Bq}\cdot\text{l}^{-1}$, $0.2 \text{ Bq}\cdot\text{l}^{-1}$, and $0.1 \text{ Bq}\cdot\text{l}^{-1}$ for $^{222}$Rn, $^{210}$Pb, and $^{210}$Po, respectively;
- to introduce, for all other uses of spa waters in spas, an upper value of $500 \text{ Bq}\cdot\text{l}^{-1}$ for the activity concentration of $^{222}$Rn;
- to recommend adequate procedures and methods for sampling water and measuring radioactivity;
- to recommend the use of the simple and effective methods available for removing $^{222}$Rn from water, in order to reduce $^{222}$Rn well below the above upper values;
- to recommend that the competent authorities verify the advertisements of spas and, if declarations on the positive health effects of radon are present, to take appropriate action.

3. Radioactivity in mineral waters

3.1. Summary of the Istituto Superiore di Sanità report

Using the same approach as for spa waters, regulations applying to similar topics, i.e. drinking waters, were adapted to mineral waters, with regard for the general principles and rules of radiation protection. The use of drinking water regulations for mineral water seems to be justified by the high and increasing consumption of bottled mineral water. The reported evaluations concerning radiation protection and dosimetry were also applied to bottled non-mineral water, in view of its increasing use and because it is included in the general definition of drinking water in the pertinent European and Italian regulations [12,13,15]. Therefore, in the next sections the term "mineral water" will generally mean bottled water, mineral and non.

3.1.1. Dose calculations for mineral water

The next sections will deal with natural radionuclides potentially present in mineral water and the highest doses associated [8], in particular: 1) natural uranium isotopes; 2) $^{228}$Ra and $^{226}$Ra; 3) $^{222}$Rn; 4) $^{210}$Po and $^{210}$Pb.

The effective dose calculations reported in this section were generally carried out for radionuclides activity concentrations of $1 \text{ Bq}\cdot\text{l}^{-1}$, but $100 \text{ Bq}\cdot\text{l}^{-1}$ was used for $^{222}$Rn. Moreover, all age groups present in the Italian Basic Safety Standards were considered [4], unlike what was done for spa waters, for which only adult consumers were assumed. For all radionuclides except $^{222}$Rn, the committed effective dose values per unit intake via ingestion were taken from the European (and Italian) Basic Safety Standards. As for spa waters, the $^{222}$Rn coefficients reported in the NRC report [7] were considered. All calculations were carried out assuming an annual intake of mineral water of $250\text{l}\cdot\text{y}^{-1}$ for age $\leq 1$
year, 350 l\cdot y^{-1} for ages between 1–10 years, 750 l\cdot y^{-1} for age >17 years, intake values chosen by the European Group of Expert (art. 31 Euratom Treaty) [16]. For the age group 10–12 years the same consumption as the 1–10 year group was assumed; for the 12–17 year group, the interpolated value of 550 l\cdot y^{-1} was used.

3.1.1.1. Natural Uranium

In case of ingestion the committed effective dose per 1 Bq\cdot l^{-1} of natural uranium activity concentration in water is shown in the following Table I:

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>\leq 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>&gt;17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient (10^{-8} Sv\cdot Bq^{-1})</td>
<td>35.5</td>
<td>12.5</td>
<td>8.4</td>
<td>7.1</td>
<td>7.0</td>
<td>4.7</td>
</tr>
<tr>
<td>Effective dose (mSv\cdot y^{-1})</td>
<td>0.088</td>
<td>0.044</td>
<td>0.030</td>
<td>0.025</td>
<td>0.039</td>
<td>0.035</td>
</tr>
</tbody>
</table>

3.1.1.2. $^{228}$Ra and $^{226}$Ra

The most abundant radium isotopes are $^{228}$Ra and $^{226}$Ra. For $^{228}$Ra, the committed effective dose per 1 Bq\cdot l^{-1} of activity concentration in water is:

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>\leq 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>&gt;17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient (10^{-7} Sv\cdot Bq^{-1})</td>
<td>300</td>
<td>57</td>
<td>34</td>
<td>39</td>
<td>53</td>
<td>6.9</td>
</tr>
<tr>
<td>Effective dose (mSv\cdot y^{-1})</td>
<td>7.5</td>
<td>2.0</td>
<td>1.2</td>
<td>1.4</td>
<td>2.9</td>
<td>0.5</td>
</tr>
</tbody>
</table>

For $^{226}$Ra the corresponding values are:

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>\leq 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>&gt;17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient (10^{-7} Sv\cdot Bq^{-1})</td>
<td>47</td>
<td>9.6</td>
<td>6.2</td>
<td>8.0</td>
<td>15</td>
<td>2.8</td>
</tr>
<tr>
<td>Effective dose (mSv\cdot y^{-1})</td>
<td>1.2</td>
<td>0.3</td>
<td>0.2</td>
<td>0.3</td>
<td>0.8</td>
<td>0.2</td>
</tr>
</tbody>
</table>

3.1.1.3. $^{222}$Rn

The same approach used for ingestion of bottled spa water was applied. In fact, two consumption hypotheses were considered, “A” and “B”, the NRC dose coefficient were used [7] and mineral water doses were calculated for all ages. Table IV, V and VI show results of calculations based on the assumption that $^{222}$Rn activity concentration at bottling time was 100 Bq\cdot l^{-1}, whereas $^{210}$Po and $^{210}$Pb were not present.

Table IV. Effective doses for ingestion hypothesis “A”: radon gas only. This gives the highest – but unrealistic – dose values.

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>\leq 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>&gt;17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient (10^{-9} Sv\cdot Bq^{-1})</td>
<td>40</td>
<td>23</td>
<td>10</td>
<td>5.9</td>
<td>4.2</td>
<td>3.5</td>
</tr>
<tr>
<td>Effective dose (mSv\cdot y^{-1})</td>
<td>1.0</td>
<td>0.81</td>
<td>0.35</td>
<td>0.21</td>
<td>0.23</td>
<td>0.26</td>
</tr>
</tbody>
</table>

Concerning ingestion hypothesis “B”, in Table V the $^{210}$Pb contribution is shown. The $^{210}$Pb activity concentration reaches the highest value of 0.047 Bq\cdot l^{-1} after about 40 days and remains almost constant for the following two years with an average value of 0.045 Bq\cdot l^{-1}.
Table V. Effective doses for ingestion hypothesis “B”: $^{210}$Pb contribution.

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>$\leq$ 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>$&gt;$17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient ($10^{-6}$ Sv·Bq$^{-1}$)</td>
<td>8.4</td>
<td>3.6</td>
<td>2.2</td>
<td>1.9</td>
<td>1.9</td>
<td>0.69</td>
</tr>
<tr>
<td>Effective dose (mSv·y$^{-1}$)</td>
<td>0.095</td>
<td>0.057</td>
<td>0.035</td>
<td>0.030</td>
<td>0.046</td>
<td>0.023</td>
</tr>
</tbody>
</table>

In Table VI the similar evaluations are presented for the $^{210}$Po contribution. The $^{210}$Po activity concentration reaches the highest value of 0.044 Bq·l$^{-1}$ after about 2 years with an average value of 0.045 Bq·l$^{-1}$ in this period.

Table VI. Effective doses for ingestion hypothesis “B”: $^{210}$Po contribution.

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>$\leq$ 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>$&gt;$17</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose coefficient ($10^{-9}$ Sv·Bq$^{-1}$)</td>
<td>26.0</td>
<td>8.8</td>
<td>4.4</td>
<td>2.6</td>
<td>1.6</td>
<td>1.2</td>
</tr>
<tr>
<td>Effective dose (mSv·y$^{-1}$)</td>
<td>0.215</td>
<td>0.102</td>
<td>0.051</td>
<td>0.030</td>
<td>0.029</td>
<td>0.029</td>
</tr>
</tbody>
</table>

3.1.1.4. $^{210}$Po and $^{210}$Pb

If $^{210}$Po and $^{210}$Pb are present in the mineral water at bottling time, with the same consumption hypotheses and effective dose coefficients utilized in the two previous paragraphs, the committed effective doses per 1 Bq·l$^{-1}$ of activity concentration are:

Table VII.

<table>
<thead>
<tr>
<th>Age (years)</th>
<th>$\leq$ 1</th>
<th>1–2</th>
<th>2–7</th>
<th>7–12</th>
<th>12–17</th>
<th>$&gt;$17</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{210}$Pb effective dose (mSv·y$^{-1}$)</td>
<td>2.10</td>
<td>1.26</td>
<td>0.77</td>
<td>0.67</td>
<td>1.03</td>
<td>0.50</td>
</tr>
<tr>
<td>$^{210}$Po effective dose (mSv·y$^{-1}$)</td>
<td>6.50</td>
<td>3.08</td>
<td>1.54</td>
<td>0.91</td>
<td>0.86</td>
<td>0.88</td>
</tr>
</tbody>
</table>

3.1.2. Regulations and Criteria used to elaborate the natural radionuclide activity concentration levels.

Due to the absence of specific regulations on natural radioactivity in mineral and spa waters, the effective doses presented in the previous section were compared with reference values extracted from regulations concerning drinking water.

In Italy, the natural radioactivity in drinking water, also if bottled, is regulated by Legislative Decree 31/2001 [15] which enforces European Council Directive 98/83/EC [13]. The Directive introduces the concept of “total indicative dose” (TID), an indicator/parameter equal to 0.1 mSv·year$^{-1}$. This is not a limit but a reference for monitoring purposes and for evaluation of whether any risk to human health is present. The Italian regulation is more conservative as regards bottled drinking water. In fact, when bottled drinking water does not comply with the TID, the Italian local authorities, in collaboration with the local health service, have to determine the causes, indicate remedial action to restore the quality of the drinking water and give priority to enforcement action, taking into account the extent to which the TID has been exceeded and the potential danger to people. It is worthy of note that the TID does not include doses for potassium-40, radon and radon decay products.

For the latter, radon and radon decay products - particularly $^{210}$Pb and $^{210}$Po - references can be found in the European Recommendation 2001/928/Euratom [12]. For radon, a reference value of between 100-1000 Bq·l$^{-1}$ should be used to consider whether remedial action is needed to protect human health. For $^{210}$Po and $^{210}$Pb, reference concentrations are 0.1 Bq·l$^{-1}$ and 0.2 Bq·l$^{-1}$, respectively; these values correspond to an effective dose per adult of 0.1 mSv·year$^{-1}$, i.e. the parametric value for the TID.

The committed effective dose calculations presented above show that the highest doses are those related to infants. In order to protect all members of the public, it was proposed that infants be
considered the reference - or critical - group. Therefore, reference concentrations were worked out in compliance with the reference doses of 0.1 mSv·year⁻¹ for infants.

As regards natural uranium and radium, Italian regulations for drinking water set the TID at 0.1 mSv·year⁻¹. Based on this value, the maximum admitted concentration (MAC) for any single radionuclides was calculated on the assumption that no other radionuclides are present. In the event of the contemporary presence of other radionuclides, their concentration must be such that the total dose of 0.1 mSv·year⁻¹ is not exceeded. In order to obtain this condition, the following expression must be satisfied:

\[ \frac{C_U}{MAC_U} + \frac{C_{Ra-228}}{MAC_{Ra-228}} + \frac{C_{Ra-226}}{MAC_{Ra-226}} \leq 1 \]  

(1)

3.1.2.1. Natural uranium

Based on the effective dose calculations and considerations reported above, the natural uranium concentration should not exceed 3 Bq·l⁻¹ for adults and 1.2 Bq·l⁻¹ for infants. It should be noted that these concentration values correspond to 0.16 mg·l⁻¹ and 0.064 mg·l⁻¹, respectively, figures much higher than the 0.002 mg·l⁻¹ limit recommended by the WHO [17] as regards the toxicological effects of uranium. In conclusion, the proposed MAC for natural uranium is \( MAC_U = 1.2 \text{ Bq·l}^{-1} \).

3.1.2.2. ²²⁸Ra and ²²⁶Ra

Applying the same method used for natural uranium to ²²⁸Ra, concentrations calculated for adults and for infants were 0.20 Bq·l⁻¹ and 0.01 Bq·l⁻¹, respectively. For ²²⁶Ra, the corresponding concentrations were 0.50 Bq·l⁻¹ and 0.09 Bq·l⁻¹. On the basis of these results the proposed MAC values for the two radium isotopes are 0.01 Bq·l⁻¹ for ²²⁸Ra and 0.09 Bq·l⁻¹ for ²²⁶Ra.

3.1.2.3. ²²²Rn

The European Recommendation [12] on the protection of the public against exposure to radon in drinking water supplies sets the radon concentration action level between 100–1000 Bq·l⁻¹. The corresponding effective doses are 0.26–2.56 mSv·year⁻¹ for adults and 1–10 mSv·year⁻¹ for infants. These fairly high dose values are probably accepted by the European Recommendation because drinking water is a primary good. This is not true of mineral water, especially since lots of mineral waters with very low radioactivity levels are available on the market. On the other hand, the dose values cited above are applicable to tap water and to mineral water just bottled. If the radon concentration at bottling time is 100 Bq·l⁻¹, the unavoidable delay between bottling and consumption makes the actual radon concentration very low and the consequent effective dose negligible even for infants. Therefore, the ²²²Rn present at bottling time can be assumed to be 100 Bq·l⁻¹. Yet, hypothesising a ²²²Rn concentration of 100 Bq·l⁻¹ at bottling time, ²²²Rn decay produces a growth in ²¹⁰Pb and ²¹⁰Po concentrations (averaged over the two years of the mineral water’s saleability) corresponding to a total dose of 0.064 mSv·year⁻¹ for adults and 0.388 mSv·year⁻¹ for infants. Therefore, in order to protect infants to the same degree as required for natural uranium and radium isotopes (i.e. an effective dose lower than 0.1 mSv·year⁻¹), an upper level of about 30 Bq·l⁻¹ is required, a value easy to achieve given the availability of simple and effective systems for reducing ²²²Rn in water.

3.1.2.4. ²¹⁰Pb and ²¹⁰Po

The evaluations reported in the previous section were based on the hypothesis that ²¹⁰Pb and ²¹⁰Po are not present in mineral water at the time of bottling. If this is not the case, reference values can be found in the already cited Recommendation [12] on drinking water, where 0.2 and 0.1 Bq·l⁻¹ are proposed for ²¹⁰Pb and ²¹⁰Po, respectively, corresponding to 0.1 mSv·year⁻¹. But these concentrations are suitable for adults only, whereas the corresponding effective doses for infants would be much higher than 0.1 mSv·year⁻¹. In order to reach this dose for infants, ²¹⁰Pb and ²¹⁰Po activity concentrations should not exceed 0.048 e 0.015 Bq·l⁻¹, respectively (see section 3.1.1.4.).
3.2. Summary of the Consiglio Superiore di Sanità opinion

On the basis of the dosimetric evaluations and radiation-protection remarks presented in the report of the Istituto Superiore di Sanità summarized above, the CSS proposed activity concentration upper values, taking the following aspects into account:

- high and increasing consumption of mineral water;
- the evolution of knowledge on risks related to ionizing radiations;
- the absence of Italian or European regulations concerning the presence of natural radionuclides in mineral water;
- the effective doses due to natural radionuclides calculated for infants substantially higher than for adults.

Consequently, in the absence of specific regulations, the CSS considered Italian and European regulations concerning radioactivity in drinking water as a reference [12,13,15]. Moreover, the CSS decided to adopt a general line aimed at containing and reducing the effective doses due to the consumption of radioactive mineral water. Therefore, considering that effective systems for reducing concentrations of uranium, radium and radon in water are widely used in many countries, the CSS proposed the following upper values for mineral water: 2.9 Bq·l⁻¹, 0.2 Bq·l⁻¹ and 0.5 Bq·l⁻¹ for natural uranium, ²²⁸Ra and ²²⁶Ra, respectively, in case these radionuclides are present alone. Otherwise, the following formula should be satisfied in order to obtain a total effective dose lower than 0.1 mSv·year⁻¹ for adults:

\[
\frac{C_U}{MAC_U} + \frac{C_{Ra-228}}{MAC_{Ra-228}} + \frac{C_{Ra-226}}{MAC_{Ra-226}} \leq 1
\]  

(1a)

where MACs (Maximum Activity Concentrations) are the already cited upper values for natural uranium, ²²⁸Ra and ²²⁶Ra, respectively. These values could be lowered by Italian or European regulations on the basis of the chemical toxicity of these radionuclides (e.g. uranium [17]).

As regards radon and its decay products in bottled mineral water, the CSS proposed the following levels: 100 Bq·l⁻¹, 0.1 Bq·l⁻¹ and 0.2 Bq·l⁻¹ for ²²²Rn, ²¹⁰Po and ²¹⁰Pb, respectively. Moreover, it stated that infants should not consume mineral water with natural radionuclide activity concentrations exceeding the following values:

- 1.1 Bq·l⁻¹ for natural uranium
- 0.01 Bq·l⁻¹ for ²²³Ra
- 0.09 Bq·l⁻¹ for ²²⁹Ra
- 32 Bq·l⁻¹ for ²²²Rn
- 0.02 Bq·l⁻¹ for ²¹⁰Po
- 0.05 Bq·l⁻¹ for ²¹⁰Pb

Finally, the Consiglio invited the legislative office of the Ministry of Health to look into the possibility of introducing regulations to implement this opinion and suggested providing for a re-examination of the issue after three years, in the light of possible new scientific knowledge.

4. Conclusions

In the absence of specific regulations to manage the problem of natural radioactivity content in mineral and spa waters, the Ministry of Health, on the basis of the two reports of the Istituto Superiore di Sanità and of the two CSS opinions, undertook important actions. Indeed, upper levels were introduced for the ²²²Rn concentration of spa waters to be complied with for local use and marketing authorizations.
REFERENCES


