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Radionuclides in Italian Drinking Water and Regulations: Data Collection to Improve Risk Assessment

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Abstract: Drinking water, in addition to the best-known chemical and biological agents, contains radionuclides of both natural and artificial origin, which can contribute significantly to the overall effective dose received by the population. The Italian Decree Law 28/2016, implementing 2013/51/EURATOM Directive, establishes the activities for risk management and the parameter values for different radionuclides activity concentrations. In addition to the institutions involved, annually the National Inspectorate for Nuclear Safety and Radiation Protection (ISIN), publishes the monitoring reports of environmental radioactivity in Italy, including radioactivity in drinking water. The purpose of the study was to integrate ISIN reports with 2018 to 2020 data by collecting measurements performed by institutional laboratories to obtain a more complete information and adding, for the Campania region, some data not yet published. This new updated report was not significantly different from ISIN's one, meaning that those publications are nevertheless extremely representative of the radioactivity in Italian drinking water. However, the study allowed to obtain a more detailed data also including measurements not considered in ISIN reports, for instance, radon-222 activity concentrations. This may be of great usefulness for all radiation protection stakeholders in order to ensure environmental protection, pollution prevention and the population safety.

Keywords: environmental radioactivity; drinking water; regulation; monitoring; risk assessment; data analysis; Italy

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1. Introduction

Radionuclides can be found in air, water, soil, and living organisms as consequence of natural and artificial contamination [1]. However, sources of natural origin mostly contribute to environmental radioactivity. In particular, radioisotopes in soil and rocks are those from uranium-238 (U-238) and thorium-232 (Th-232) chains, and potassium-40 (K-40). These nuclides, together with any artificial radionuclides due to accidental contamination [2], are released also into groundwater through erosion and dissolution processes [3,4].

Given the great heterogeneity of radionuclides in drinking water, the radiological risk management is done by performing measurements of gross alpha and gross beta activity concentrations [5–8]. Moreover, particular attention is also given to activity concentration of radon (Rn-222) and tritium (H-3) [9].

Rn-222, originating from U-238 series, is dissolved in water given its physio-chemical characteristics [10]. The biological hazard associated with radon exposure is related to inhalation and ingestion since the alpha particles generated by the decay of radon and its daughters can damage the cells of bronchioles and intestinal tract [11–15]. Despite these two different internal exposure

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scenarios, it has been concluded that about 90% of dose from Rn-222 in drinking water is due to inhalation because of the propriety of the gas to exhale from the water itself [16].

H-3 is a radioisotope of hydrogen with a mainly cosmogenic origin formed in the high atmosphere that reaches the ground with rain [17,18]. A small fraction is H-3 of artificial origin produced by human activities as research and nuclear power plants. Even if its radiotoxicity is low compared to other radionuclides, there is an interest in the biological consequences of low dose H-3 exposures since it can occur in three different forms as tritium water, tritium gas and bounded to organic molecules, each with a different contribution to tissue irradiation because of different distribution modality inside the organism [19,20]. However, given the few data available in literature regarding the risk correlated with tritium exposure, it's hard to assess the related biological effects [21,22].

In this complex framework, it appears essential to regulate and monitor radioactivity in drinking water in order to ensure radiological safety.

Despite the origin of radioactivity, the intake of radionuclides by drinking water is considered as a planned exposure situation rather than a source of environmental background radiation [23]. Globally, the World Health Organization (WHO) [24] and the International Commission on Radiological Protection (ICRP) [25] advise an Individual Dose Criterion (IDC) of 0.1 mSv/year as individual dose limit, a value considered sufficiently low to pose minimal risk and not lead to any detrimental health consequences compared to the total exposure to environmental radioactivity.

In order not to exceed the IDC of 0.1 mSv/year, the WHO recommends screening levels of of 0.5 Bq/l and 1 Bq/l for gross alpha and beta activities respectively. If the gross beta activity is higher than 1 Bq/l, the K-40's contribution is evaluated, and the residual beta activity is calculated by subtraction. The result of this additional assessment might indicate that no action is needed, or it may suggest that further evaluation is necessary before the implementation of measures to decrease the dose.

For low energy beta emitters, as H-3, no monitoring guidelines are given.

Even about Rn-222, WHO does not provides instructions on radiological risk management related to water ingestion while it only highlights the need of monitoring the radon concentration in air and the hazards associated with its inhalation. Consequently, screening levels on radon concentration in drinking water must be based on those currently valid for air indoor activities.

By EURATOM Council Directive n°51 of 2013 [26], European Union adopted a parameter value, i.e. screening level, of 100 Bq/l for Rn-222 and, accordingly, for H-3 too, with an Indicative Dose (ID) of 0.1 mSv/year which takes into account the annual effective dose from ingestion resulting by all natural and artificial nuclides but H-3, K-40 and Rn-222 [27]. Furthermore, compared to WHO, the European Commission recommends a more severe parameter value for gross alpha activity and the same for gross beta activity. These are, in fact, set to 0.1 and 1.0 Bq/l respectively.

In Italy, the legislative framework which regulates the radioactivity in drinking water is given by the Decree Law n°28 of 2016 [28] which implemented the 51/2013/EURATOM Directive [26] lowering the screening level for gross beta activity at 0.5 Bq/l. If the measured gross alpha and beta activity concentrations are lower than their parameter values, no further actions are required since ID<0.1 mSv. In case one of these two parameter values is exceeded, it's required to determine the activity concentration of specific nuclides to calculate the ID, which is evaluated assuming an annual water intake of 730 l with Equation 1:

$$ID\ (mSv) = \sum_{i=1}^{n} \frac{A_i^{(m)}}{A_i^{(d)}}\ 0. \tag{1}$$

with n being the number of radionuclides, while $A_i^{(m)}$ and $A_i^{(d)}$ are the measured and derived (tabulated) activity concentrations for the i-th nuclide, respectively.

If ID<0.1 mSv, no further actions are required. In case ID>0.1 mSv, corrective actions are required in order to decrease the activity concentration and make them in compliance with the law. Risk management is carried out by the Ministry of Health in collaboration with the Istituto Superiore di Sanità (ISS) which, through the measurement and control activities of the territorial Regional Agencies for Environmental Protection (ARPAs), and Autonomous Provinces Agencies for

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Environmental Protection (APPAs), can check compliance with the provisions of the law and apply any activities for public health protection.

Furthermore, National Inspectorate for Nuclear Safety and Radiation Protection (ISIN), for each annual campaign of monitoring, publishes a report summarizing all the collected information and measured values about environmental radioactivity. Data are presented cohesively but measurements are performed independently by 19 ARPAs, 2 APPAs and 10 Experimental Zooprophylactic Institutes (IIZZSSs). This work is not only required by law but it also is essential for all the stakeholders for radiation protection and monitoring of drinking water.

However, published reports do not always include data regarding all 20 Italian regions, especially for radionuclides in drinking water where the measurements are typically inhomogeneous and partially lacking. In addition, ISIN does not provide any information on Rn-222 activity concentration in drinking water, but only gross alpha, gross beta and H-3 activity concentrations.

The purpose of this work is to implement and integrate the latest reports published by ISIN by adding, whenever available, lacking data from missing regions provided by institutional laboratories to obtain a more complete outline of radioactivity in Italian drinking water.

2. Materials and Methods

2.1. Institutional data collection

Institutional data published by ISIN about environmental radioactivity monitoring in Italy were considered. The data are published within reports freely available online on the official website (www.isinucleare.it). To date, these reports contain measurements performed in years 2018 [29], 2019 [30] and 2020 [31].

In each of these reports, average of measured values of gross alpha, gross beta and H-3 activity concentrations in drinking water performed by individual regions are presented together with the total number of measurements, including and specifying those with a value lower than the Minimum Detectable Activity (MDA).

Activity concentration values from missing regions were added by searching official data published independently by individual ARPAs and APPAs. By accessing to publication section for each official ARPA/APPA's website, data over radionuclides in drinking water were sorted, whenever available, according to:

- Type of parameter (gross alpha, gross beta, tritium, radon and other radioisotopes activity concentrations).
- Measurement year.
- Type of data (average activity concentration values, total number of measurements, number of measurements with values lower than the MDA).
- Presentation of data (tabular format, spreadsheet, histogram/graphical, interactive map).

For this work gross alpha, gross beta, H-3 and Rn-222 activity concentrations measured during the three-year period 2018-2020 were considered. Given a specific parameter, if the average activity concentration values were available, all types of data were collected, otherwise they were excluded. Moreover, some of the ARPAs and APPAs also give a description of the methodology used to perform the measurements together with provided data.

For Campania region, measurements carried out in a certificated ISO 9001:2015 laboratory [32] by La Verde et al. [33–35] during the years 2018 and 2019 were considered. For year 2020, given the lack of data, measurements performed by the same authors with the same methodology were included despite not being published. Details on materials and methods for gross alpha, gross beta, H-3 and Rn-222 activity concentration measurements in Campania are given in subsection 2.2.

After all the data have been collected, weighted average activity concentration values were calculated considering the total number of measurements for each Italian macro-area. According to ISIN reports, the three macro-areas are defined as follow:

a) North: Emilia-Romagna, Friuli-Venezia Giulia, Liguria, Lombardy, Piedmont, Trentino-Alto Adige, Aosta Valley and Veneto.
b) Center: Lazio, The Marches, Tuscany, Umbria and Sardinia.
c) South: Abruzzo, Basilicata, Calabria, Campania, Molise, Apulia and Sicily.

2.2. Sampling and radionuclides Activity Concentration Evaluation in Campania

The same methodology described in the work by La Verde et al. [33] has been used to perform measurements in 2020 in Campania.

2.2.1. Sample preparation

During 2020, a total of 13 samples were collected from sites (dwells) distributed in three water subsystems in Campania region [33]. Additionally, 7 more samples were collected only for Rn-222 measurements from the same sampling points.

Sample preparation was performed according to standard techniques and procedures [36–38] for each parameter measurement. The procedures used for each parameter together with measurement techniques and detection limits are summarized in Table 1.

Table 1. Procedures and techniques used to perform measurements of gross alpha, gross beta, H-3 and Rn-222 activity concentrations in Campania. Calculated detection limits for each parameter are lower or equal than those required by Decree Law 28/2016 [28].

Parameter	Standard Procedure	Measurement Technique	Detection Limit (Bq/l)
Gross alpha	EPA 900 [36]	Alpha spectrometry	0.02
Gross beta	EPA 900 [36]	Proportional chamber	0.2
Tritium	ISO 9698 [37]	Liquid scintillation	3
Radon	ISO 13164-1 [38]	Electret	1

2.2.2. Activity concentration measurements

For details on instrumentations and methods used to perform the measurements refer to La Verde et al. [33]. Some specifications are following:

1. For gross alpha activity concentration A_α measurements, the Ortec® Alpha Duo spectrometer (Peschiera Borromeo, Milan, IT) and the detectors ULTRA-AS, with an efficiency of 0.0332, were used. For gross beta activity concentration A_β measurements, the proportional counter Berthold Technologies Umo LB 123, with an efficiency of 0.105 was used. Both activities were calculated with equation 2:

$$A_{a,\beta}\left(\frac{Bq}{l}\right) = \frac{CPS_{net}}{Efficiency_{a,\beta}} \cdot \frac{1}{V} \tag{2}$$

where V=0.05 l is the sample volume and CPS_{net} is the net counts per second obtaining after background counts subtraction.

2. For H-3 activity concentration $A_{H\text{-}3}$ measurements, the PerkinElmer® Wallac 1220 Quantulus liquid scintillator was used. After having determined the background activity A_{BKG}, equation 3 was used to evaluate the net H-3 activity after measuring the activity in the sample A_T:

$$A_{H-3}\left(\frac{Bq}{l}\right) = A_T - A_{BKG} \tag{3}$$

3. For Rn-222 in water activity concentration A_{Rn-222}^{water} measurements, the Electret Ion Chamber (EIC) E-Perm® system was used. Equations 4, 5 and 6 were used for the evaluation:

$$A_{Rn-222}^{water} = A_{Rn-222}^{air} \cdot B_1 \cdot B_2 \cdot B_3 \tag{4}$$

$$A_{Rn-222}^{air} = \left[\frac{V_i - V_f}{CF \cdot T} - G_\gamma \cdot C_1\right] \cdot 37 \tag{5}$$

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$$CF = C_2 + C_3 \cdot \frac{V_i + V_f}{2} \qquad (2)$$

where A^{air}_{Rn-222} is the radon concentration in the air inside the jar; B_1 considers the delay period between the collection of the sample and the start of the measurement; B_2 is a constant based on analysis period; B_3 is the ratio between the jar and water sample volumes; V_i and V_f are the electret voltages before and after the exposure respectively; T is the exposure time; G_γ is the background signal due to gamma radiation and C_1=0.097, C_2=1.670 and C_3=5.742×10^{-4} are constants provided by the instrumentation manufacturer; CF is the calibration factor.

3. Results and discussions

Among all the 21 ARPAs and APPAs, only 12 of them published results of at least one measurement campaign since 2016 (i.e., the year of the Italian Decree Law n°28 [28]). However, among these 12, only 6 met the following criteria for at least one parameter:

- To provide the precise result of average activity concentration for at least one year between 2018 and 2020.
- To provide the total number of measurements.
- Not to be included into any considered ISIN report.

Subsections with summarized data for each parameter will follow. More specifically, the average activity concentration values (A), the total number of measurements performed (N) and the number of measurements with values lower than the MDA (N<MDA) (annex III table 2 of [28]) will be reported for each parameter, relatively to years 2018, 2019 and 2020.

3.1. Gross alpha activity concentration

For gross alpha activity concentrations data from four regions were added with respect to ISIN reports. Measurements performed in Veneto [39], Tuscany [40], Sardinia [41] and Campania [33] between 2018 and 2020 together with values provided by ISIN for gross alpha activity concentration are showed in Table 2.

Table 2. Measurements of Gross alpha activity concentrations in Italian drinking water in 2018-2020. The sign "-" indicates that data were already published by ISIN; "na" stands for "not available"; colored in gray are the updated data with respect to ISIN ones.

Gross Alpha		2018			2019			2020			Ref.
Macro-areas	*Region*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	
North	*Lombardy*	112	17	<0.054	86	12	<0.078	67	5	<0.087	
	Piedmont	28	17	<0.049	306	110	<0.035	322	167	<0.042	
	Emilia-Romagna	4	4	<0.018	63	39	<0.032	na	na	na	
	Liguria	na	na	na	144	65	<0.056	401	304	<0.044	
	Friuli-Venezia Giulia	na	na	na	74	8	<0.043	77	4	<0.046	[29]
Center	*Lazio*	2	1	<0.054	2	0	0.040	2	1	<0.061	
	The Marches	29	17	<0.035	38	17	<0.042	18	6	<0.029	[30]
	Tuscany	47	23	<0.034	65	25	<0.036	na	na	na	
	Umbria	na	na	na	96	2	<0.047	67	0	0.043	[31]
	Sardinia	na	na	na	na	na	na	41	40	<0.072	
South	*Basilicata*	4	3	<0.022	6	3	<0.046	2	2	<0.152	
	Calabria	189	77	<0.062	208	107	<0.055	179	68	<0.076	
	Campania	29	14	<0.123	1	0	0.079	na	na	na	
	Apulia	na	na	na	na	na	na	10	4	<0.033	
North	*Veneto*	228	175	<0.040	471	293	<0.042	407	284	<0.041	[39]

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Center	*Tuscany*	-	-	-	-	-	-	221	na	<0.038	[40]
	Sardinia	14	9	<0.052	56	22	<0.044	-	-	-	[41]
South	*Campania*	55	20	<0.038	28	14	<0.035	13*	4*	<0.055*	[33]

* Unpublished data.

3.2. Gross beta activity concentration

For gross beta activity concentrations data from three regions were added with respect to ISIN reports. Measurements performed in Tuscany [40], Sardinia [41] and Campania [33] between 2018 and 2020 together with values provided by ISIN for gross beta activity concentration are showed in Table 3.

Table 3. Measurements of Gross beta activity concentrations in Italian drinking water in 2018-2020. The sign "-" indicates that data were already published by ISIN; "na" stands for "not available"; colored in gray are the updated data with respect to ISIN ones.

Gross Beta		**2018**			**2019**			**2020**			**Ref.**
Macro-areas	*Region*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	
North	*Lombardy*	112	64	<0.100	86	59	<0.137	67	50	<0.133	
	Piedmont	40	30	<0.141	326	162	<0.114	322	152	<0.110	
	Emilia-Romagna	4	0	0.084	63	0	0.067	na	na	na	
	Liguria	na	na	na	155	102	<0.292	401	389	<0.257	
	Friuli-Venezia Giulia	na	na	na	74	32	<0.193	77	5	<0.231	[29]
Center	*Lazio*	2	1	<0.153	2	1	<0.170	2	0	0.741	
	The Marches	29	2	<0.189	38	22	<0.121	18	13	<0.083	[30]
	Tuscany	47	22	<0.125	65	43	<0.145	na	na	na	
	Umbria	na	na	na	96	15	<0.116	67	4	<0.153	[31]
	Sardinia	na	na	na	na	na	na	41	12	<0.147	
South	*Basilicata*	4	0	0.204	7	0	0.295	2	1	<0.310	
	Calabria	189	152	<0.218	208	184	<0.209	179	142	<0.221	
	Campania	29	1	<0.423	na	na	na	na	na	na	
	Apulia	na	na	na	1	0	0.156	10	1	<0.168	
Center	*Tuscany*	-	-	-	-	-	-	221	na	<0.161	[40]
	Sardinia	14	9	<0.251	56	40	<0.136	-	-	-	[41]
South	*Campania*	-	-	-	28	17	<0.221	13*	12*	<0.212*	[33]

* Unpublished data.

3.3. Tritium activity concentration

For H-3 activity concentrations no data was found from all the ARPAs and APPAs websites, therefore only measurements performed by La Verde et al. [33] were included for years 2019 and 2020. Table 4 presents all the reported values of H-3 activity concentration in the three years.

Table 4. Measurements of H-3 activity concentrations in Italian drinking water in 2018-2020. The sign "-" indicates that data were already published by ISIN; "na" stands for "not available"; colored in gray are the updated data beyond ISIN ones.

H-3		**2018**			**2019**			**2020**			**Ref.**
Macro-areas	*Region*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	
North	*Lombardy*	47	47	<5.42	14	14	<4.66	14	14	<6.20	[29]
	Piedmont	24	24	<1.97	25	25	<1.86	20	20	<2.86	

	Emilia-Romagna	5	4	<1.33	4	4	<1.65	na	na	na	[30]
	Veneto	4	4	<2.50	na	na	na	na	na	na	
Center	*Lazio*	2	1	<8.60	2	0	8.05	2	2	<4.85	
South	*Basilicata*	4	2	<2.76	8	6	<2.16	2	2	<2.03	[31]
	Calabria	187	177	<10.05	199	199	<10.00	156	156	<10.00	
	Campania	29	29	<3.00	na	na	na	na	na	na	
	Apulia	na	na	na	1	0	5.90	7	1	<6.40	
South	*Campania*	-	-	-	28	28	<2.72	13*	13*	<10.0*	[33]

* Unpublished data.

3.4. Radon activity concentration

For Rn-222 activity concentrations data from seven regions were included. Measurements performed in Lombardy [42], Veneto [39], Lazio [43], The Marches [44], Tuscany [40], Sardinia [41] and Campania [33] between 2018 and 2020 are showed in Table 5. Note that none of these measurements is reported in any ISIN report.

Table 5. Measurements of Rn-222 activity concentrations in Italian drinking water in 2018-2020. "na" stands for "not available".

Rn-222		**2018**			**2019**			**2020**			**Ref.**
Macro-areas	*Region*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	*N*	*N<MDA*	*A (Bq/l)*	
North	*Lombardy*	na	na	na	72	0	11.90	66	3	<11.36	[42]
	Veneto	264	27	<8.70	475	82	<8.10	393	48	<8.30	[39]
Center	*Lazio*	6	0	37.34	15	0	27.69	3	0	21.26	[43]
	The Marches	17	na	<2.56	na	na	na	na	na	na	[44]
	Tuscany	na	na	na	181	12	<7.278	156	8	<12.5	[40]
	Sardinia	4	0	2.60	22	3	<8.00	na	na	na	[41]
South	*Campania*	55	0	16.21	28	0	11.71	20*	0*	12.82*	[33]

* Unpublished data.

3.5. Comparisons between ISIN and updated data

To perform a comparison between measurements provided by ISIN and updated values the weighted average for each parameter in all the three Italian macro-areas were calculated using the total number of measurements as weights. Results are showed in Table 6.

Table 6. ISIN and updated values of weighted average H-3, gross alpha, gross beta and Rn-222 activity concentrations in Italian drinking water in 2018-2020 for individual macro-areas. The sign "-"for Rn-222 values in ISIN columns indicates that data are lacking in reports.

		2018		**2019**		**2020**	
Parameter	*Macro-area*	*ISIN*	*Updated*	*ISIN*	*Updated*	*ISIN*	*Updated*
Tritium (Bq/l)	*North*	<3.97	<3.97	<2.80	<2.80	<4.24	<4.24
	Center	<8.60	<8.60	<8.10	<8.10	<4.85	<4.85
	South	<9.07	<8.75	<9.70	<8.90	<9.88	<9.77
Gross alpha (Bq/l)	*North*	<0.052	<0.044	<0.044	<0.044	<0.047	<0.045
	Center	<0.035	<0.037	<0.043	<0.043	<0.051	<0.043
	South	<0.070	<0.064	<0.053	<0.053	<0.075	<0.073
Gross beta (Bq/l)	*North*	<0.11	<0.11	<0.16	<0.16	<0.19	<0.19
	Center	<0.15	<0.16	<0.13	<0.13	<0.15	<0.16
	South	<0.24	<0.24	<0.21	<0.21	<0.22	<0.22
Radon (Bq/l)	*Nord*	-	<8.7	-	<8.6	-	<8.7

	Center	-	<10.3	-	<8.8	-	<12.7
	South	-	<16.2	-	<11.7	-	<12.8

As can be seen from values in Table 6, updated values are not significantly different from the ones published by ISIN. This highlights how, despite not including measurements from all the 20 Italian regions, those reports are extremely effective and representative of the radioactivity distribution in drinking water.

However, results indicate that small variations are still present, meaning that including more regions, that is more measurements for each macro-areas, allows to obtain a more detailed information on parameter values with higher statistics.

Furthermore, independent research of official published data allowed to collect additional information as the Rn-222 distribution in Italian drinking water, which was missing in ISIN reports.

4. Conclusions

This work integrates official data of radioactivity monitoring in Italian drinking water. For the specific case, measurements elaborated and published by ISIN in annual reports were implemented with values taken by institutional sources and laboratories. Comparisons between ISIN and updated values showed that published reports are representative of the Italian framework on radionuclides in drinking water although the same reports are incomplete since many regions aren't included. For the next future it could be interesting to create a network of information flows in order to merge them into a single document that could be continuously updated to obtain a more detailed distribution of radioactivity in Italian drinking water. Institutional laboratories, for example, could have access to a database and upload data. This would also promote more in-depth investigations for Italian regions with few/no measurements, providing not only for regulatory compliance but also for a more extensive radio protection program.

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