IL NUOVO CIMENTO **43 C** (2020) 143
DOI 10.1393/ncc/i2020-20143-y

Colloquia: SIRR 2020

Radon measurements in drinking water using electret according to Italian legislation and mapping of Campania region (Southern Italy)

G. La Verde(1)(2)(3), A. Desiderio(4)(*), M. La Commara(1)(2), V. D'Avino(1), V. Roca(1), F. Coppola(5), M. Panico(6), S. Polichetti(6)
and M. Pugliese(1)(3)

(1) *National Institute of Nuclear Physics, Section of Naples - Naples, Italy*
(2) *Department of Pharmacy, University of Naples Federico II - Naples, Italy*
(3) *Department of Physics "E. Pancini", University of Naples Federico II - Naples, Italy*
(4) *Department of Electrical Engineering and Information Technologies, University of Naples Federico II - Naples, Italy*
(5) *Center for Advanced Metrological Services, University of Naples Federico II - Naples, Italy*
(6) *Gori S.p.A. - Ercolano (Naples), Italy*

received 25 January 2021

Summary. — In this study the radon-222 activity concentration in drinking water over a large area of the western Campania region was investigated. Water samples were collected over a period of three years between May 2017 and May 2019 in compliance with Italian legislation. The measurement of radon activity concentration using electret detector (E-Perm® system) in short-short term configuration was performed. The purpose has been to identify the sampling points and report them on a map, studying the presence of any correlations between variations in the concentration of activity, seasonality and specific characteristics of the plant. The results obtained revealed that the concentrations of radon activity in the drinking water of the investigated sites are below the parameter value established by the law. Furthermore, activity concentration of radon in well water does not depend on seasonality, unlike the tanks, for which there is a relationship with the temperature that must be studied separately on the basis of the single tank.

1. – Introduction

Human being exposure to ionizing radiation is continuous and inevitable due to the presence of different sources such as cosmic radiation and naturally radioactive elements

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(*) Corresponding author. E-mail: `ari.desiderio@studenti.unina.it` that are present in the environment, in addition to human-made sources. Also, drinking water contains radionuclides that could be a risk for human health so its quality must be strictly controlled in terms of natural and anthropogenic radionuclides concentration. Among these radionuclides, the most important is radon gas. It is an inert, noble and ubiquitous gas that comes from the decay of uranium and thorium. The longest-lived isotope and also most abundant in nature is radon-222, which comes from the radioactive decay chain of uranium-238. Its progeny, in particular polonium-210 is the principal source of dose from ingestion of radon in drinking water supplies [1]. Radon is soluble in water and its solubility decreases with increasing temperature [2, 3], so groundwater that passes through uranium-bearing soils and rocks contains radon. When radon-rich groundwater is used as drinking water, people are exposed to radon both by ingesting water and by inhaling the radon exhaled from the water itself [4]. The UNSCEAR 2000 Report [5] has concluded that on average, 90% of the dose attributable to radon in drinking water comes from inhalation rather than ingestion. Radon has been classified by the World Health Organization as the second leading cause of lung cancer death after cigarette smoking and many studies have clearly shown that long-term exposure to high radon concentrations in indoor air increases the risk of lung cancer [6-9]. On the other hand, the relationship with stomach cancer is not so clear. Some studies suggest that there is no causal relationship between radon ingestion and an increased risk of stomach cancer [10-14] while other studies say the opposite [15, 16]. However, the radiological aspect of drinking water quality is widely considered on the international scale [1]. Thus, in 2013, the European Union published the Directive 2013/51 EURATOM [17] and many countries have determined the natural radioactivity in their drinking water [18-22]. The Italian decree n. 28/2016 [23] represents the implementation of directive 2013/51/EURATOM, laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption. It establishes parametric values, frequencies and methods for monitoring radionuclides in drinking water. For measurements of radon activity concentration, surveys must be realized in order to collect information about the geology and hydrology of the area, radioactivity of rock or soil and type of well or plant. The frequency of sampling is determined on the basis of the volume of water distributed or produced each day within a supply zone (Italian decree n. 28/2016 annex II, table I). The parametric value is 100 Bq/L (Italian decree n. 28/2016 article 5, paragraph 1). Any failure to comply with a parametric value must be immediately investigated in order to identify the cause and it must be assessed whether failure represents a risk to human health. The health risk assessment must take into account, when the parametric value is exceeded, the calculation of the indicative dose. If, on the other hand, the average annual concentration of radon activity exceeds the reference value of 1000 Bq/L, the risk assessment must not be performed but corrective measures must be taken directly [24].

Data about activity concentration of radon used in this work come from a monitoring campaign realized by Gori S.p.A., a local company that manages and distributes drinking water. As there are not many studies that integrate measurement data with geographic information [25], the goal of this study has been to use data about sampling points to produce a map of radon activity concentration and to identify any trends.

2. – Materials and methods

In the period between May 2017 and May 2019, 246 samples were collected and analysed. The collection of samples was carried out by specialized Gori operators that manage the water supply, complying with the standard procedures [26]. The municipalities involved have been 38 and the sampling points 99, for a total of 246 samples. Clearly, some points were investigated several times over the three years based on the frequency defined by the legislation. The measurements were performed in the Laboratory of Radioactivity (LaRa) of the Department of Physics “E. Pancini” of the University of Naples Federico II, certified UNI EN ISO 9001:2015 for measures of concentration of activity of radon gas [27]. To perform radon activity concentration measurement an E-Perm$^{\circledR}$ EIC system was used [28]. As reported by Kotrappa *et al.* [29], instrumentation includes:

- electrometer (Rad. Elec. Inc. Mod. 6383-01, Frederick, MD, USA);
- 140 mL glass sample bottle with screw cap;
- 4 L glass jar with hermetically sealed ring cap;
- E-Perm$^{\circledR}$ chamber in Short-Short Term (SST) configuration.

After transport to the laboratory, within about 24 h of sampling, each 140 mL bottle was opened and immediately placed in the glass jar with a suspended E-Perm chamber in Short-Short Term configuration. The jar containing the electret and water sample was sealed (airtight) for 94 h to allow radon to reach equilibrium with its daughters. The radon activity concentration in the water sample was calculated with a formula provided by the manufacturer [30]:

$$C_{\mathrm{Rn}}(\mathrm{water}) = C_{\mathrm{Rn}} + B_1 + B_2 + B_3, \tag{1}$$

where:

- C_{Rn}: radon concentration measured in the air inside the jar with eq. (2) and (3) [Bq/L];
- B_1: period between the collection of the water sample and the start of the measurement [h];
- B_2: period from the time of inserting the sampling bottle into the jar until the E-Perm$^{\circledR}$ is removed [h];
- B_3: ratio between the volume of the jar and the water sample.

The radon concentration was calculated applying the appropriate calibration factor and the exposure time, according to the following equations [31]:

$$C_{\mathrm{Rn}} = \left[\frac{(V_{\mathrm{i}} - V_{\mathrm{f}})}{(CF \cdot T)} - G_\gamma C_1\right] \cdot 37, \tag{2}$$

$$CF = C_2 + C_3 \cdot (V_{\mathrm{i}} + V_{\mathrm{f}})/2, \tag{3}$$

where

- V_{i} and V_{f}: electret voltage readings before and after exposure respectively [V];
- T: exposure time [d];
- G_γ: gamma dose rate [$\mu\mathrm{R} \cdot \mathrm{h}^{-1}$];
- $C_1 = 0.097$, $C_2 = 1.670$, $C_3 = 0.0005742$, constants given by the manufacturer depending on configuration and volume of the E-Perm$^{\circledR}$ chamber.

3. – Results and discussion

The samples are distributed in three water subsystems of the western Campania region: Monti Lattari, Ausino e Vesuviano as shown in table I.

The category "Other" in the table includes sampling points for which no association with water systems has been detected or which are supplied by endogenous sources. All activity concentration values are smaller than the parameter value of 100 Bq/L. Results for each system are shown in table II and the dispersion of values for each system was analysed as shown in fig. 1.

The second analysis addressed the type of plant from which the samples were taken, in particular the activity concentration values for 37 wells and 39 tanks were examined in relation to the season: cold period (from October to March) hot period (from April to September). The results for each plant in the two periods are reported in detail in tables III and IV, furthermore, the comparison between the two plants on the basis of seasonality is shown in fig. 2.

As can be seen from the graph in fig. 2, on average the concentration of radon activity is higher for wells than for tanks, result already known from the literature [2]. In addition, the activity concentration of wells does not change with the season. This result can be explained with a consideration: wells are underground at a great depth and the water inside them is not affected by seasonal climatic variations, so its temperature does not

Table I. – *Number of samples for each system and relative percentage of the total samples analysed.*

System	Number of samples	Percentage
Monti Lattari	33	13%
Ausino	100	41%
Vesuviano	89	36%
Other	24	10%

Table II. – *Activity concentration values for the systems.*

System	Minimum value (Bq/L)	Maximum value (Bq/L)	Medium value (Bq/L)
Monti Lattari	3.4 ± 0.5	42.5 ± 2.2	10.8 ± 0.7
Ausino	3.2 ± 0.3	57.0 ± 2.9	15.5 ± 0.9
Vesuviano	1.3 ± 0.3	55.7 ± 2.8	10.7 ± 0.7
Other	0.7 ± 0.5	55.2 ± 2.8	11.3 ± 0.7

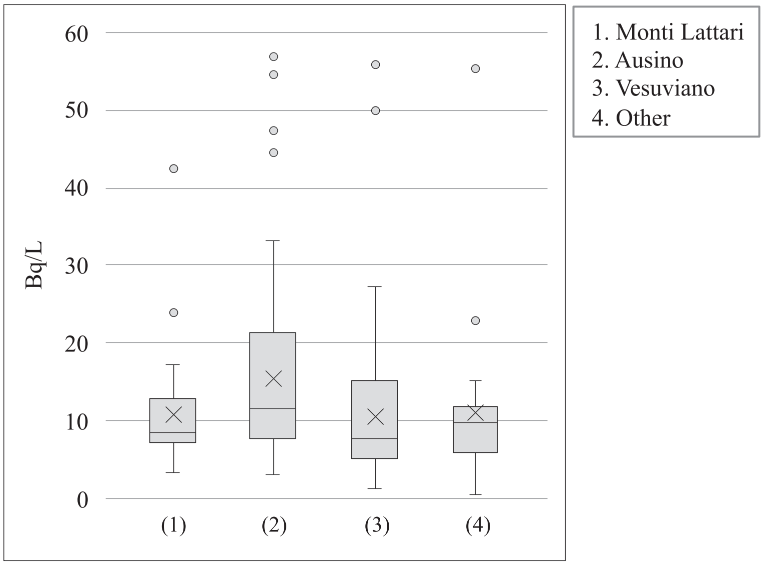

Fig. 1. – Dispersion of radon activity concentrations in the sampling period for each water system.

vary during the year and does not interfere with the solubility of radon in water. This is not true for tanks, for which the activity concentration varies with the season. This can be understood by considering that a single tank can have different characteristics, it can be underground, partially underground, external and in gallery. For this reason, a final analysis only for the tanks was realized. The average activity concentration for the hot and cold period for each water sub-system was determined as reported by fig. 3.

Results show that there may be a dependency on the location of the tank. For example, the tanks of the Ausino system could be external so the water inside them is affected by seasonal climatic variations. In fact, activity concentration of radon is higher in the cold period and lower in the hot period, in accordance with the variation in radon solubility with the temperature. The tanks of the Monti Lattari system, on the other hand, could be partially underground since there are no large variations between the two periods. Nothing can be said for the tanks of the last system. However, this aspect must

Table III. – *Activity concentration values for tanks.*

Tanks	Minimum value (Bq/L)	Maximum value (Bq/L)	Medium value (Bq/L)
Cold period	2.0 ± 0.4	50.0 ± 2.5	9.7 ± 0.7
Hot period	3.5 ± 0.3	42.5 ± 2.2	11.3 ± 0.7

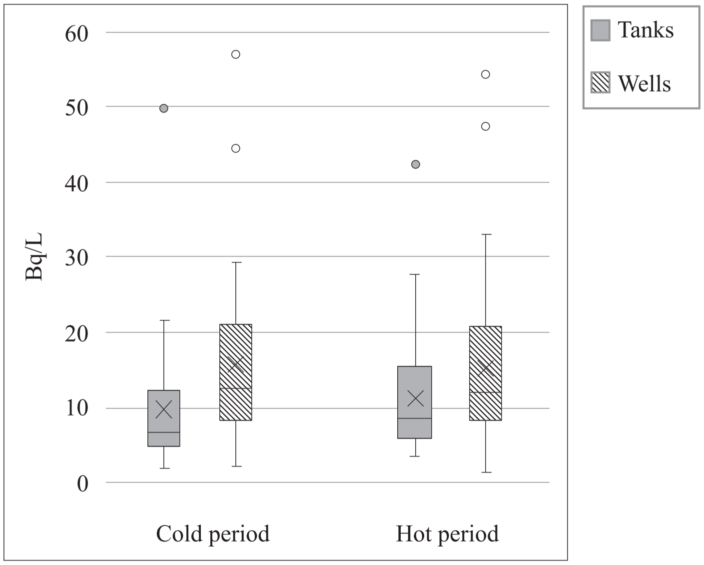

Fig. 2. – Comparison between tanks and wells, analysis based on the type of sampling site and the temperature variation.

be investigated with future studies. Finally, all the sampling points were included in a map (fig. 4) in order to geographically describe the distribution of the systems and the number of samples. This turns out to be really useful for managing the measures and identifying any anomalies with accuracy.

4. – Conclusions

This study should be intended as an initial step. It has been verified that all values of radon activity concentration in drinking water comply with the Italian legislation. Then, the assessments that have been done allow to lay the foundations for a more targeted study in this area. In fact, the production of a map turns out to be fundamental for the individuation and the choice of future monitoring points only where it is really needed, as reported by the decree n. 28/2016 [23], annex II and it should be the starting point for the realization of an additional map in terms of risk level. Among the points involved

Table IV. – *Activity concentration values or wells.*

Wells	Minimum value (Bq/L)	Maximum value (Bq/L)	Medium value (Bq/L)
Cold period	2.2 ± 0.5	57.0 ± 2.9	15.7 ± 0.9
Hot period	1.3 ± 0.3	54.5 ± 2.8	15.1 ± 0.9

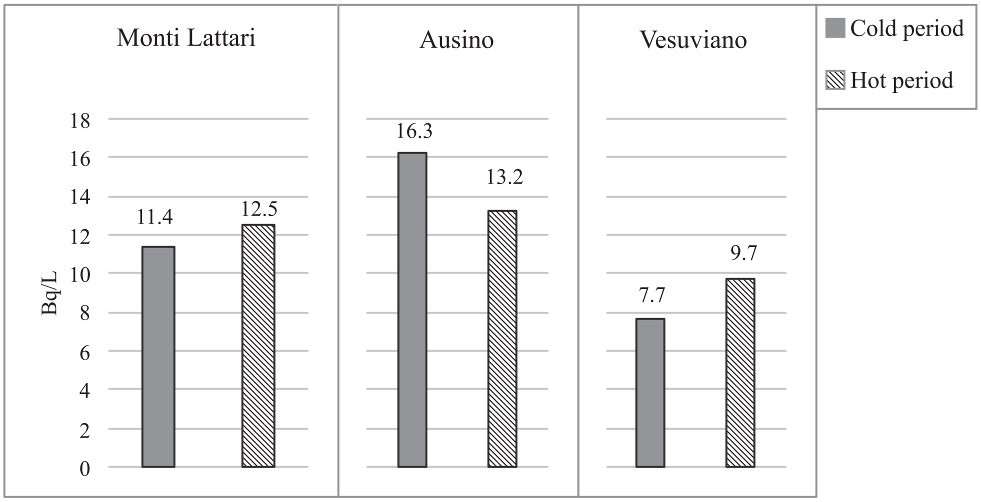

Fig. 3. – Average activity concentration of radon in the tanks of the three water subsystems.

in the monitoring, wells and tanks were selected and the activity concentrations of radon were analysed. It was found that activity concentration is higher in wells and is not affected by seasonal variations. On the other hand, tanks are affected by seasonality but a further investigation must be carried out in terms of characteristics of the single tank.

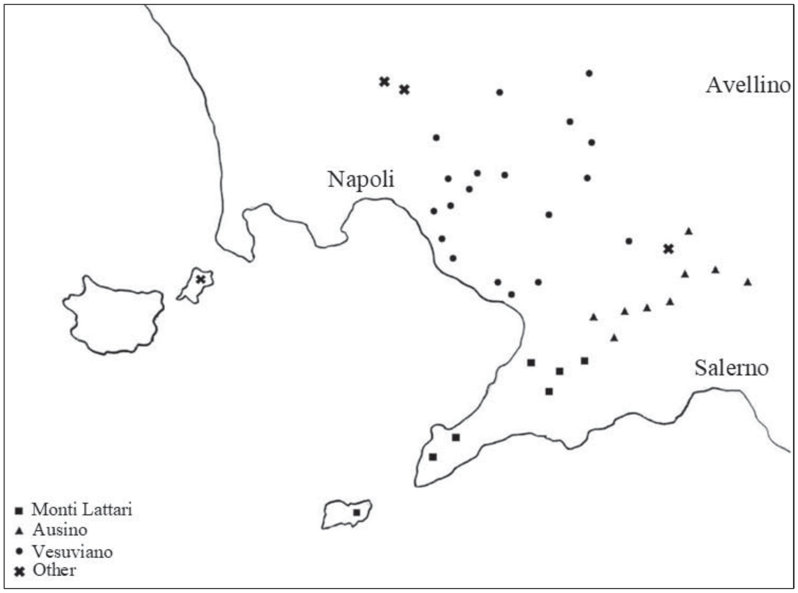

Fig. 4. – Measuring points of water supply systems.

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