

Maximum recommended and allowable Radon-222 limits in water and air: Systematic review

Limites máximos recomendados e permitidos de Radon-222 na água e no ar: Revisão sistemática

Límites máximos recomendados y permisibles de radón-222 en agua y aire: Revisión sistemática

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Abstract

Radon-222 is a radioactive gas that, when inhaled at high concentrations, can harm human health. However, there are several recommended and maximum allowable Radon-222 limits in water and air in the international community. Thus, this research aimed to evaluate the maximum recommended and allowed Radon-222 limits in drinking water and indoor air in the international community, indicating the most referenced organizations in Radon-222 studies through a systematic review. The results indicate that there is variation of up to 1000% between limits established for Radon concentrations in the air indicated by ICRP and US EPA. For water, the maximum limit established by EURATOM is 90 times higher than that established by US EPA. Greater relevance regarding the presence of Radon-222 in the air was also evidenced, due to its potential to detach from various physical means, such as water agitation, its occurrence in building materials and its release by contaminated soil. Finally, it was also found that the limits imposed by US EPA for the presence of Radon-222 in water and air were the parameters most used for comparison in the scientific community and that about 80% of evaluated studies reported from one two sources that proposed Radon-222 limits in air and water.

Keywords: Natural radiation; Laws for Radon 222; Contamination of drinking water and air by radiation; Lung cancer by radiation;²²²Rn; Radon limits.

Resumo

O radônio-222 é um gás radioativo que, quando inalado em altas concentrações, pode prejudicar a saúde humana. No entanto, existem vários limites recomendados e máximos permitidos de Radon-222 na água e no ar na comunidade internacional. Assim, esta pesquisa teve como objetivo avaliar os limites máximos recomendados e permitidos de Radon-222 em água potável e ar interno na comunidade internacional, indicando as organizações mais referenciadas em estudos de Radon-222 por meio de uma revisão sistemática. Os resultados indicam que há variação de até 1000% entre os limites estabelecidos para as concentrações de Radon no ar indicados pelo ICRP e US EPA. Para a água, o limite máximo estabelecido pela EURATOM é 90 vezes superior ao estabelecido pela US EPA. Evidenciou-se também maior relevância quanto à presença do Radon-222 no ar, devido ao seu potencial de se desprender de diversos meios físicos, como agitação da água, sua ocorrência em materiais de construção e sua liberação por solo contaminado. Por fim, verificou-se também que os limites impostos pela US EPA para a presença do Radon-222 na água e no ar foram os

parâmetros mais utilizados para comparação na comunidade científica e que cerca de 80% dos estudos avaliados reportaram de uma duas fontes que propuseram Limites de Radon-222 no ar e na água.

Palavras-chave: Radiação natural; Leis para Radon 222; Contaminação da água potável e do ar por radiação; Câncer de pulmão por radiação; 222Rn; Limites de radônio.

Resumen: El radón-222 es un gas radiactivo que, cuando se inhala en altas concentraciones, puede dañar la salud humana. Sin embargo, existen varios límites de radón-222 recomendados y máximos permitidos en el agua y el aire en la comunidad internacional. Por lo tanto, esta investigación tuvo como objetivo evaluar los límites máximos recomendados y permitidos de Radon-222 en agua potable y aire interior en la comunidad internacional, indicando las organizaciones más referenciadas en los estudios de Radon-222 a través de una revisión sistemática. Los resultados indican que existe una variación de hasta un 1000% entre los límites establecidos para las concentraciones de Radón en el aire indicados por ICRP y US EPA. Para el agua, el límite máximo establecido por EURATOM es 90 veces superior al establecido por la US EPA. También se evidenció mayor relevancia en cuanto a la presencia de Radón-222 en el aire, debido a su potencial para desprenderse de diversos medios físicos, como la agitación del agua, su ocurrencia en materiales de construcción y su liberación por suelo contaminado. Finalmente, también se encontró que los límites impuestos por la US EPA para la presencia de Radon-222 en agua y aire fueron los parámetros más utilizados para la comparación en la comunidad científica y que cerca del 80% de los estudios evaluados reportaron de una dos fuentes que proponían Límites de radón-222 en aire y agua.

Palabras clave: Radiación natural; Leyes para el Radón 222; Contaminación del agua potable y del aire por radiación; Câncer de pulmón por radiación; 222Rn; Límites de radón.

1. Introduction

Part of the radiation present in the world is generated through the environment. Called natural radiation, it is characterized by being the main source of radiation exposure for the world population (UNSCEAR 2008; Raghu et al. 2017). There are two types of natural radiation: cosmic radiation, coming from the solar system, which contributes to 16% of the total exposure, and terrestrial radiation, which accounts for 84% and originates from natural elements present in the soil (Mazzilli 2011).

Even though it originates from materials coming from the Earth's crust, terrestrial radiation can permeate to different media, such as building materials, air, water, food and the human body itself, all through the use of natural resources (Ahmad et al., 2014).

Thorium ^{232}Th , Actinium ^{235}U and Uranium ^{238}U are among the elements that emit terrestrial radiation. Uranium ^{238}U is found in soils and rocks, such as rock phosphate, which obtains high amounts of natural terrestrial radionuclides (UNSCEAR 2000). Radon 222 (Rn^{222}) is a radionuclide resulting from several successive Uranium ^{238}U decays, characterized as a colorless, odorless and tasteless gas (Desideri 2005; Thabayneh 2015; Abuelhia 2017).

Due to its gaseous form, it can be rapidly diffused in media in which it comes into contact, allowing it to be inhaled through the air and/or ingested in the aqueous medium. When ingested or inhaled, this element can cause serious harm to human health, such as lung cancer (Khan et al. 2000; Duggal et al. 2016; Torres-Duran et al. 2014).

In the United States, Radon 222 was found to be the second leading cause of death from lung cancer, killing 21,000 inhabitants/year, being surpassed only by deaths resulting from smoking (National Cancer Institute 2010). When present in drinking water, Radon 222 can also pose a significant risk of stomach and gastrointestinal cancer, especially when water comes from underground sources (Duggal 2019).

In the world scientific literature, there are records from several countries that confirmed environmental contamination by Radon 222, among them Venezuela, Iran, China, India, Brazil, Japan and Mexico. However, in all of these countries, there is lack of national laws and guidelines that address the maximum permitted Radon 222 concentrations in drinking water and air (Horvahth et al. 2000; Keramati et al. 2018; Wang et al. 2017; Kumar et al. 2016; Janik et al. 2009; Espinosa et al. 2009). In these cases, the standards imposed by developed nations such as the United States or organizations such as the United Nations, are used as comparative limits (WHO 2001; UNSCEAR 2008).

The maximum Radon 222 limits proposed for drinking water and air differ widely among countries and organizations that have their own legislation. The United States sets limit of 11.1 Bq/L for the presence of Radon 222 in drinking water. In contrast, in the European Union, values from 100 to 1000 Bq/L are used, and intervention cases are considered only for concentrations above 1000 Bq/L (EPA 2009; European Commission 2001). For air, the maximum concentrations established in the international community are also quite different, and may present, in some countries, values four times higher than those recommended by WHO (2007), which is 100 Bq/m³.

Given the above, it was observed that despite the existence of studies on Radon 222 contamination, they are still scarce and related to local realities. So far, no convergences have been observed in relation to the maximum recommended and permitted Radon 222 limits in drinking water and indoor air, and this standardization is still a little explored and discussed issue worldwide, despite its indisputable importance.

Therefore, considering the relevance of the topic and the absence of a worldwide consensus regarding safe Radon 222 concentrations in water and air, this study aims to assess, through a systematic review, the values used by the most referenced worldwide organizations regarding Radon 222 concentrations aiming to propose discussions on the subject and contribute to the convergence of values to be adopted.

2. Research Design

The question that guided this work is: Is there a stipulated standard of radon 222 in the air in the world? The survey was carried out from January to June 2021 and was divided into two stages, the first is a theoretical overview of laws and guidelines of countries and international organizations that establish limits, which can be minimum or maximum, for Radon 222 concentrations in air and water. The second stage evaluated through a systematic review which international organizations and/or local laws would be most cited by published articles, either in full or abstract, being selected here.

According to Castro (2001), a systematic review consists of a planned review with the aim of responding a specific question using explicit and systematic methods, in order to deepen the knowledge regarding an already prepared material, that is, a research with secondary database, which in this case are scientific works.

To obtain eligibility of articles to be evaluated, the PRISMA methodology was applied, which consists of a four-step flowchart (Figure 1) in order to help improve the reporting of systematic reviews. This systematic review was built based on the PRISMA methodological guideline (Preferred Reporting Items for Systematic Reviews and meta-analyses), which consists of a checklist with the aim of helping to improve reporting of systematic reviews (Moher et al. 2015).

The criteria used to select articles for the systematic review were: studies that evaluated Radon 222 concentrations in drinking water; studies that evaluated Radon 222 concentrations in indoor air; studies that cited international laws or guidelines on Radon 222 limits.

All articles selected for this review were published in the last twenty years and indexed in the following databases: Science Direct, PubMed, Taylor and Francis and Springer Link. The keywords used for the systematic review research were: Radon 222 in drinking water; Radon 222 in indoor air; Radon 222 limits in indoor air and drinking water.

The selection of studies was carried out in two stages. First, titles and abstracts of articles were read by the two main reviewers, individually and independently, in order to select studies that, according to inclusion and exclusion criteria, had the potential to respond the guiding question. After reading titles and abstracts, there was a first consensus meeting to assess the inter-examiner agreement, using the kappa statistical test. At that moment, the two researchers tried to reach consensus in the selection of articles, and when there was disagreement, a third evaluator performed the tiebreak. This first step selected articles to be read in full.

The second stage consisted of reading articles in full, aiming to choose those to be included in the systematic review, whose data were extracted. This stage was also guided by eligibility criteria of studies and, at the end, there was a consensus meeting, in which the third evaluator performed the tiebreak when there were divergences between the two main reviewers.

Data extraction was performed in tables collecting data of interest to respond the guiding question. Data collected were the country in which the study was conducted, date of publication and maximum Radon 222 concentrations allowed in air and water at the location where the study was conducted.

Data synthesis was qualitatively performed, with graphs and tables being generated to discuss the results, because, due to the insufficiency or absence of data, it was not possible to perform the meta-analysis.

3. Results

This review selected 2,816 studies, 1,587 of them were repeats. In the first stage, the title and abstract of 1229 articles were read, in which only 35 presented direct approach to Radon 222 in air and water. In the first stage, the Kappa test was performed, specified in Table 1, which obtained result of 0.89, considered excellent assimilation of choice criteria between evaluators.

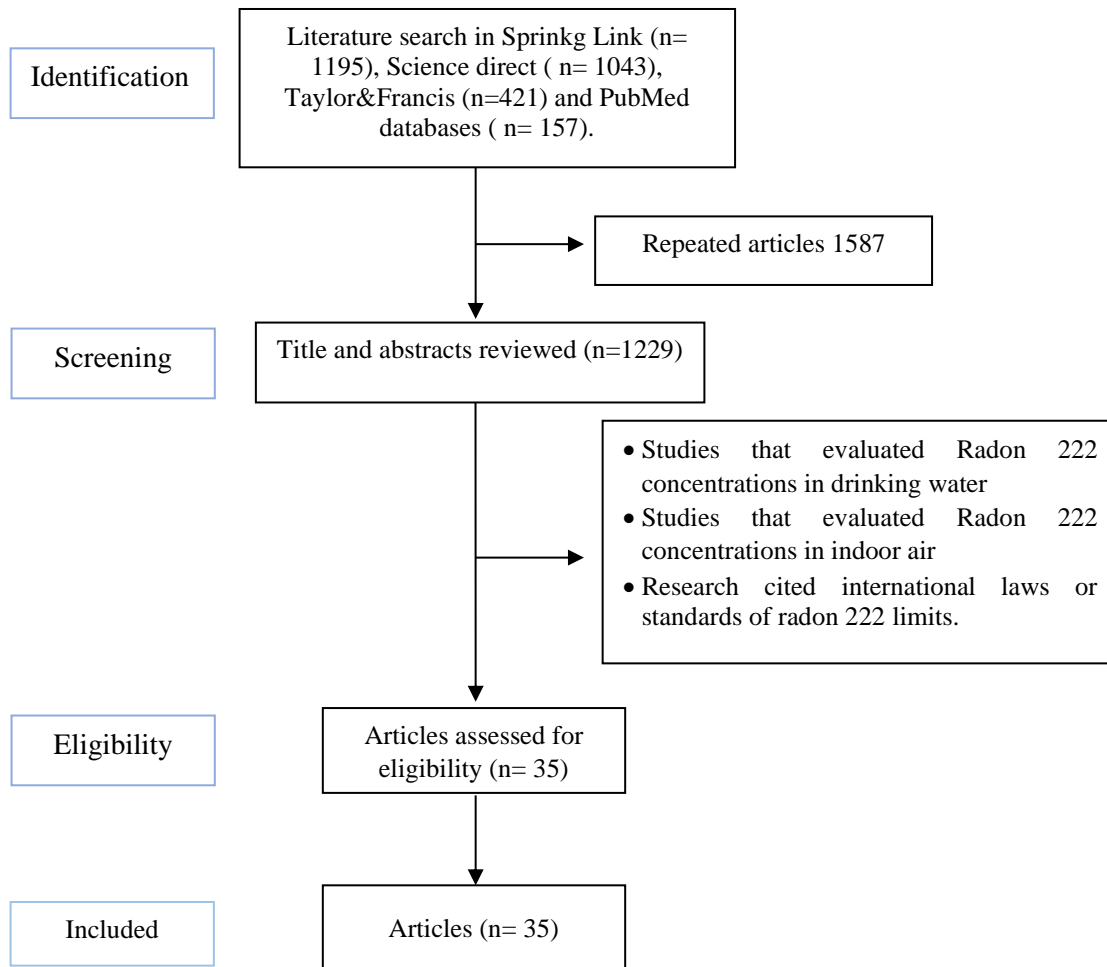
Table 1 - Symmetric measures of the Kappa test.

		Significance			
		Value	Standard Error ^a	Approx. X ^b	Approx. Sig.
Measure of agreement	Kappa	0.893	0.040	31,295	0.000
No. of Valid Cases		1229			

a. Not considering the null hypothesis. b. Use of asymptotic standard error considering the null hypothesis.
Source: Own authorship.

In the second stage, the eligibility criteria were evaluated and all 35 articles were included in the final analysis.

Figure 1 - Flow diagram of study identification according to PRISMA.



Source: Own authorship.

When listing the most cited institutions, organizations that presented restrictive standards for Radon 222 concentrations in water and air were considered. The National Ordinances and Resolutions published by the Federation of each country and cited as comparative limits of results of articles included in this review were also considered as sources of data. The entire graphic part of the review was prepared in Excel 2012 software.

4. Analysis of Global Limits Established for Radon 222 in Air and Drinking Water

In 2003, the World Health Organization (WHO) published a guideline recommending the maximum values for Radon 222 concentrations in drinking water and indoor air, establishing values of 100 Bq/L and 100 Bq/m³, respectively. The value of 100 Bq/L recommended by WHO has been adopted by Italy, France, Germany and the Czech Republic as maximum allowable Radon 222 concentration in drinking water.

Countries such as Austria, Sweden and the United Kingdom, chose to adopt values ranging from 100 Bq/L, indicated by WHO, to maximum limit of 1000 Bq/L. The range from 100 to 1000 Bq/L of Radon 222 in drinking water was established by the European Union (EU) as a guideline for member countries.

However, some EU countries (Norway, Portugal, Spain and Ireland) have established their own limits in internal legislation, being more restrictive as to the maximum concentrations allowed. Portugal and Spain determined intermediate values

compared to those established by EURATOM (European Atomic Energy Community), allowing maximum Radon 222 concentration of 500 Bq/L in drinking water.

In 2009, WHO launched a new Guideline titled Handbook on Indoor Radon - A public health perspective. This handbook does not refer to the already established limits regarding the presence of Radon 222 in drinking water, as it indicates greater relevance in relation to the contamination of this radioactive element in the air, especially when treated indoors (WHO 2009).

The greatest concern with the presence of Radon 222 in the air is based on studies that show the occurrence of lung cancer due to the inhalation of this gas and the ease of calculating the effective Radon 222 dose inhaled by each human being through simple air measurements (Elío et al. 2018; Martínez et al. 2018; US EPA, 2000).

Some other factors also reinforce the greater relevance of the incidence of Radon 222 in the air such as its high volatility, since when present in water, part of this radioactive element can be dissipated into the air through agitation, which can occur during the manipulation of the aqueous medium in daily activities inside households, contributing to increase of Radon 222 concentrations in indoor air (WHO 2009).

There are also scientific reports that demonstrate the increased influence of Radon 222 in indoor air through the contamination of building materials used in masonry finishes, being another cumulative factor of ionizing radiation in the air (Ahmad et al. 2017).

Another argument that reinforces the idea of greater concern with Radon concentration in the air is that despite the fact that the scientific literature associates stomach and gastrointestinal cancer with the ingestion of water contaminated by Radon 222, so far, there are no definitive conclusions that the element is one of the actual causes of this pathology. Unlike ingestion contamination, the occurrence of lung cancer due to Rn²²² gas inhalation has already been found through scientific research (López-Abente et al. 2018; Vogeltanz-Holm et al. 2018; Ha et al. 2017; Vienneau et al. 2017).

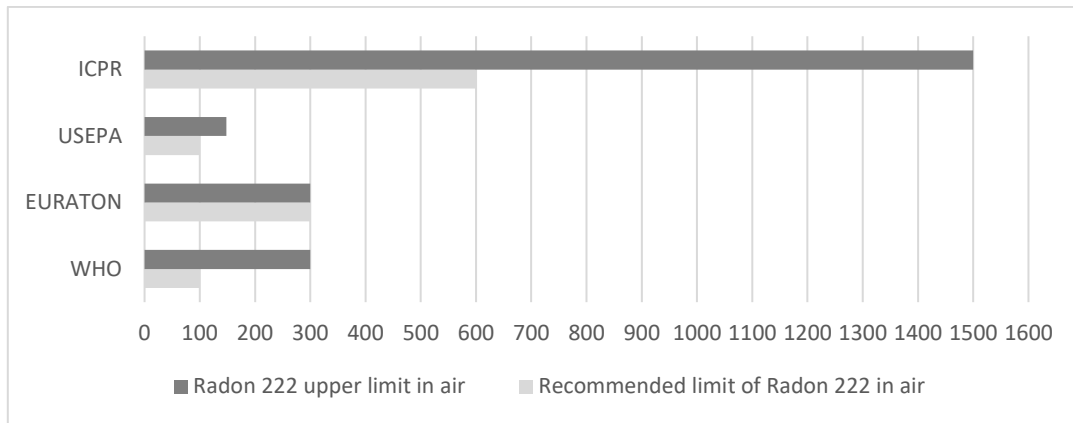
Greater control of the presence of Radon 222 in the air becomes increasingly evident; however, the recommended limit for its presence in the air remained the same established in the 2003 guideline, 100 Bq/m³ (WHO, 2001). Even after the maximum recommended limit established by WHO, several countries with legislation on Radon 222 standards still use ranges of values and not a fixed value for air contamination.

Portugal and France, for example, establish Radon 222 values ranging from 100 Bq/m³ to maximum of 400 Bq/m³ (EURATOM 2000).

The International Commission on Radiological Protection (ICRP), which is also a well-respected organization in the study area, does not limit Radon 222 concentrations in water, but recommends maximum Radon 222 limit of 600 Bq/m³ for indoor environments for prolonged stays and maximum limit of 1500 Bq/m³ for indoor working environments (ICRP 1993, WHO 2001).

Figure 2 shows the reference limit values for Radon 222 in the air indicated by the most cited organizations in scientific studies: ICRP, US EPA (United States Environmental Protection Agency), EURATOM and WHO.

Figure 2 - Limits proposed by world organizations for Radon 222 in the air.



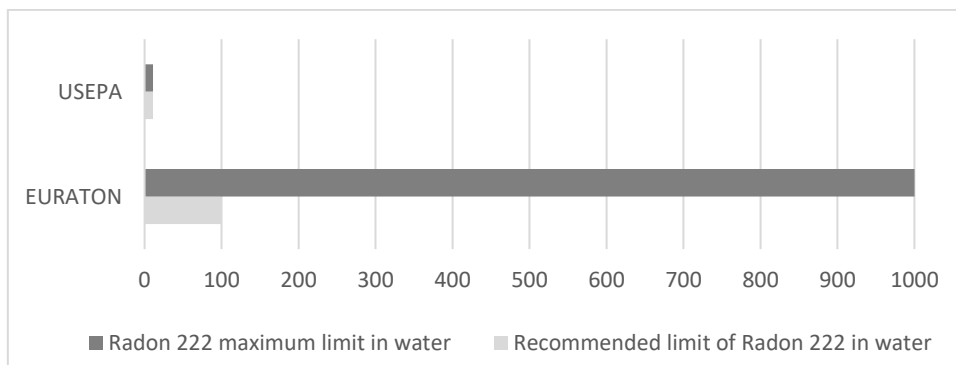
Source: Own authorship.

Comparing the maximum limits established by organizations, there is a 900% discrepancy between ICRP and US EPA values and 400% between values established by ICRP in relation to EURATON and WHO. US EPA sets maximum limit of 150 Bq/m³, half the value established by EUROCON and WHO. These last two organizations agree with maximum limit of 300 Bq/m³; however, they disagree by 200% regarding recommended values.

All divergences pointed out indicate that despite the existence of several guidelines to limit the presence of Radon 222 in the air, there is still lack of knowledge, on the part of organizations worldwide, about the safety and harmfulness limits of this element. This fact highlights the need for more advanced studies on Rn222 concentrations and possible impacts caused by the inhalation of the gas on people's health. New studies will enable to safely and scientifically establish reliable values for maximum and recommended Radon 222 limits.

Regarding the maximum and recommended Radon 222 concentrations in drinking water, only US EPA and EURATOM remain with defined values. However, like the behavior observed in relation to air, the difference between limits proposed by the two organizations is significant, with maximum limit indicated by EURATOM being ninety times greater than that standardized by US EPA (Figure 3).

Figure 3 - Limits proposed by world organizations for Radon 222 in drinking water.



Source: Own authorship.

Table 2 presents a survey of the maximum and recommended Radon 222 concentrations in the aforementioned organizations and in specific legislation in some countries around the world.

The table shows that when comparing countries that establish maximum and recommended Radon 222 limits in air and drinking water, there are also many inconsistencies and lack of standardizations between proposed limits. These divergences generate uncertainties about risks to human health arising from exposure to Radon 222.

Another worrying issue that deserves attention is the absence of specific legislation in certain countries. Areas recognized in literature as more prone to Radon 222 contamination and called High Background Natural Radiation Areas (HBNRAs), such as Poços de Caldas in Brazil, Ramsar in Iran, Orissa and Kerala in India and Yangjiang in China, do not yet have legislation that standardize maximum acceptable Radon 222 concentrations in drinking water and air (EPA, 2000; Sohrabi 2013; Aliyu, 2015). In these countries, only Ordinances that establish maximum concentrations of alpha particles present in the air and drinking water are in force, without specifying the radioactive contaminant element.

Table 2 - Summary of recommended and maximum Radon 222 concentrations in the legislation of countries and organizations.

Organizations/ Countries	Recommended concentration in drinking water (Bq/L)	Maximum concentration in drinking water (Bq/L)	Recommended concentration in indoor air (Bq/m ³)	Maximum concentration in indoor air (Bq/m ³)
WHO	100*	100*	100	300
EURATOM	100	1000	300	300
United States US EPA	11.1	-	100	148
ICRP	-	-	600	1500
Portugal	-	500	100	400
Norway	100	500	200	200
Austria	100	1000	300	400
Switzerland	-	-	140	400
Sweden	100	1000	200	400
United Kingdom	100	1000	200	200
Finland	300	1000	200	400
Belgium			200	400
Italy	100	-	200	200
Greece	-	-	-	400
Spain	-	500	200	-
Czech republic	100	100	-	400
France	100	100	100	400
Germany	100	100	100	200
Ireland	100	500	100	200
Mexico	-	-	-	-
China	-	-	-	-
Brazil	-	-	-	-
Canada	-	-	-	200

*These values were proposed by the Guidelines for Drinking-water Quality. Source: Own authorship.

The tendency to consider the presence of Radon 222 in the air as more relevant, identified in the latest WHO and ICRP publications, especially in indoor and long stay environments, was based on studies assessing the magnitude of the risk of lung cancer in underground mine workers exposed to Radon 222. These studies strongly suggested that this radioactive gas could be the cause of lung cancer in the general population due to exposure confined to households and other buildings (NCRP 1984).

There is also the perspective that Radon 222 concentrations would depend on systematic diurnal and seasonal variations such as weather patterns and behavior of individuals such as the opening of windows inside households (WHO 2001).

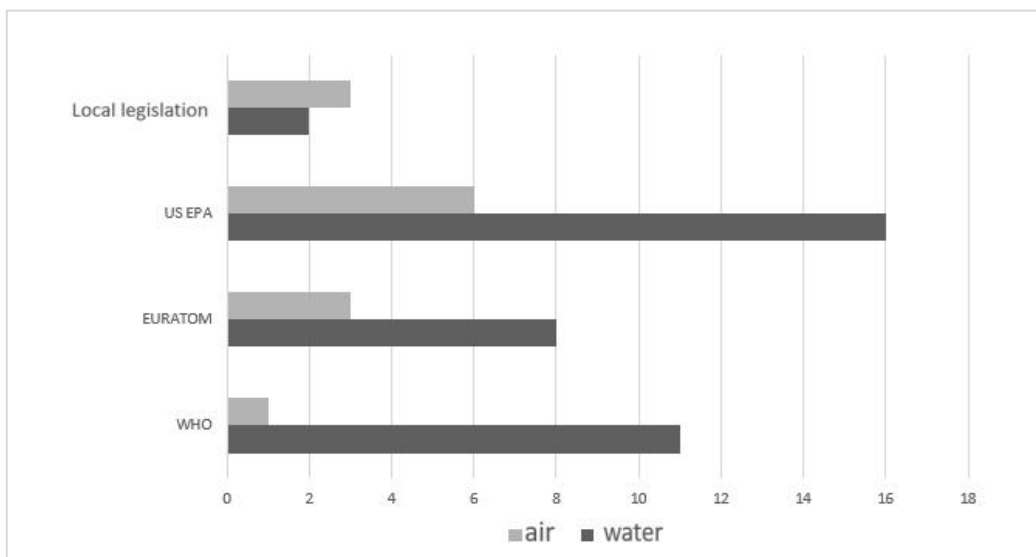
Thus, studies that examined the correlation between mean Radon 222 concentrations and mean lung cancer mortality rates in different geographic areas have been carried out. However, it was found that the usefulness of such studies was limited, as they could not adequately control other risk factors for lung cancer, such as smoking, which causes more cases of lung cancer than Radon 222 in most populations, and the use of ecological studies is controversial, which often provide biased and erroneous estimates of the risk related to Radon 222 (Puskin 2003).

In addition, researchers have found statistically significant association between Radon 222 concentration and lung cancer, even when the analysis was restricted to individuals in households with Radon concentrations below 200 Bq/m³ (WHO 2001). Babai (2012) found that the risk of lung cancer was 20% higher for individuals exposed to Radon 222 concentrations between 100 and 199 Bq/m³ compared to those exposed to Radon 222 concentrations below 100 Bq/m³.

In the scientific community, some limits established for Radon 222 in water and air are more used as reference than others, as shown in Figure 4. According to the figure, the most cited regulation among articles evaluated was US EPA, organization that limits Radon 222 concentrations in North America.

Some studies that used US EPA as reference were developed in countries like Portugal and Poland, belonging to the European continent, which have their own regulations. However, the authors chose to use external standards values as reference (Curado *et al.* 2017; Bem *et al.* 2014). In some cases, there was no report of the existence of a national standard or its use as a comparative element of results obtained in the research (Inácio *et al.* 2017). As US EPA uses more restrictive values regarding the presence of Radon 222 in air and water, it is believed that the more frequent use of values proposed by it in the scientific community is an attempt to obtain greater restriction of Radon 222 limits in countries with less restrictive regulations and increase the safety of the population exposed to the radioactive element (Figure 4).

Figure 4 - Most cited organizations regarding Radon 222 limits in water and air in the evaluated articles.



Source: Own authorship.

Regarding scientific studies of Radon 222 in water, after US EPA, limits established by WHO were the most cited in manuscripts. The same behavior was not observed in journals that dealt with the presence of Radon 222 in air, in this case, the values indicated by WHO were the least cited.

EURATOM obtained expressive citations regarding the concentration limits for water, mainly in studies carried out in countries outside its economic bloc, such as Nigeria, India, Canada, Mexico and Venezuela (Oni *et al.* 2014; Duggal *et al.* 2019;

Blanco-Novoa et al. 2018; Espinosa et al. 2008; HorvaHth. 2000). However, it obtained lower number of citations in articles focused on Radon 222 concentrations in air, equaling those described as local legislations.

ICRP was not mentioned in Figure 4 because it did not propose Radon 222 limits for the aqueous medium, as shown in Table 1.

Of the 35 articles evaluated, 15 manuscripts (42.8%) cited the limit of a single organization as a comparative factor of their results and 14% of the evaluated sample cited three or more than three organizations as comparative limits.

5. Conclusions

According to what was discussed, it could be concluded that:

- The lack of standardization of recommended and maximum allowed Radon 222 limits in water and air was evidenced, which can reach, in some cases, up to a 900% difference.
- It was found that many countries, mainly those located in Latin America and the Asian continent, such as Brazil, Mexico, Venezuela, Japan, China, among others, do not have any national regulations for the control of Radon 222 in air and water.
- Greater concern for the presence of Radon 222 in air, when compared to the presence in aqueous media, was demonstrated by WHO and ICRP guidelines, which only illustrated limits imposed in air.
- The most relevant theoretical basis of Radon 222 in air was evidenced, as it is the sum of gases released from physical media, such as the emission of gases due water agitation, the occurrence in construction materials and release that naturally occurs in contaminated soil.
- The limits established by US EPA for Radon 222 concentrations in water and air were the most referenced in the scientific community, probably due to its more restrictive values when compared to other organizations.
- It was found that some countries and economic blocs, such as the EU, despite having their own regulations regarding Radon 222 limits, often do not use their reference values as a comparative method in scientific studies, probably because their higher limits are very discrepant.
- About 80% of the articles evaluated cited at least two international organizations, comparing the results obtained with two Radon 222 limits for water or air.

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