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Water Radioactive Pollution and Related Environmental Aspects[#]

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Abstract: Radioactivity present in surface continental waters is mainly due to the presence of radioactive elements in the earth's crust. Other artificial radionuclides have appeared due to such human activities as nuclear power plants, nuclear weapons testing and manufacture and use of radioactive sources. There are two sources of radioactive contamination in drinking water. The first is naturally occurring radionuclides that are contained in the soil that water moves through. Some areas are susceptible to contamination from phosphate rich soils and rock. The second source of radioactive contamination comes from man-made sources. Radionuclides found in drinking water are members of three radioactive series, uranium, thorium, and actinium and include the naturally occurring elements radium, uranium, and the radioactive gas radon. These contaminants may cause different types of biological damage. Radium concentrates in the bones and can cause cancers. Uranium can cause cancers in the bones and can have a toxic effect on kidneys.

1. Introduction

Our world is radioactive and has been since it was created. Over 60 radionuclides (radioactive elements) can be found in nature, and they can be placed in three general categories:

1. Primordial - from before the creation of the Earth
2. Cosmogenic - formed as a result of cosmic ray interactions
3. Human produced - enhanced or formed due to human actions (minor amounts compared to natural)

Radionuclides are found naturally in air, water and soil. They are even found in us, being that we are products of our environment. Every day, we ingest and inhale radionuclides in our air and food and the water. Natural radioactivity is common in the rocks and soil that makes up our planet, in water and oceans, and in our building materials and homes. There is nowhere on Earth that you can not find Natural Radioactivity. Radioactive elements are often called radioactive isotopes or radionuclides or just nuclides. There are over 1,500 different radioactive nuclides.

2. Radioactivity in water

Radioactive substances in ground water, such as radium, uranium and thorium, occur naturally. They are present at least to some extent in almost all rocks and radium, in particular, dissolves more readily into ground water in contact with sands or soils. The acidity of the water, which may be increased by the presence of elevated levels of nitrates associated with agricultural land use, is believed to increase the amount of radium that dissolves into ground water from contact with sands and soils.

Radioactivity in drinking water is not a new phenomenon, having been present to some extent for thousands of years. Nevertheless, exposure to radium over a long period of time is believed to increase one's lifetime risk of developing certain types of cancer. Therefore, homeowners should be aware of the steps they might wish to take to test their private drinking water wells for radioactivity and to reduce their exposure.

Radioactive substances are those that are unstable in nature. Radioactive types of uranium, thorium and radium emit radiation to reach a more stable condition.

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This process is called radioactive decay. By measuring the type of radiation emitted, the specific energy level or levels of radiation and the precise rate of decay, scientists are able to identify a radioactive substance and determine how much of it is present.

Radioactivity is usually measured in Curies or in Becquerels. The unit typically used to describe the concentration of radioactivity present in drinking water is the picocurie per liter – one trillionth of a curie.

Initial water testing to determine the presence of radioactivity measures gross alpha activity. Alpha radiation is a type of particle emitted through the decay of certain radioactive substances. Additional testing may be required to measure the presence of specific types of radium, including Radium 226 and Radium 228. An improved testing procedure for gross alpha activity also will reflect the presence of Radium 224, a shortlived radioactive material. Until recently, it was not thought to be a problem, and there is no state standard for Radium 224. However, recent studies have shown Radium 224 to be present and to account for a number of instances of elevated levels of gross alpha activity.

3. Risks connected with radioactivity in water

The U.S. Environmental Protection Agency (EPA) has estimated that the additional lifetime risk associated with drinking water that contains the maximum contaminant level for gross alpha or combined Radium 226 and 228 is about 1 in 10,000. This means that if 10,000 persons were to consume two litters of this water per day for 70 years, we would expect to see one additional fatal cancer in the 10,000 people exposed.

Increased risk of bone cancers has been associated with the ingestion of Radium 224 and 228, and bone cancers and cancers of the head sinuses with Radium 226. This increased risk should be viewed in the context of current cancer statistics.

According to the American Cancer Society, approximately 4,300 people in 10,000 will develop cancer at some point in their lifetimes and approximately 2,200 of the 10,000 eventually will die of cancer. Consuming drinking water containing radioactivity at the maximum level permitted under the Safe Drinking Water Act will increase the number of deaths in the 10,000 individuals so exposed by one.

A gross alpha test is the first step in determining the level of radioactivity in drinking water. This test serves as a preliminary screening device and determines whether additional testing is advisable. A gross alpha test typically costs from $75 to $150. If the level is above 5 picocuries per litter, then further testing for Radium 226 and 228 is recommended. Testing for Radium 226 and 228, if indicated, will cost an additional $100 to $200. A typical water softener costs between $400 and $600 and offers other benefits besides reducing radium levels.

4. Radionuclide Contamination of drinking water

There are two sources of radioactive contamination in drinking water. The first is naturally occurring radionuclides that are contained in the soil that water moves through. Some areas are susceptible to contamination from phosphate rich soils and rock. The second source of radioactive contamination comes from man-made sources.

Radionuclides found in drinking water are members of three radioactive series, uranium, thorium, and actinium and include the naturally occurring elements radium, uranium, and the radioactive gas radon. These contaminants may cause different types of biological damage. Radium concentrates in the bones and can cause cancers. Uranium can cause cancers in the bones and can have a toxic effect on kidneys.

5. The Radioactivity Standards for Water: the US example and a comparison

5.1. Naturally occurring radionuclides:

- Combined radium-226 and radium-228: The Maximum Contaminant Level is 5 picoCuries (pCi) per Liter (Note that 1 Curie = $3.7\ 10^{10}$ Bq).
- Gross alpha particle activity including radium-226 but excluding radon and uranium: The Maximum Contaminant Level is 15 picoCuries per Liter

5.2. Man-made radionuclides

- The average annual concentration of beta particle and photon radioactivity from man-made radionuclides in drinking water shall not produce a total annual exposure greater than 4 milliRem/year (Note that 1 Rem = 0.01 Sv)..
- Except for those radionuclides listed below, the concentration of radionuclides in paragraph (a) above shall be calculated on the basis of a 2 liter per day drinking water intake using the 168-hour data listed in "Maximum Permissible Body Burdens and Maximum Permissible Concentration of Radionuclides in Air or Water for Occupational Exposure," NBS Handbook 69 as amended August 1963, U.S. Department of Commerce.
- Tritium in the total body - 20,000 pCi/L
- Strontium-90 in the bone marrow 8 pCi/L

Average Annual Concentration Assumed to Produce an Exposure of 4 milliRem/year:

5.3 Typical water quality criteria: a comparison

Criteria that are generally used for drinking water quality assessments can be expressed in terms of any of the following quantities:

- Dose
- Activity concentration
- Chemical concentration
- Qualitative health risk e.g. stated as increased morbidity or cancer risk
- Quantitative health risk e.g. fatality risk per year

A quantitative health risk for exposure to radioactivity can be estimated by using a nominal value of 5% per Sievert. This value is for mortality from cancer after exposure to low doses for a population of all ages and as recommended by the ICRP. Inclusion in the water quality guideline of a quantitative risk criterion expressed as a statistical mortality risk is, however, not recommended. The public generally finds it confusing when risk is expressed in statistical terms. Factors, for example, that result in increased public concern in health risk matters include the following:

- Unfamiliarity with a certain type of risk
- Mechanisms or process of how a risk arises are not understood
- Risk is scientifically uncertain and expressed in statistical terms
- Delayed effects are part of the risk, for example when a health effect only manifest itself some time after exposure to a hazardous material

These factors are all elements of ionising radiation health risks. Communication with the general public on water quality in terms of radioactivity and radiation hazards, should preferably not include unfamiliar terms such as sieverts, becquerels and statistical fatality risk. However, the method of communication must still allow a clear grasp of the radiation health hazard associated with a specific source of water.

The American Health Physics Society also recommends against quantitative estimation of radiation health risk below an individual dose of 50 mSv per year, additional to background radiation. The reason given is that there exists no conclusive evidence of health risks for low dose rate annual doses up to 50 mSv . Statistically significant risks for solid tumors in the LSS cohort (Life Span Study cohort of the Japanese A-bomb survivors) are presently seen only above a dose of 100 mSv (short-term low-energy transfer doses).

It is essential that when preparing a guideline, other international guidelines be considered, especially for those countries with which important trade links exist, for example the United States and the European Community.

The criteria used by the USA and the European Union are compared in the table that follows.

The German Commission for radiation protection, "Strahlenschutz-Kommission" (SSK), gave some recommendations on the basis of practicability to evaluate natural radioactivity in water. The relative mobility of the ions of the primordial nuclides in water is stated to be in the order $U^{6+} > U^{4+} >> Th^{4+}$. As a consequence of these different mobilities the Th-isotopes are ignored as constituents of aqueous systems. The same is mostly true for their radium progeny. Ra-226 as progeny of U-238 is more relevant than Ra-228 in the Th-232 decay chain.

The SSK has also recommended an additional annual dose of 0.500 mSv as limit from tap water where water resources are affected by former uranium mining activities.

Table 1. US and EU Guidelines

Regulatory Authority	Chemical concentration	Activity concentration	Radiation dose	Quantitative health risk	Qualitative health risk	Comments
US EPA	Uranium: 30 µg/L	Alpha particles: 555 mBq/L Combined Ra-226 and Ra-228: 185 mBq/L	Beta particles and photon emitters: 0.040 mSv/a	–	Levels stated for MCL (Maximum Contaminant Level), the highest level of a contaminant that is allowed in drinking water	Standards apply to public water distribution networks.
European Union	–	–	0.100 mSv/a	–	–	Radiation dose is the indicator parameter for water intended for human consumption.

The SSK further defines relevant natural nuclides in tap water to be U-238, U-234, Ra-226, Rn-222, Pb-210 and Po-210. No Th-232 and U-235 progeny is included.

In South Africa domestic water refers to all uses to which water can be put in the domestic environment (drinking, food preparation, bathing, washing dishes, laundry and gardening). The EU uses the concept of "water intended for human consumption" and is defined as follows:

- "all water either in its original state or after treatment, intended for drinking, cooking, food preparation or other domestic purposes, regardless of its origin and whether it is supplied from a distribution network, from a tanker, or in bottles or containers;
- "all water used in any food-production undertaking for the manufacture, processing, preservation or marketing of products or substances intended for human consumption unless the competent national authorities are satisfied that the quality of the water cannot affect the wholeness of the foodstuff in its finished form."

The EU Directive also states that the directive does not apply, or a member state is exempt, when "water intended for human consumption from an individual supply providing less than 10 m3 a day as an average or serving fewer than 50 persons, unless the water is supplied as part of a commercial or public activity."

The EU Directive value of 0.100 mSv/a is described as an indicator parameter and the value has been selected to ensure that water intended for human consumption can be consumed safely on a lifelong basis. The same value of 0.100 mSv/a is described by WHO as a screening value.

UNSCEAR provides information on nuclides in drinking water for different regions and countries in the world. The information in Table hereunder indicates that most countries report U-238 and/or Ra-226. It is deduced that mostly these two nuclides are used for purposes of screening water for its radioactivity content.

The wide ranges that are reported for some countries stem from the fact that the data reflect all kinds of water sources, e.g. ground water, lakes and treated water.

6. Water treatment and its effect on NORM concentrations

Water treatment processes that are part of large water distribution networks, for example: in towns and cities, cause a significant decrease in NORM when compared to the concentrations in the raw feed water. These processes include aeration, flocculation, sedimentation, pH-adaption and filtration. It is therefore only necessary to perform comprehensive nuclide analysis in the initial phase of monitoring. But once typical nuclide concentrations have been established, it could be justified that routine monitoring comprise of only a few indicator nuclides.

Table 2. International Drinking Water Radioactivity Concentrations; mBq/L (Note: Empty cells in the table indicate that no values are reported.)

Country	U-238		Th-230		Ra-226		Pb-210		Po-210		Th-232		Ra-228		Th-228		U-235	
	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max
U.S.A.	0.3	77		0.1	0.4	1.8	0.1	1.5				0.05	0	0.05				0.04
China	0.1	700			0.2	120					0.04	12						
India	0.09	1.5																
Finland	0.5	150000			10	49000	0.2	21000	0.2	7600			18	570				
France	4.4	930			7	700					0	4.2						
Germany	0.4	600			1	1800	0.1	200	0.1	200								
Italy	0.5	130			0.2	1200												
Poland		7.3		1.4	1.7	4.5		1.6		0.5	0.06							
Romania	0.4	37			0.7	21	7	44	7	44	0.04	9.3						
Switzerland	0	1000			0	1500							0	200			0	50
Spain	3.7	4.4			20	4000												
U.K.					0	180	40	200										

Remark: The wide range in water activities for some EU countries, e.g. Finland, and some low minimum values should be noted.

Another process that efficiently removes heavy metals and therefore also uranium, lead, polonium and radium, is flocculation. The nuclides are co-precipitated with other unwanted constituents and most of the radioactivity reports to the sludge that is generated by water treatment facilities

Assessments of health effects of pollutants generally use models in which exposure variables and model parameters are point values, often chosen as conservative estimates. A more realistic approach is to characterise the uncertainty of each variable and parameter explicitly as a probability distribution. Quantification of uncertainty should be an integral part of the estimation of annual doses, especially where high doses may exist.

7. Radon and other pollutants in water: a comparison

Underground water always contains a certain radon amount, while the radon concentration in surface water is negligible. Radon enters into water from rocks that contain uranium and radium, and radon enters together with water into houses. Radon releases to the atmosphere if you use water in houses (during showering and washing about 50 percent, and during cooking and washing nearly 100 percent); and short-lived radon daughters are also generated. Their inhalation contributes to the exposure to the population. Nevertheless, from the standpoint of exposure, drinking water is less significant.

The measurement of the radon concentration in water is carried out in a laboratory. The samples of water with a volume of about few tenths litres are taken into the special containers that are sealed to prevent radon release, and the measurement should be performed within four days after sampling. The measurement of the radon concentration in water is classified as the most significant performance from the standpoint of radiation protection

In addition to radon, the other natural radionuclides are present in water; however, in lower concentrations. These are mainly radium (Ra-226) and uranium isotopes (U-234 and U-238). They enter water in the same way as radon, and drinking water that contains such nuclides will also cause a certain exposure to the population.

The determination of individual natural radionuclides (about some tens of radionuclides) in water is time-consuming and expensive work. Hence, two group indicators were specified, i.e., gross alpha counting and gross beta counting that characterise the content of natural radionuclides (other than radon) in water. The guidance level of 0.2 Bq/l for the gross alpha counting and the guidance level of 0.5 Bq/l for gross beta counting may be fixed for water that is supplied to the public water mains. If any of these values are exceeded, it is useful to determine the concentration of individual radionuclides in water (and legal regulations require such measurements).

The gross alpha/beta counting and the measurement of the concentrations of the individual natural radionuclides in water are done in a laboratory. The samples of water with a volume of some litres are taken into plastic containers, and immediately after sampling, nitric acid is added. The measurement of natural radionuclide concentrations is classified as the most significant performance from the standpoint of radiation protection.

The removal of radon and uranium off water is a time consuming and expensive matter, and the problems are also with the existing radioactive waste disposal. The most suitable solution is to replace this water source.

As for radon, usually regulations do not require counting the gross alpha/beta in drinking water for individual water supplies, and no limit is specified either for the gross alpha/beta counting. There are only the guidance levels, *i.e.*, 2 Bq/l for gross alpha and 5 Bq/l for gross beta. If the guidance levels are exceeded, it is recommended that water be used only as service water.

8. Conclusions

Among different pollution causes for water, we find radioactive pollution, the most undetectable one.
Water quality criteria must take into account radioactivity of water too.
Radioactive water pollution may be due to natural or artificial radionuclides.
Among the natural ones, the main hazard source is uranium decay chain, and in particular Radon.
Among the artificial ones, Chernobyl fallout is the main cause, however local sources may occur: nuclear submarines presence must be monitored.

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