

Investigation and Treatment of Natural Radioactivity in Large-Scale Sandstone Aquifer Systems

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Abstract: In many parts of the world, major regional drinking water resources are represented by large-scale sandstone aquifers. In particular in arid climate zones, society depends on the water from such aquifer systems because of the lack of alternative renewable drinking water resources. The often fossil (and hence non-renewable) groundwaters of deep sandstone aquifer systems are not likely to show any anthropogenic contamination. However, sandstone tends to contain elevated concentrations of naturally occurring radionuclides, which has potential impact on the water quality with regard to the groundwater use, giving rise to health related issues. The activity concentration of a certain radionuclide in groundwater depends on (1) the occurrence of the radionuclide or its parent nuclides in the aquifer matrix, (2) the intensity of element mobilization from the mineral grains and (3) the potential ways of natural element removal from the groundwater. As a result, the overall radionuclide concentration in groundwater is determined by the mineralogical composition of the aquifer matrix and by the existing geochemical/hydrochemical conditions, which might change with time as a result of groundwater mining (e.g. due to overexploitation). If elevated radionuclide concentrations in a groundwater sample have to be suspected, an undifferentiated analysis and assessment of all potentially present radionuclides is not appropriate. Whereas some radionuclide species represent a disproportionately high risk and have to be taken into account, most radionuclides do not have to be considered from a radio-ecological point of view. That is due to the poor solubility of some elements, due to the very short half-lives of many of the radionuclides and due to very low natural concentrations of many radionuclide species. Hence, designing a smart approach for the assessment and handling of elevated radionuclide activities in groundwater is compulsory. Such an approach must consider smart ways of (1) localizing and assessing the contamination, (2) careful groundwater mining, (3) state-of-the-art water treatment and (4) after-treatment NORM disposal. Especially the smart treatment of waters containing radionuclides is crucial to maximize the usable fraction of the resources. Typical removal of radionuclides employs ion exchange, reverse osmosis, as well as precipitation with hydroxides, carbonates, or sulphides. However, selective sorbents specifically tailored for radionuclide removal might represent a cost effective alternative to these well established techniques.

Key words: Groundwater radioactivity · Sandstone aquifers

INTRODUCTION

In many parts of the world, major groundwater resources that are used for drinking water production and/or irrigation purposes are represented by large-scale

sandstone aquifer systems. While some of these aquifers are regularly recharged, others, especially those in arid and hyper-arid climates, do not receive significant recharge and have thus to be considered non-renewable or “fossil” resources.

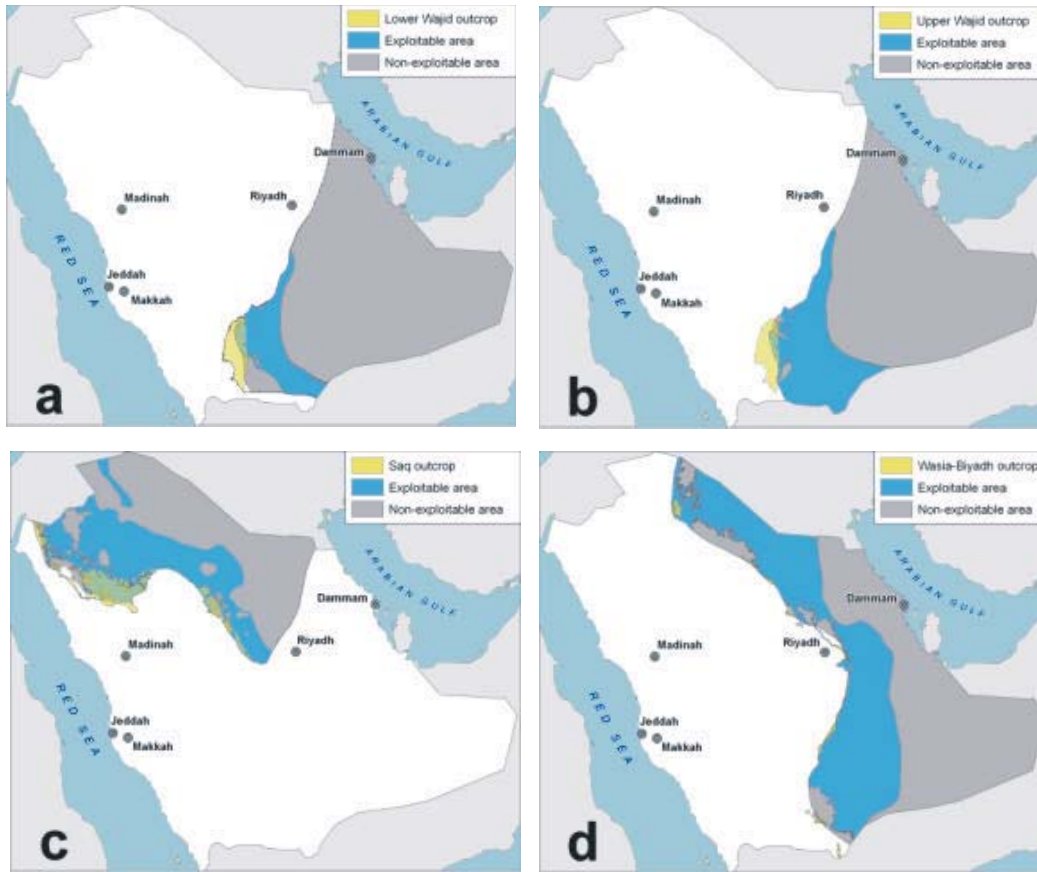


Fig. 1: Maps showing the outcrop and exploitable as well as non-exploitable areas of the major Saudi Arabian sandstone aquifers Lower Wajid (a), Upper Wajid (b), Saq (c) and Wasia-Biyadh (d). The assessment of the exploitability is based on criteria such as water salinity (<2,000 mg/l), drilling depth (<2,000 m) and pumping height (<300 m)

An outstanding example for a *non-fossil*, unconfined large-scale sandstone aquifer is the Triassic/Jurassic Guarani Aquifer, which is, as one of the largest single groundwater bodies in the world, geographically shared by Argentina, Brazil, Paraguay and Uruguay. It covers an area of about 1.2 million km² and supplies some 15 million people [1, 2]. An example for a major fossil water resource is the so called "Nubian Sandstone Aquifer System". It represents the world's largest *fossil* water resource and contains groundwater with ages of up to one million years [3, 4]. It is covering about 2.2 million km² of land and is shared by north-western Sudan, north-eastern Chad, south-eastern Libya and most of Egypt.

Also the sandstones occurring on the Arabian Peninsula host several large-scale aquifers containing big amounts of non-renewable water resources, namely the Lower Wajid and Upper Wajid aquifer in the south-east, the Saq aquifer in the north and the Wasia-Biyadh aquifer in the east of the peninsula (Figure 1). Together, these

aquifers cover more than 2.4 million km². Nevertheless, it has to be kept in mind that parts of the aquifers are not exploitable, either due to significant salinities (threshold value 2,000 mg/l) or due to technical/economical limitations such as drilling depth and pumping height (threshold values of 2,000 m and 300 m, respectively).

Whereas renewable water resources bound to unconfined aquifers are generally vulnerable to anthropogenic contamination, fossil groundwaters are not likely to exhibit any manmade pollution. However, the presence of certain naturally occurring components may prove disadvantageous or even harmful if fossil waters are used for drinking and irrigation purposes. That puts substantial pressure on the authorities that are in charge of groundwater quality assessment and management, particularly in arid regions where, due to the lack of renewable drinking water resources, society depends on the non-renewable water resources.

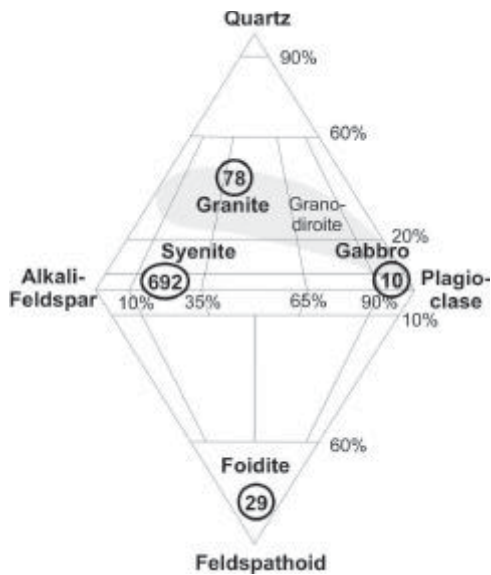


Fig. 2: QAPF Diagram with area of major rock types shaded in gray; encircled figures represent typical ²²⁶Ra activity concentrations [Bq/kg]

A common and widely discussed problem in this regard is the presence of elevated salinities. However, elevated salinities are easily detectable and do not pose any threat to the health of the consumer. A much more critical groundwater constituent that might cause problems in sandstone aquifers is natural radioactivity. Due to the generally considerable size of the aquifer domains that are potentially affected, a key issue for successful groundwater quality assessment and management is the implementation of an efficient methodology that allows straightforward investigation of natural radioactivity in large-scale sandstone aquifer systems. Such “screening” approach should be based on easily attainable data that allow surveying a large number of monitoring wells at limited amounts of time and effort.

The paper presents an efficient methodology for localization and preliminary evaluation of natural radioactivity in groundwater, focussing on the radionuclides that are most critical for human health.

Radioactivity in Sandstone Derived Groundwater:

The activity concentration of a certain radionuclide in a certain groundwater depends on (1) the occurrence of the radionuclide or its parent nuclides in the aquifer matrix, (2) the intensity of nuclide (i.e. element) mobilization from the aquifer matrix and (3) the potential ways of natural element removal from the groundwater

by adsorption and/or precipitation. Thus, the overall radionuclide concentration in groundwater is governed by the mineralogical composition of the aquifer matrix and by the geochemical/hydrochemical conditions present.

Mineralogical Composition: Due to its composition, sandstone tends to contain elevated activity concentrations of naturally occurring radionuclides. Although sandstone does not have a defined mineral composition, quartz can be named its major constituent. Thus, sandstone can generally be characterized as weathering derivative of quartz-dominated rocks (mainly plutonites). On the Earth’s crust these are mainly represented by granites.

In Figure 2, the major rock types on the Earth crust (Gabbro/Diorite, ca. 42 %; Granodiorite, ca. 11%; Granite, ca. 10 %; Matthes, [5]) are plotted in a Quartz–Alkali-Feldspar–Plagioclase–Feldspathoid Diagram (QAPF Diagram). The area representative for the major crustal rock types is roughly shaded in gray. Two rock types with lesser frequency of crustal occurrence are plotted as well (Syenite, ca. 0.4 % and Foidite, 0.2 %; Matthes, [5]).

Also shown in Figure 2 are the radium activity concentrations (²²⁶Ra) that are typical for the rock types displayed. ²²⁶Ra is part of the ²³⁸U decay chain and can thus stand as robust proxy for the activity concentration present in a certain rock type in general.

It can be seen that among the major rock types, granites generally show high (radium) activity concentrations. Higher activities are normally only found in rare rock types rich in alkali-feldspar (such as Syenites and Phonolites, their vulcanitic equivalent), which do, however, not contribute considerably to the development of major sandstone (or any other) aquifer formations. It has to be pointed out that the concentrations given in Figure 2 represent mean values of wide concentration ranges. For granites, for instance, radium activity concentrations between 30 and 500 Bq/kg are known, concentrations for gabbros range between 5 and 50 Bq/kg.

The elevated abundance of natural radionuclides in more differentiated rock types such as granites (or syenites) is due to the high natural valences of uranium and thorium, which head the three natural decay chains. Since sandstone can in general be characterized as granite derivative, the elevated activity concentrations found in granites are a typical feature of sandstones as well.

Geochemical/hydrochemical Conditions: Whereas the mineralogical composition of an aquifer matrix is a given and invariable condition, the hydrochemical environment in an aquifer may change with time, for instance due to the aeration of a formerly anoxic aquifer section as a result of extensive groundwater withdrawal. Therefore, in particular overexploitation of a groundwater reservoir might result in considerably increased element mobility in the aquifer.

As it will be pointed out in more detail later on, uranium and radium are elements with high (radio) toxic relevance. Thus, their mobilization behaviour in aquifers shall be briefly summarized in the following.

Uranium is very mobile in its hexavalent form. Under circum-neutral conditions, it forms soluble complexes primarily in the presence of carbonate, phosphate, or chloride. At lower pH, it tends to form soluble complexes in the presence fluorides and sulphate. Thus, in saline groundwaters under oxidizing conditions the affinity of hexavalent uranium to form soluble complexes results in a generally high uranium mobility. In its tetravalent form, i.e. under reducing conditions, uranium is stable and precipitates as poorly soluble $U(OH)_4$ or UO_2 (uraninite). Besides that natural element removal from the groundwater by precipitation, uranium can also be subject to (partly reversible) adsorption onto clay minerals, iron or manganese oxides/hydroxides and also carbonates [6].

In nature, radium exists only as bivalent cation. In low salinity solutions, radium occurs as Ra^{2+} with its solubility not being influenced by the RedOx conditions. Complexing to highly insoluble $RaSO_4$ or $RaCO_3$ only occurs, if high concentrations of the respective inorganic ligands are present (e.g. in brines). In such systems, co-precipitation with barium and calcium salts (barite, gypsum, calcite) is also likely. In normal groundwaters, however, it is not solubility but adsorption that exerts a strong control on radium mobility. In groundwaters of low ionic strength, radium can readily be adsorbed onto the surfaces of the aquifer mineral matrix. The intensity of adsorption is strongly dependent upon the aquifer material, the groundwater composition (Eh, pH, other cations in solution) and the water temperature. Increasing salinity generally decreases radium adsorption since the available adsorption sites of the mineral matrix are occupied by the more abundant cations [6, 7].

Radioactivity Levels in Saudi Arabian Sandstone Aquifers: Several studies have demonstrated that also Saudi Arabian sandstone aquifers show elevated radioactivity concentrations. Out of 11 water samples

taken from the Lower Wajid aquifer 10 showed ^{228}Ra activity concentrations exceeding the corresponding WHO [8] guideline level of 100 mBq/l. With respect to the Upper Wajid aquifer, the guideline level was violated in 24 out of 30 samples [9]. Also in the Saq aquifer, ^{228}Ra and, moreover, ^{226}Ra activities tend to be high – among 34 samples taken, 29 and 7 samples exceeded the corresponding suggested threshold values, respectively [10]. Finally, also the Wasia-Biyadh aquifer system seems to be concerned [9].

Health Related Data Evaluation: If elevated radionuclide concentrations in groundwater have to be suspected, an undifferentiated radiochemical analysis and data assessment for all radionuclides that are potentially present in the aquifer/groundwater is not appropriate. For evaluating the quality of a groundwater from the radio-ecological point of view, some radionuclide species have necessarily to be taken into account because they represent a disproportionately high health risk, whilst many others do not have to be considered at all. The latter is due to the very scarce natural occurrence of many radionuclide species, their very short half-life, or due to the very poor solubility of the respective element in water.

For revealing health related problems of a groundwater, choosing the right radionuclides for evaluation is essential. The reason for that is that the total human ingestion dose (H_{ing} [mSv/a]), which is the measure for radiation protection issues, is not a parameter that can be measured but that has to be calculated (the WHO suggests a total human ingestion dose for drinking water of $H_{ing} = 0.1$ mSv/a). As given in Equation 1, H_{ing} is a function of the ingestion rate (U [l/a]) and of the sum of the products of the activity concentrations of all considered radionuclides (c_i [Bq/l]) and the respective Dose Conversion Factor (DCF_i [mSv/Bq]).

$$H_{ing} = U \cdot \sum c_i \cdot DCF_i \quad (1)$$

The DCF combines three parameters specific for each radionuclide: (1) its decay half-life/biologic half-life ratio, (2) its immediate tailing of short lived progeny and (3) the kind of radiation it is emitting. The DCF is increased, if the decay half-life is significantly shorter than the biologic half-life (e.g. ^{210}Po), if the considered radionuclide has many short-lived progeny (e.g. ^{226}Ra) and if it disintegrates via alpha decay (e.g. all Ra and Po species). DCF's have been estimated for all relevant radionuclides and are available in the literature.

Table 1: Dose Conversion Factors for ingestion of radionuclides with radiation protection relevance (for adults >17a, ICRP [11])

Radionuclide	DCF [Sv/Bq]
²³⁴ U	4.9×10^{-8}
²³⁸ U	4.5×10^{-8}
²²⁶ Ra	2.8×10^{-7}
²²⁸ Ra	6.9×10^{-7}
²¹⁰ Po	1.2×10^{-6}
²¹⁰ Pb	6.9×10^{-7}

The ingestion rate depends on the age of the consumer (Table 1) as well as on the geographic location and can easily be assessed. The activity concentration depends, as discussed above, on the mobilization potential and the solubility under the given conditions in the aquifer and can be determined. Thus, the most uncertain parameter in Equation 1 is the choice of radionuclides that are taken into consideration (i) out of the 40 radionuclides being present in the natural decay chains.

For selecting the radionuclides that are relevant for health related issues, their natural abundance, aqueous solubility and half-lives have to be taken into account. By doing that systematically, the number of radionuclides that are relevant can be reduced significantly.

Step 1: The three natural decay chains are headed by the radionuclides ²³⁸U, ²³⁵U and ²³²Th. The nuclides that are comprised by the ²³⁵U chain (13 in total) occur naturally in (from the health perspective) negligible concentrations and do not have to be considered (the natural ²³⁸U/²³⁵U activity ratio amounts to 21.7/1).

Step 2: Of the two remaining decay chains, the isotopic species that belong to the elements thorium and protactinium (5 in total) can be neglected if groundwater issues are discussed since both elements exhibit very poor aqueous solubilities.

Step 3: Most of the remaining radioisotopes show half-lives of less than 5 days (16 in total). Their activity concentrations in groundwater are thus in decay equilibrium with the respective long-lived parent nuclides and do not have to be evaluated individually.

Reducing the number of radionuclides as demonstrated above brings six radionuclides into the focus of investigation: ²³⁴U, ²³⁸U, ²²⁶Ra, ²²⁸Ra, ²¹⁰Po and ²¹⁰Pb. The Dose Conversion Factors for the six radionuclides are given in Table 1.

If the DCF's and the respective mobilities are taken into consideration, it can be concluded that the relevance of the six radionuclides, seen from the radiation protection perspective, decreases in the following order: ²²⁸Ra and ²²⁶Ra (fairly high concentrations in groundwater and high DCF's), ²¹⁰Pb and ²¹⁰Po (low concentrations in groundwater but very high DCF's; Pb is chemo-toxic) and finally ²³⁴U and ²³⁸U (low concentrations in groundwater and fairly low DCF's with U being highly chemo-toxic).

Localization and Preliminary Evaluation Approach:

Radioactivity in spacious sandstone aquifer systems is not a small-scale issue. Thus, successful groundwater management and quality assessment necessitates the design and the implementation of an efficient approach that allows a straightforward large-scale investigation based on easily attainable data that allow surveying a large number of monitoring wells at limited amounts of time and effort.

Easily attainable parameters that give indication of the mobility of elements (and thus of radionuclides) in an aquifer are the standard water parameters Eh, pH, electrical conductivity (EC) and temperature. Whilst the related data allow a rough assessment of element solubilities in an aquifer in general, they do however not reveal any information on the actual presence of elevated activity concentrations.

As discussed above, radium (²²⁶Ra) can stand as robust proxy for the radionuclide concentration present in a certain aquifer domain. The direct progeny of radium is radon (²²²Rn). Due to its short half-life of only 3.8 days, radon can in general be assumed to be in decay equilibrium with the radium available for radon emanation in the aquifer matrix.

Radon exhibits certain characteristics that predestines it for being used as a tracer and radiation indicator: (1) it is the only element in the natural decay chains that appears as a (dissolved) gas, (2) it behaves chemically inert due to its noble gas configuration and thus gets not retarded by any aquifer material and (3) it is, even at low concentrations, easily detectable on site by using mobile radon-in-water detection equipment as described by Schubert *et al.* [12].

Thus, as a first step for a large-scale screening approach aiming at the localization of elevated activity concentrations in an aquifer domain, a radon survey could be appropriate. Elevated radon concentrations in the groundwater indicate qualitatively the presence of

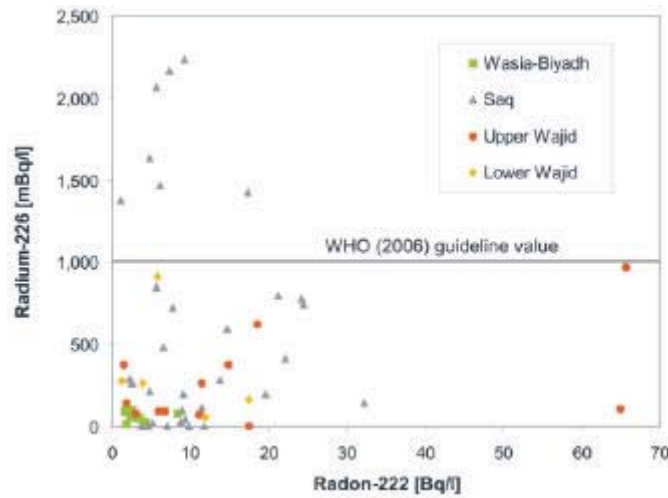


Fig. 3: Diagram showing ^{222}Rn and ^{226}Ra activity concentrations in the Lower and Upper Wajid, Saq and Wasia-Biyadh aquifers; Note the ^{226}Ra guideline violations and the general lack of correlation

elevated concentrations of radium in the aquifer matrix in the neighbourhood of the sampled well. The spatial range of radon signal is dependent on the groundwater flow velocity (and the radon half-life of 3.8 days). Besides the radium activity concentration of the aquifer matrix (A_{Ra}) the radon concentration in the water (C_{Rn}) is also dependent on the porosity (n), the emanation coefficient (ϵ) and the dry density of the aquifer matrix (ρ) (Equation 2), i.e. on parameters that can be considered nearly constant on the small scale in homogeneous aquifers.

$$C_{\text{Rn}} = \frac{A_{\text{Ra}} \cdot \epsilon \cdot \rho}{n} \quad (2)$$

As mentioned above, the radium mobility in the aquifer is, besides the radium occurrence, of vital importance. First indication of the mobility of the indirectly detected radium is given by the Eh, pH, EC and temperature data that are recorded simultaneously.

Own measurements in the Saq (BRGM/ATC, 2008), Lower and Upper Wajid [9] and Wasia-Biyadh aquifers (GTZ/DCo, in progress) revealed that no significant correlation between radon-in-water and radium in solution are found if the radium solubility is not taken into consideration. At wells with significantly high ^{222}Rn concentrations of up to 65 Bq/l (Upper Wajid), relatively low ^{226}Ra levels of only 105 mBq/l were found. On the other hand, at low ^{222}Rn concentrations of only 1 Bq/l (Saq), ^{226}Ra concentrations of approximately 1,400 mBq/l were detected (Figure 3).

The obviously missing correlation between ^{222}Rn and ^{226}Ra in solution is due to the fact that the radon that is present in the groundwater is predominantly produced and emanated by the aquifer matrix and is thus not in decay equilibrium with the radium in solution. Hence, radon activity concentrations in groundwater samples that are detected immediately on-site cannot be used as unequivocal proxy for radium in solution. They only give indication of the radium inventory present in the mineral matrix of the local aquifer. Although the radium concentration in the aquifer matrix is a key parameter for assessing the radiation related groundwater quality, the prevailing hydrochemical conditions, which govern element (i.e. radium) solubility in the aquifer, have also (and in particular) to be taken into account. Future research is needed that focuses on the combined quantitative evaluation of radon-in-water concentrations and the water parameters Eh, pH, EC and temperature for answering questions concerning groundwater related radiation protection.

Radon activity concentrations (^{220}Rn , ^{222}Rn) detected in groundwater samples can, however, be used as proxy for radium in solution (^{224}Ra , ^{226}Ra) after the radon/radium decay equilibrium in the water is reached, i.e. after all radon that is not supported by dissolved radium (“excess radon”) has decayed or degassed. In case of $^{226}\text{Ra}/^{222}\text{Rn}$, dissolved radon will show the same (98.5%) activity concentration as dissolved radium after 23 days (six ^{222}Rn half-lives), in case of $^{224}\text{Ra}/^{220}\text{Rn}$, decay equilibrium is reached after six minutes (six ^{220}Rn half-lives). Commercially available mobile radon detection

equipment allows detection limits that meet the requirements set by the guideline-relevant activity concentrations for radium.

Hence, a suitable approach for uncomplicated radium (^{224}Ra , ^{226}Ra) detection by means of mobile and easy to handle radon detection equipment would be to

- Measure radon on-site (including “excess radon”),
- Take a water sample (e.g. 6 litres), “degas” its radon content (e.g. by means of a bubble stone) and store it in a radon-tight container and.
- Measure the water sample in the container for radon (^{220}Rn , ^{222}Rn) after at least six half-lives of the respective nuclide.

If the ^{232}Th and ^{238}U activity concentrations of the aquifer matrix are known, e.g. from spectral gamma ray borehole loggings or measurements with portable radiation detectors, the ^{226}Ra activity concentration of the groundwater can roughly be estimated based on the ^{226}Ra values detected in the groundwater.

Smart Mining: Several studies have shown that elevated radionuclide concentrations frequently occur in agricultural areas, i.e. in areas where the intensive groundwater withdrawal causes modifications of the flow patterns (e.g. BRGM/ATC, [10]). Two reasons can be assumed for the triggering of radionuclide mobilization due to extensive water withdrawal. One possibility is, as mentioned above, the aeration of formerly anoxic aquifer sections resulting in a significant change of the local hydrochemical environment. Another option is that due to the hydrostatic pressure drop at the base of the aquifer, cross-formational flow can be induced and waters from formations of low hydraulic conductivity with elevated radioactivity levels (due to the long contact times) can be mobilized.

Hence, aquifer sections that are known for elevated radionuclide concentrations in the underlying aquitards should be avoided when drilling wells for public water supply. Furthermore, if new areas are developed, abstraction rates should be kept at a level that allows on the one hand to keep interference with the natural groundwater flow pattern low and on the other hand to distract a maximum amount of water (smart mining). The pump stream should continuously be monitored for changes in the standard water parameters, in particular Eh, pH and EC.

Table 2: Radon, radium and uranium removal efficiencies of common water treatment techniques [13].

Treatment Method	Removal Efficiency [%]		
	Radon	Radium	Uranium
Activated alumina			90
Aeration, packed tower	to 99+		
Aeration, diffused bubble	to 99+		
Aeration, spray	70-95+		
Coagulation/filtration			80-98
GAC adsorption/decay	62-99+		
Electrodialysis		90	
Greensand		25-50	
Hydrous manganese oxide filter		90	
Ion exchange		81-99	90-100
Lime softening		80-92	85-99
Reverse osmosis		90-95+	90-99

However, it is not always possible to completely avoid contaminated waters. In these cases, appropriate treatment techniques have to be applied.

Treatment and NORM Disposal: If elevated radionuclide concentrations lead to a total human ingestion dose exceeding the guideline level of 0.1 mSv/a recommended by the WHO [8], measures have to be taken in order to enable the use of the groundwater for drinking water purposes. The water can either be blended with non-contaminated water or treated utilizing standard treatment techniques that also address radionuclides. Table 2 gives an overview of potential techniques and the respective element-specific removal rates that can be achieved.

If the general water quality is good and radionuclides are the only parameters showing guideline value violations, techniques addressing only specific elements, such as selective sorbents, might be a cost-effective alternative.

When applying water treatment in order to remove radioisotopes, one has to consider that solving this problem produces another one. While radon removal (degassing) is not associated with the production of liquid or solid Naturally Occurring Radioactive Material (NORM) containing waste, the techniques addressing radium or uranium produce such wastes. Examples of NORM waste from municipal water treatment are radioactive contaminated brines from reverse osmosis facilities, spent ion exchange resins, filter sludges and waters from filter backwash.

The radioactivity levels of the resulting contaminated materials depend on the amount of treated water, the radionuclide concentrations of the water and the removal efficiencies – the more specific and efficient the method of removal, the higher the specific activities.

After concentration by evaporation and/or precipitation (liquid wastes) or mechanical dewatering (sludges) these materials can be disposed in landfills or, in case of higher concentrations, in licensed radioactive waste disposal facilities [14]. In case of adsorption based techniques it is, with regard to the classification of the resulting solid NORM waste and the related disposal option, not necessarily advisable to use the chosen sorption material until its maximum sorption capacity of is reached.

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