

Radon-222 content of groundwater in Western Australia

by P. M. Thorpe

Abstract

Radon-222 (radon) is a naturally occurring, inert, radioactive gas of short half-life. It is derived from the decay of radium-226 which in turn originates from uranium-238, both of which are found in trace quantities in all rock-forming minerals. The radon gas released from radium-bearing minerals is either dissolved in groundwater or included within soil air.

There is a potential domestic and occupational health risk associated with the inhalation of radon and its short-lived progeny. However, the extent of the contribution of radon by exsolution from domestic water sources to the radon in air inside houses is uncertain. In Australia the recently proposed draft guideline level for radon in drinking water is 100 Bq/L. A Statewide survey of groundwater radon levels in Water Authority schemes and in other groundwater bores shows that radon levels in untreated groundwater range widely from 0.9 to 233 Bq/L, and average 16.2 Bq/L.

The wide variation in groundwater radon levels is attributed to the type of aquifer. Older, fractured-rock aquifers generally contain higher radon levels than younger sedimentary aquifers with intergranular porosity, which usually contain larger groundwater resources and supply large population centres.

The results of this study suggest that the primary source of radon in groundwater is solid-phase radium-226 contained within aquifer minerals. Radium-226 dissolved in groundwater is unlikely to represent a significant source of radon. This aspect of radon generation is important in that once the radon in the groundwater is separated from the aquifer it will be lost rapidly over a period of days or weeks by both exsolution to air and radioactive decay, and will not regenerate to a significant extent. Specific rock types which have a relatively high radon-producing potential include phosphate nodules that contain uranium, heavy-mineral sands, and fractured igneous rocks. Further research is required to identify aquifer types likely to yield groundwater with high radon levels.

KEYWORDS: Ground water, aquifers, radon-222, radium-226, polonium, uranium, ground water management.

Radon-222, hereafter referred to as radon, is the longest-lived isotope of radon and is a naturally occurring inert gas with a half-life of 3.8 days. It is soluble in water and accumulates in groundwater and soil air. It is exsolved from groundwater to the atmosphere when the confining hydrostatic pressure is released. Radon is not ionized in solution, nor does it precipitate in solid phases; it adsorbs onto organic material to a limited extent, especially charcoal.

There is increasing concern for the potential domestic and occupational health risks associated with the inhalation of radon and its short-lived decay products in confined areas, such as dwellings or underground mine galleries. When radon in air is inhaled into the lungs, the radon decay products, chiefly polonium isotopes, adhere to lung tissue. Radon-derived polonium atoms also adhere electrostatically to particulate matter in air, which, when breathed in, may also deposit on lung tissue. The polonium isotopes emit alpha particles, which can cause lung damage and lung-cancer deaths. It is therefore important to investigate the possible contribution of radon exsolved from public water supplies to the overall exposure of the population to radon gas. The ingestion of radon in drinking water is not considered to be a significant risk to health.

This report summarizes the results of a survey of the radon content of groundwater in Western Australia based upon measurements from Water Authority of Western Australia (WAWA) schemes (Thorpe, 1994), and various other production and observation bores. The survey was undertaken jointly by WAWA and the Geological

Survey of Western Australia (GSWA) in 1992, and follows an earlier study in the Perth Metropolitan Area (Thorpe and Davidson, 1991).

Sources of radon

The parent isotope of radon, radium-226 (half-life of 1620 years), is the fifth member of the uranium-238 decay series and occurs in trace quantities within a variety of rock-forming minerals, in addition to the principal uranium-ore minerals. Uranium and radium are commonly contained in accessory minerals that are common in aquifers, such as monazite, zircon, phosphates, and clay minerals. Uranium is mobile in solution in groundwater under oxidizing conditions, and can be incorporated within secondary mineral deposits. These secondary precipitates coat the surfaces of fractures and mineral grains in a variety of rock types.

Radium-226 is chemically insoluble in water (Michel, 1990); however, its solubility in a groundwater environment is enhanced by the process of alpha recoil. This process involves the recoil effect of the release of an alpha particle from a nucleus, which propels a newly created progeny radio-nuclide with sufficient energy to break chemical bonds. Atoms undergoing radioactive decay on the surface of a mineral grain can eject their progeny directly into the surrounding pore water. In this way, the decay of thorium-230 atoms on mineral-grain surfaces can eject radium-226 atoms into groundwater. This process physically alters the internal crystal structure of minerals containing radioactive isotopes and accelerates their chemical breakdown. This alteration commonly occurs in zircon, which contains traces of uranium and thorium.

Radon is a noble gas and is soluble in water, though not ionized in solution. The solubility of radon in water increases with increasing pressure and decreases with increasing temperature. Numerous studies have shown that radon in groundwater is produced almost entirely from solid-phase radium-226 contained within aquifer minerals, rather than from radium-226 in solution (Wanty and

Gunderson, 1987; Michel, 1990). This is supported by recent work involving detailed radon analysis of groundwater during purge pumping of deep observation bores in the northern Perth Basin (Thorpe, 1994, in prep.).

Below the watertable a state of secular equilibrium develops between the rate of radon production by radium-226 decay and the rate of radon decay. This balance is almost complete after a period of 30 days. The transport of radon by groundwater flow in an aquifer from a zone of high radon production is likely to be minimal because the half-life of radon is short in the unsupported state in terms of rates of groundwater flow. This aspect of the occurrence of radon is important when considering the location of groundwater production bores in areas that contain zones of high radon production.

The physical properties of an aquifer have an important control on the radon content of groundwater. In general, sedimentary-rock aquifers with high intergranular porosity

produce groundwater with lower radon levels than fractured-rock aquifers (Wanty and Gunderson, 1987; Michel, 1990). This difference is attributed to two factors:

- (i) The volumetric ratio between water and rock is substantially lower in fractured-rock aquifers; hence, the radon produced throughout the rock mass is concentrated into a small volume of pore water, producing high radon concentrations.
- (ii) Fractured-rock aquifers, such as granitoids, commonly contain higher concentrations of uranium-bearing minerals (Michel, 1990).

Exceptions to the above are karst limestones and highly lithified sandstones, which behave as fractured-rock aquifers with a low primary permeability and high secondary permeability. Radon levels in karst limestones are likely to be low due to the rapid rate of groundwater flow through the rock mass. This rapid rate of flow, following a high recharge event,

Table 1. Summary of radon-222 concentrations in groundwater in Western Australia

| Region | Type | Number of samples | Radon (Bq/L) | | |
|----------------|--------------------|-------------------|--------------|------------|-------------|
| | | | Average | 1 σ | Range |
| North West | Source | 15 | 35.4 | 58.9 | 1.8 – 233 |
| | Retic. | 13 | 13.8 | 21.2 | <0.8 – 83.9 |
| Mid West | Source | 28 | 24.4 | 40.5 | 0.9 – 220 |
| | Retic. | 22 | 9.8 | 9.7 | 0.7 – 36.3 |
| Central | Source | 11 | 22.7 | 19.7 | 1.6 – 64.7 |
| | Retic. | 9 | 9.4 | 9.1 | 1.2 – 27.0 |
| Goldfields | Source | 4 | 15.6 | 15.4 | 2.8 – 41.8 |
| | Retic. | 4 | 13.3 | 14.9 | 2.5 – 39.1 |
| Perth | Unconfined aquifer | 67 | 8.3 | 10.8 | 1.7 – 81.2 |
| | Confined aquifer | 25 | 11.8 | 5.8 | 4.1 – 31.9 |
| | Retic. | 6 | 1.6 | 0.4 | 0.9 – 2.0 |
| Great Southern | Source | 6 | 31.2 | 18.3 | 3.1 – 53.8 |
| | Retic. | 4 | 3.0 | 1.6 | 1.2 – 5.6 |
| South West | Source | 20 | 14.9 | 7.6 | 5.2 – 38.4 |
| | Retic. | 12 | 5.8 | 7.3 | 1.1 – 24.4 |
| Overall | Source | 176 | 16.2 | 27.0 | 0.9 – 233 |
| | Retic. | 70 | 9.0 | 12.7 | 0.7 – 83.9 |

Retic. : post-treatment sample from the reticulation
1 σ : population standard deviation

may not give sufficient time for secular equilibrium to be achieved between the rock mass and adjacent groundwater.

High radon levels are likely to be found in sedimentary-rock aquifers that contain secondary uranium-rich carbonate cement, heavy minerals, or phosphatic material. Localized high radon-producing zones, associated with uranium mineralization, are likely to be found in all rock types.

Sampling and analysis

Sampling

Groundwater samples for analysis were collected by air-free pumping of bores. The bores were purged of the groundwater standing in the bore casing immediately prior to sampling (otherwise anomalously low radon levels will be recorded; Thorpe, in prep.). Radon gas is rapidly lost by exsolution, so a simple sampling technique was developed to prevent radon loss (Thorpe, 1994). Radon measurements were performed in the GSWA Isotope Hydrogeology Laboratory within two days of field collection and were corrected for radioactive decay to the time of field collection (Thorpe, 1994). About 80% of the samples were retained for three months after the initial analysis for remeasurement to assess whether radon was being regenerated from radium-226 in solution. The measurements showed no detectable increase in activity above that of laboratory background samples. This indicates that most of the radon initially present in the groundwater samples was produced by the decay of solid-phase radium-226 in the aquifer. The remeasurement-emanation technique can only detect radium-226 concentrations greater than about 5 Bq/L. These tests therefore do not indicate that dissolved radium-226 is entirely absent in all of the groundwater sources sampled or that gross alpha radiation and radium-226 activities in the sources are below the draft National Health and Medical Research Council (NHMRC) guideline levels of 0.1 and 0.5 Bq/L respectively.

In the Perth Metropolitan Area, samples were collected from all of the production bores in the confined

aquifers and from a selection of bores within the unconfined aquifer. Post-treatment samples were also collected. In country areas, each aquifer used in a town water supply scheme was sampled. Generally one bore within each aquifer was sampled together with the treated reticulated water. Some rural schemes were not included, although similar aquifer types were sampled elsewhere.

Results

Overall radon concentrations in production and observation bores

(Table 1) from the sampling site locations shown in Figures 1 and 2 vary considerably from 0.9 to 233 Bq/L, and average 16.2 ± 27.0 (1σ) Bq/L (Thorpe, 1994). This variation in concentration is similar to that observed by the major groundwater surveys conducted in the United States of America (Longtin, 1990; Dixon et al., 1991).

Radon levels in groundwater in the confined and unconfined aquifers in the Perth Metropolitan Area (Perth Basin) average 11.8 ± 5.8 and 10.8 ± 8.3 Bq/L respectively and range from 1.7 to 81.2 Bq/L. Radon levels in country-scheme sources average

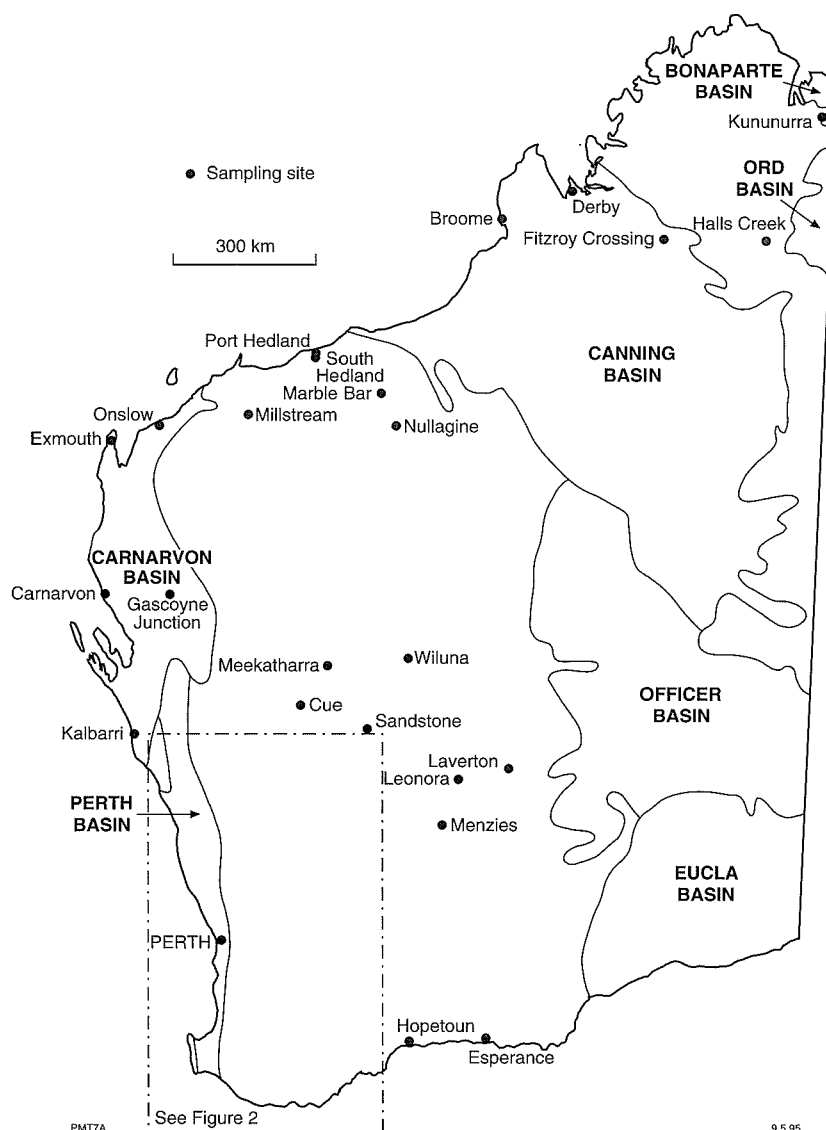


Figure 1. Location of radon sampling sites in Western Australia

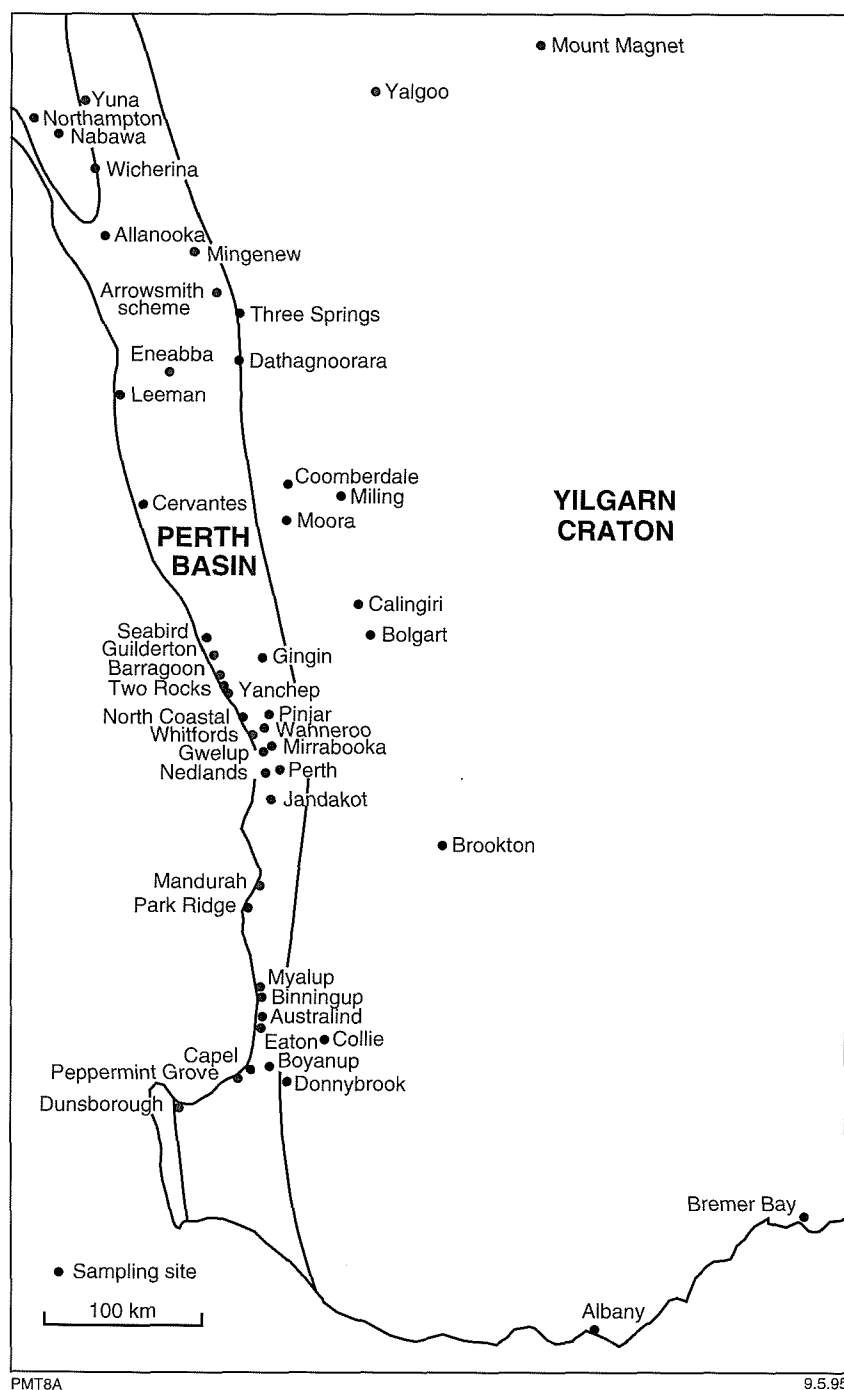


Figure 2. Location of radon sampling sites in southwestern Western Australia

24.0 ± 36.2 Bq/L and range widely from 0.9 Bq/L at Exmouth to 233 Bq/L at Halls Creek.

Country areas

In general, higher groundwater radon levels are associated with fractured-rock aquifers. The

highest radon concentrations were found in bores screened within fractured Proterozoic limestone and sandstone at Halls Creek (233 Bq/L) and fractured granite and dolerite at Northampton (220 Bq/L). High levels were also observed at Coomberdale (64.7 Bq/L) in Proterozoic chert, at Hopetoun (52.2 Bq/L) in

weathered granite, at Yuna and Kalbarri (51.7 and 31.5 Bq/L) in jointed Silurian sandstone aquifers, and at Menzies and Marble Bar (41.8 and 41.5 Bq/L) in fractured basalt.

Moderate concentrations were measured at Gingin within the aquifer in the Leederville Formation in the Perth Basin (22.7 Bq/L), particularly at shallow depth, and in aquifers within the Tertiary Plantagenet Group.

Lower groundwater-radon levels are generally associated with sedimentary rock aquifers, which have intergranular porosity. The lowest levels were measured at Millstream (1.8 Bq/L) in Tertiary calcrete; at Hopetoun (3.1 Bq/L) in Quaternary Tamala Limestone; in aquifers within the Mesozoic and younger sediments in the Perth Basin; and within shallow alluvial aquifers overlying basement rocks on the Yilgarn Craton. At Laverton, in the Goldfields, the Telegraph Shaft source has a relatively low radon concentration of 8.3 Bq/L, considering it is located in a fractured-rock aquifer. The Telegraph Shaft draws water from a network of flooded mine galleries and sumps, and radon levels are likely to be low due to a combination of loss by exsolution of radon from groundwater drainage into the air space underground; and loss by radioactive decay in the large volume of water stored in the sumps. Low and relatively uniform radon concentrations were measured at Collie in samples collected from Permian confined sand aquifers (8.8–13.8 Bq/L) located between major coal seams within the Collieburn and Cardiff Members.

Perth Metropolitan Area

The results from the Perth Metropolitan Area cover all of the confined- and most of the unconfined-aquifer production bores both within and outside the major groundwater schemes. Results are also included from observation bores constructed for studies of the unconfined aquifer at Whitfords, Nedlands, and Barragoon, and for the proposed North Coastal scheme. Post-treatment samples were collected at the groundwater treatment plants.

Overall, radon concentrations in the operational production bores were low, averaging 10.0 ± 6.1 Bq/L, and ranged from 1.9 Bq/L at Two Rocks in the unconfined Quaternary aquifer to 31.9 Bq/L at Mirrabooka in the confined Leederville Formation. Radon concentrations in the unconfined aquifers, were slightly lower on average than those in the confined aquifers, possibly due to a higher proportion of clays and heavy minerals in the latter.

The radon concentration in the Nedlands observation bores in the unconfined aquifer are low, averaging 6.0 ± 2.6 Bq/L. Similar low levels exist in the North Coastal scheme, Whitfords, and Barragoon bores. These bores are located within the unconfined aquifer which comprises Quaternary Tamala Limestone and Tertiary Ascot Limestone. Moderate radon concentrations (up to 81.2 Bq/L) were measured in the Tertiary Ascot Limestone at Whitfords and in the North Coastal scheme at specific depths where phosphatic nodules were abundant in the strata, adjacent to the bore screen. Analysis of a sample of phosphorated bone fragments from the Ascot Limestone showed the presence of significant concentrations of uranium (265 ppm). The phosphate nodules within the Ascot Limestone are probably derived from the Molecap Greensand (Coolyena Group). The distribution of uranium within the phosphate-nodule beds is unknown, and its effect upon radon levels in the unconfined aquifers is therefore uncertain.

Post-treatment samples contain low radon concentrations, except those from Halls Creek and Nullagine where, possibly, detention times are short in the treatment process and aeration is minimal. In addition, a number of schemes have post-treatment levels similar to or higher than the selected source bores. This probably results from short detention time, minimal aeration,

and the influence of supplies from other source bores. At Collie, groundwater drainage in the mine sumps has a low radon content due to radon loss by both exsolution through pressure release and radioactive decay during detention. Artificial ventilation of the mine galleries will continually remove the radon in the air that has come from groundwater drainage.

In the Perth Metropolitan Area, samples of the reticulated groundwater from the schemes have a very low radon content (averaging 1.6 ± 0.4 Bq/L), which indicates that the primary spray-aeration treatment for iron and hydrogen sulfide is effective in removing about 95 % of the dissolved radon in the source water.

Conclusions

Levels of radioactive radon-222 gas in public groundwater sources in Western Australia are low on average, though highly variable. Of the 69 WAWA schemes surveyed, only two have source (production

bore) radon levels exceeding the draft NHMRC guideline level for drinking water of 100 Bq/L. Post-treatment samples collected from the schemes all contain radon levels below the draft NHMRC guideline level. The survey results show that simple spray aeration in the treatment process is a very effective and low-cost means of reducing groundwater radon levels.

Variations in groundwater radon levels are controlled by local hydrogeological factors including the type and mineral composition of the aquifer. The main source of radon in groundwater is considered to be traces of radium-226 within the aquifer matrix. Generally, fractured-rock aquifers give rise to higher groundwater-radon levels than sedimentary-rock aquifers with intergranular porosity; however, evidence from this study indicates that sedimentary-rock aquifers that contain accumulations of uranium-bearing phosphate nodules and heavy-mineral sands are likely to impart higher than average radon levels to groundwater.

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