

# The use of Radon ( $\text{Rn}^{222}$ ) isotopes to detect groundwater discharge in streams draining Table Mountain Group (TMG) aquifers

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Environmental isotopes have been used for decades as natural tracers in studies aimed at understanding complex hydrogeological processes such as groundwater and surface water interactions. Radon ( $\text{Rn}^{222}$ ) is a naturally occurring, radioactive isotope which is produced from radium ( $\text{Ra}^{226}$ ) during the radioactive decay series of uranium ( $\text{U}^{238}$ ). Since  $\text{U}^{238}$  is present in most geological substrates,  $\text{Rn}^{222}$  is produced in various lithological structures and subsequently transported with groundwater through fractures and pore spaces in an aquifer towards surface water discharge points in rivers and springs. This study aimed to determine (i) the concentration of  $\text{Rn}^{222}$  within both surface water and groundwater in Table Mountain Group (TMG) aquifer systems, and (ii) the feasibility of using  $\text{Rn}^{222}$  isotopes as a natural tracer in groundwater-surface water interaction studies. This study was conducted in a highly fractured TMG aquifer system near Rawsonville, South Africa. Surface water from two perennial rivers (i.e. Gevonden and Molenaars), together with groundwater from a nearby borehole, were sampled and their corresponding  $\text{Rn}^{222}$  concentrations measured. Our study found median  $\text{Rn}^{222}$  concentrations in the Gevonden River of  $76.4 \text{ Bq}\cdot\text{L}^{-1}$  and  $67.2 \text{ Bq}\cdot\text{L}^{-1}$  in the dry and wet seasons, respectively. Nearly 12% of surface water samples exceeded  $100 \text{ Bq}\cdot\text{L}^{-1}$ . These abnormally high  $\text{Rn}^{222}$  concentrations can only be attributed to the influx of groundwater with extremely high  $\text{Rn}^{222}$  concentrations. Under ambient (no pumping) conditions,  $\text{Rn}^{222}$  concentrations in groundwater range between  $130\text{--}270 \text{ Bq}\cdot\text{L}^{-1}$ . However, when the borehole was pumped, and inflowing water from the surrounding aquifer was sampled, even higher  $\text{Rn}^{222}$  concentrations ( $391\text{--}593 \text{ Bq}\cdot\text{L}^{-1}$ ) were measured. These extremely high  $\text{Rn}^{222}$  concentrations in groundwater are believed to be attributed to the underlying granitic geology and the prevalence of faults. The use of  $\text{Rn}^{222}$  isotopes as an environmental tracer in groundwater-surface water interaction studies is therefore regarded as a feasible option in similar highly fractured aquifer systems.

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## INTRODUCTION

The interaction between surface water and groundwater is dynamic and complex (Sophocleous, 2002; Hunt et al., 2005). Understanding the linkages between groundwater and surface water bodies is critical for sustainable utilization and management of these complex systems. According to Wu et al. (2004), it is critical to establish the flow paths, patterns, water quantity and quality of the water flowing between surface water and groundwater, to develop and manage water resources efficiently. Since the properties of surface water and groundwater are usually chemically, physically and biologically different, the exchange of water between the two entities may have a significant impact on the water quality of either of these hydrological units (Kalbus et al., 2006).

Radioactive and stable isotopes have been applied as tracer techniques in many Earth systems studies world-wide (Thomas and Rose, 2003; Gibson et al., 2005; Kalbus et al., 2006; Praamsma et al., 2009). For roughly half a century, environmental isotopes have been used as natural tracers in studies aimed at understanding hydrogeological processes (Thomas and Rose, 2003). This field has developed in its scope and currently environmental isotopes are also used to study the exchange of water between groundwater and surface water sources (Midgley and Scott, 1994; Thomas and Rose, 2003; Wu et al., 2004). Other hydrological tracer studies include the application of stable isotopes such as  $^{18}\text{O}$  and  $^2\text{H}$  (e.g. Harrington et al., 2002; Hunt et al., 2005; Praamsma et al., 2009), as well as radioactive isotopes such as tritium (e.g. Scanlon, 2000),  $^{14}\text{C}$  (e.g. Harrington et al., 2002) and radon ( $\text{Rn}^{222}$ ) (e.g. Ellins et al., 1990; Kalbus et al., 2006; Moreno et al., 2014; Srinivasamoorthy et al., 2018).

$\text{Rn}^{222}$  is a naturally occurring, odourless, radioactive noble gas with a half-life of 3.83 days (Ellins et al., 1990; Wu et al., 2004).  $\text{Rn}^{222}$  isotopes are daughters produced from radium ( $\text{Ra}^{226}$ ) during the radioactive decay series of uranium ( $\text{U}^{238}$ ) (Broecker et al., 1967; Schubert et al., 2008). Since  $\text{U}^{238}$  is present in most rocks and has a half-life of 4.5 billion years,  $\text{Ra}^{226}$  and  $\text{Rn}^{222}$  are readily available in geological substrates such as granites, carbonaceous shale and metamorphic rocks (Hoehn and Von Gunten, 1989; Ellins et al., 1990; Schubert et al., 2008; Moreno et al., 2014).  $\text{Ra}^{226}$ , and subsequently  $\text{Rn}^{222}$  concentrations, tend to increase along faults and fractures, particularly when aquifers have been exposed to tectonic activities (Ellins et al., 1990; King et al., 1996; Vogiannis et al., 2004). Therefore, the lithology of the aquifer material influences the concentration of  $\text{Rn}^{222}$  found in the aquifer. The radioactive decay of  $\text{Ra}^{226}$  alongside and within the aquifer is the principal means for the accumulation of  $\text{Rn}^{222}$  in groundwater, whereby the  $\text{Rn}^{222}$  gases continuously diffuse into the pore spaces of the aquifer resulting in  $\text{Rn}^{222}$ -enriched groundwater (Loomis, 1987; Hoehn and Von Gunten, 1989; Cecil and Green, 2000; Wu et al., 2004). Consequently,  $\text{Rn}^{222}$  concentrations are significantly

higher in groundwater than in surface water bodies, with rainfall containing no  $Rn^{222}$ . High  $Rn^{222}$  concentrations measured in surface water may therefore be indicative of groundwater influx into the surface water source.

The application of radioactive isotopes, such as  $Rn^{222}$ , in hydrogeological studies has been mostly limited to studies in Asia, Europe, North and South America (e.g. Loomis, 1987; Hoehn and Von Gunten, 1989; Almeida et al., 2004; Moreno et al., 2014; Singh et al., 2018). Our study is one of the first in South Africa using  $Rn^{222}$  as an environmental tracer in hydrogeological studies between groundwater and rivers. Other studies, such as Eilers et al. (2015), Masevhe et al. (2017) and Botha et al. (2019), quantified  $Rn^{222}$  concentrations in either surface water or groundwater (not both) in different parts of South Africa. The objective of this study is to determine (i)  $Rn^{222}$  concentrations within both surface water and groundwater, and (ii) the feasibility of using  $Rn^{222}$  isotopes as a natural tracer in groundwater-surface water interaction studies in Table Mountain Group (TMG) aquifer systems.

### Study site

This study was conducted in the Rawsonville area (33° 42' 55" S; 19° 14' 48" E) of the Western Cape, which is situated in the winter rainfall region of South Africa (Fig. 1). The area is characterized

by a Mediterranean climate with cold, wet winters and warm, dry summers. The mean annual precipitation (MAP) for the area is approximately 800 mm, most of which occurs between the months of June and August (Conradie, 1995). The vegetation type may be described as mountainous fynbos (Mucina and Rutherford, 2006). The surrounding geology in this area comprises mainly of highly fractured TMG sandstone forming part of the Cape Supergroup, with scattered granitic outcrops (Thamm and Johnson, 2006).

The Gevonden and Molenaars rivers were studied to determine the interaction between groundwater and these surface water bodies using  $Rn^{222}$  isotopes. The Gevonden River is a small, perennial headwater stream which flows along the Waterkloof Fault before its confluence with the Molenaars River. The Molenaars River is a major perennial river with its headwaters situated in the Klein Drakenstein Mountains. The Molenaars River eventually joins the Breede River just northeast of Rawsonville. During the dry summer months, the Gevonden River is characterized by a low stream stage but often floods its banks during the wet season, which is a common characteristic of many fynbos mountain rivers (Brown et al., 2004). A borehole (BH3) situated within 100 m of the Gevonden River was sampled to determine groundwater  $Rn^{222}$  concentrations under both ambient (no pumping) and pumping conditions between both wet and dry seasons (Fig. 2).

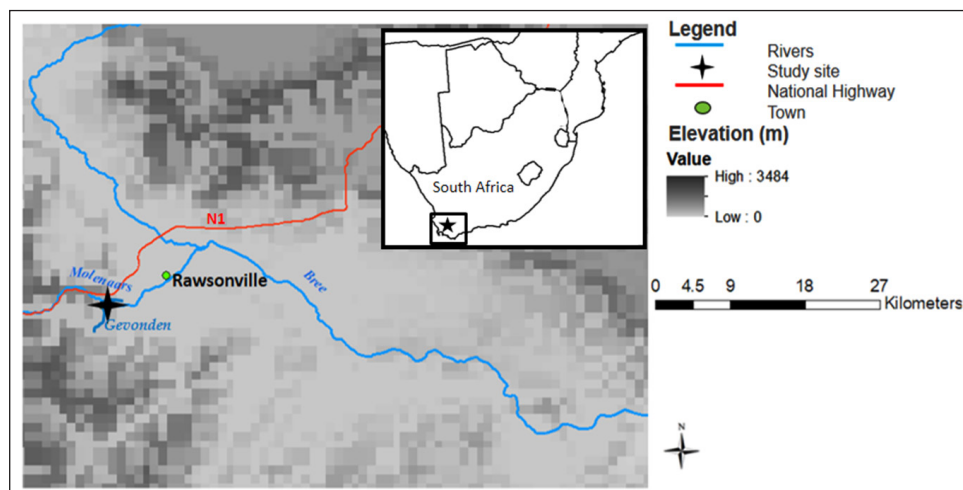


Figure 1. The study site location in relation to Rawsonville town and South Africa

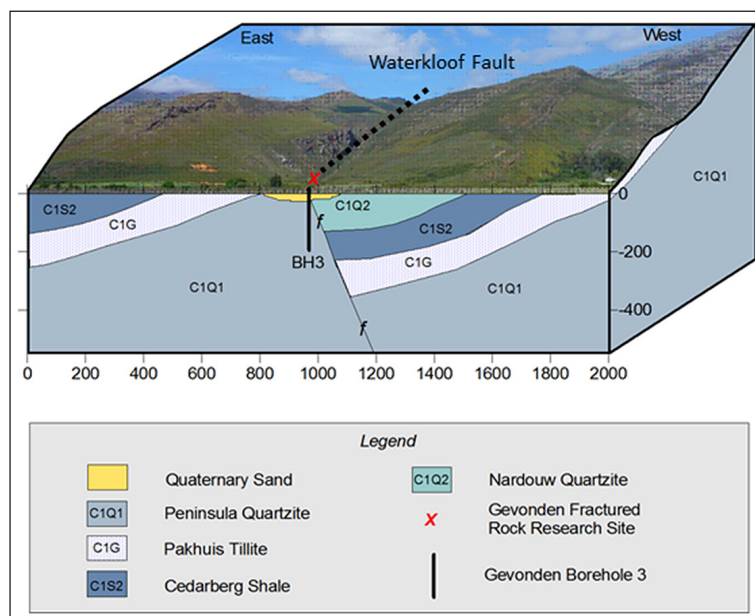


Figure 2. Conceptual geological cross-section for the Gevonden research site near Rawsonville

## METHODS

Surface water samples ( $n = 32$ ) were collected from the Gevonden and Molenaars rivers during both dry and wet seasons. Sampling points were randomly selected but also confined by accessibility constraints. Water samples were collected using 250 mL sample bottles, which were sealed under water to prevent  $\text{Rn}^{222}$  gases from escaping (Weaver et al., 2007). Samples were collected 15 cm below the water surface as to avoid the air-water interface where gaseous exchange would influence  $\text{Rn}^{222}$  concentrations (Ellins et al., 1990).

Groundwater samples were collected to represent various depths down the borehole profile (average of 25 m intervals) under both ambient conditions (i.e., no pumping) and pumping conditions (at 2 L/s pumping rate), with the use of a depth-specific sampler. This device is connected to a power source, and when lowered to a specific depth in the borehole a small pump displaces the water between two non-return valves and the sample is secured when the pump is switched off. Water was then carefully transferred into the plastic sample bottles.

The  $\text{Rn}^{222}$  concentrations were then analysed in a controlled laboratory environment using a RAD-7 electronic radon detector and RAD-H2O accessory (DurrIDGE Company, USA). This instrument is known to be very precise and capable of measuring concentrations as low as 1 mBq (Masevhe et al., 2017). Tests were conducted at normal room temperature with sufficient desiccant in the air sample path of the equipment (see Botha et al., 2019 for detailed protocol).

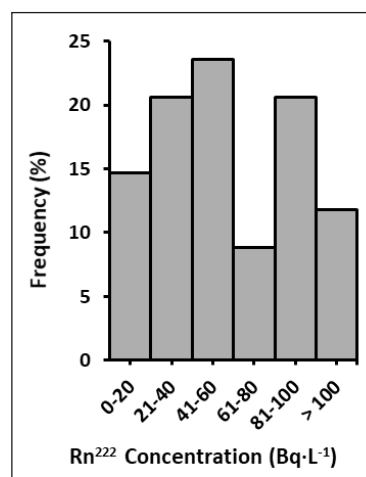
In order to correct for the radioactive decay between the time the sample was collected and the time at which  $\text{Rn}^{222}$  concentration was measured, the results were corrected using a decay correction factor (DCF) which is a simple exponential function (Eq. 1) adapted from DurrIDGE RADH20 Owner's Manual (2012). Initial  $\text{Rn}^{222}$  concentrations at time of sampling can be calculated based on the following formula:

$$C_{\text{Rn}} = \frac{C_M \cdot \exp^{\frac{t}{132.4}}}{1000} \quad (1)$$

Where  $C_{\text{Rn}}$  is the corrected  $\text{Rn}^{222}$  concentration ( $\text{Bq}\cdot\text{L}^{-1}$ ),  $C_M$  is the  $\text{Rn}^{222}$  concentration measured by the RAD-7 detector ( $\text{Bq}\cdot\text{m}^{-3}$ ),  $\exp$  refers to the exponential function and  $t$  is the time since the sample was collected (hours).

## RESULTS AND DISCUSSION

The results from this study suggest that  $\text{Rn}^{222}$  enriched groundwater discharges into both the Gevonden and Molenaars rivers. Less than 15% of the surface water sampled from the Gevonden and Molenaars rivers had  $\text{Rn}^{222}$  ranging between 1 and 20  $\text{Bq}\cdot\text{L}^{-1}$  (Fig. 3). Studies conducted in other rivers in various geologies around the world have found relatively low  $\text{Rn}^{222}$  concentrations typically ranging between 0.3 and 20  $\text{Bq}\cdot\text{L}^{-1}$  (Hall et al., 1985; Ellins et al., 1990; Wu et al., 2004; Schubert et al., 2008). During both wet and dry seasons,  $\text{Rn}^{222}$  ranged between 1 and 118.8  $\text{Bq}\cdot\text{L}^{-1}$  (Table A1, Appendix).  $\text{Rn}^{222}$  concentrations were considerably higher at suspected groundwater discharge sites along the rivers and would decrease substantially further away from these sites as  $\text{Rn}^{222}$  gases



**Figure 3.** The frequency of  $\text{Rn}^{222}$  concentrations measured in the Gevonden and Molenaars rivers

escape into the atmosphere. A recent study conducted in South African rivers in Gauteng Province by Masevhe et al. (2017), found low  $\text{Rn}^{222}$  concentrations ranging between 0.13 and 2.87  $\text{Bq}\cdot\text{L}^{-1}$ . The median  $\text{Rn}^{222}$  concentration in the Gevonden River is 76.4  $\text{Bq}\cdot\text{L}^{-1}$  and 67.2  $\text{Bq}\cdot\text{L}^{-1}$  in the dry and wet seasons, respectively (Table 1). With median  $\text{Rn}^{222}$  concentrations of 38.5  $\text{Bq}\cdot\text{L}^{-1}$  and 39.8  $\text{Bq}\cdot\text{L}^{-1}$  in the dry and wet seasons, respectively, the Molenaars River also exceeds typical  $\text{Rn}^{222}$  concentrations measured in rivers in other studies (Ellins et al., 1990; Schubert et al., 2008).

Under ambient (no pumping) conditions,  $\text{Rn}^{222}$  concentrations in groundwater range from 130–270  $\text{Bq}\cdot\text{L}^{-1}$  (Table 2). These are very high concentrations compared to groundwater samples from other global studies, which found  $\text{Rn}^{222}$  concentrations as low as 12  $\text{Bq}\cdot\text{L}^{-1}$  measured in the United Kingdom (Mullinger et al., 2007), 112.6  $\text{Bq}\cdot\text{L}^{-1}$  measured in Transylvania in Romania (Cosma et al., 2008), 26.6  $\text{Bq}\cdot\text{L}^{-1}$  detected in the volcanic regions of Spain (Moreno et al., 2014) and 16.1  $\text{Bq}\cdot\text{L}^{-1}$  in northern India (Singh et al., 2018). The use of  $\text{Rn}^{222}$  isotopes may not have been the most appropriate method in those studies due to the low concentrations detected. At our study site, slightly lower concentrations were detected at roughly 125 m, which may be attributed to a different fracture network contributing to the flow of water resulting in a decrease in  $\text{Rn}^{222}$ . Note the low concentration measured at 35 m closer to the surface where degassing may have occurred.

When the borehole was pumped and purged, the water entering the pump from the surrounding aquifer was sampled, and even higher  $\text{Rn}^{222}$  concentrations (391–593  $\text{Bq}\cdot\text{L}^{-1}$ ) were measured (Fig. 4). Generally,  $\text{Rn}^{222}$  concentrations in the pumped groundwater further increased during the wet season, probably due to active movement of water within the aquifer after being recharged and thus transporting  $\text{Rn}^{222}$  gases through the aquifer. It is likely that the Waterkloof Fault, on which the Gevonden and Molenaars rivers intersect, is the primary transfer mechanism for  $\text{Rn}^{222}$  gases to be mobilized by groundwater moving between  $\text{U}^{238}$  and  $\text{Ra}^{226}$  deposits within the TMG and released into the rivers via groundwater

**Table 1.** The variation in  $\text{Rn}^{222}$  concentrations in surface water samples in Rawsonville, South Africa

River	Season	N	Median	Min.	Max.
Gevonden	Dry	13	76.4	1	118.8
	Wet	12	67.2	33.5	104.9
Molenaars	Dry	4	38.5	36	43
	Wet	5	39.8	29.6	41.5

**Table 2.** Ambient Rn<sup>222</sup> concentrations in groundwater samples during the dry season

Borehole depth (m)	Rn <sup>222</sup> (Bq·L <sup>-1</sup> )
35	130
65	222
80	242
105	270
125	237
160	264

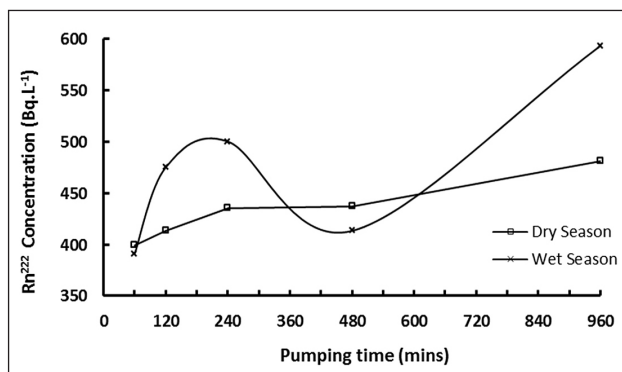
discharge points, which was particularly evident in the Gevonden River where very high Rn<sup>222</sup> concentrations were measured and decreased as the river flowed away from the fault. Similarly, Rn<sup>222</sup> concentrations in the Molenaars River spiked as it flowed over the Waterkloof Fault (see MR\_29 in Table A1, Appendix).

Other hydrogeological studies conducted within regions underlain by granitic geologies and along faults also measured high Rn<sup>222</sup> concentrations. For example, concentrations in the range of 2.1–653.5 Bq·L<sup>-1</sup> was measured in the Himalayas, India (Choubey et al., 2007), with 0.99–226.74 Bq·L<sup>-1</sup> measured along the North Anatolian Fault in Turkey (Akkaya et al., 2016) and 0.6–346 Bq·L<sup>-1</sup> was measured in the north-eastern region of Spain (Moreno et al., 2018). In the Karoo Basin in South Africa, a study by Eilers et al. (2015) found Rn<sup>222</sup> concentrations in groundwater ranging between < 10–163 Bq·L<sup>-1</sup> while Botha et al. (2019) found that 78% of their groundwater samples had Rn<sup>222</sup> concentrations below 60 Bq·L<sup>-1</sup>. Both these studies found considerably lower Rn<sup>222</sup> concentrations than we had measured in our study in the TMG.

Since the majority of the river samples had high Rn<sup>222</sup> concentrations, with roughly 12% of samples exceeding 100 Bq·L<sup>-1</sup>, it is suggested that Rn<sup>222</sup>-enriched groundwater discharged into the Gevonden and Molenaars rivers, leading to an increase in Rn<sup>222</sup> detected. Ellins et al. (1990) and Cook et al. (2003) found similar results which indicated an instant increase in Rn<sup>222</sup> concentrations in the immediate areas surrounding groundwater discharge points into rivers. Therefore, this study suggests that Rn<sup>222</sup> may be applied as a useful, natural radioactive isotope to determine groundwater discharge into surface water bodies in highly fractured aquifer systems such as the TMG. Furthermore, there is potential to use the Rn<sup>222</sup> concentration measurements to estimate groundwater flow volumes, both within the borehole as well as rate of groundwater discharge into surface water, improving our understanding of groundwater–surface water interaction in complex systems such as the TMG (Schubert et al., 2011; Kafri, 2001).

## CONCLUSIONS

Very high Rn<sup>222</sup> concentrations were measured in the Gevonden and Molenaars rivers due to Rn<sup>222</sup>-enriched groundwater discharging into the rivers. Groundwater in this region has extremely high Rn<sup>222</sup> concentrations (well above the global average) due to the underlying geology and proximity of nearby faults. This case study illustrates and verifies the application of Rn<sup>222</sup> isotopes as an environmental tracer in the assessment of groundwater–surface water interaction. It is particularly useful to apply in similar highly fractured aquifer systems such as the TMG. These radioactive isotopes have many of the required characteristics of an ideal environmental tracer, i.e., easily detectable in trace concentrations, chemically stable or inert for the required period of time, not typically found in large concentrations in surface water, present in most geological substrates and more importantly not filtered, absorbed nor adsorbed by the medium through which the water travels (Davis et al., 1980; Cecil and Green, 2000; Wu et al., 2004).



**Figure 4.** Rn<sup>222</sup> concentrations in groundwater samples under pumping conditions

Future studies could use Rn<sup>222</sup> isotopes in order to quantify groundwater influx into surface water, or vice versa, in order to promote the sustainable utilization of these water resources within the TMG region, which has gained particular focus in recent years due to the 2015–2017 drought in the Western Cape. However, we suggest that future studies invest in a high-intensity sampling campaign with increased sampling, both in groundwater and the river, in order to improve estimates and analytical rigour as well as provide greater detail on spatial variation of Rn<sup>222</sup> concentrations along specific river reaches.

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## REFERENCES

- AKKAYA G, KAHRAMAN A, KORAY A and KAYNAK G (2016) Variation in the radon concentrations and outdoor gamma radiation levels in relation to different geological formations in the thermal regions of Bursa, Turkey. *J. Radiol. Protect.* **36** (3) 490.
- ALMEIDA RMR, LAURIA DC, FERREIRA AC and SRACEK O (2004) Groundwater radon, radium and uranium concentrations in Região dos Lagos, Rio de Janeiro State, Brazil. *J. Environ. Radioact.* **73** (3) 323–334. <https://doi.org/10.1016/j.jenvrad.2003.10.006>
- BROECKER WS, LI YH and CROMWELL J (1967) Radium-226 and Radon-222: Concentration in Atlantic and Pacific Oceans. *Science*. **158** (3806) 1307–1310. <https://doi.org/10.1126/science.158.3806.1307>
- BROWN CA, BOUCHER C, PIENAAR E and PEMBERTON C (2004) Project Report: Effects of alien invasives on the Breede river. Department of Environmental Affairs and Forestry, Pretoria, South Africa.
- CECIL LD and GREEN JR (2000) Radon-222. In: Cook PG and Herczeg AL (eds.) *Environmental Tracers in Subsurface Hydrology*. Springer, Boston, USA.
- CHOUBEY VM, MUKHERJEE PK, BAJWA BS and WALIA V (2007) Geological and tectonic influence on water–soil–radon relationship in Mandi–Manali area, Himachal Himalaya. *Environ. Geol.* **52** (6) 1163–1171.
- CONRADIE WJ (1995) Efficiency of surface-applied ameliorants for alleviating acidity in two vineyard soil types. In: Date RA, Grundon NJ, Raymet GE and Probert ME (eds) *Plant-Soil Interactions at Low PH: Principles and Management*. Springer, Dordrecht.
- COOK PG, FAVREAU G, DIGHTON JC and TICKELL S (2003) Determining natural groundwater influx to a tropical river using radon, chlorofluorocarbons and ionic environmental tracers. *J. Hydrol.* **277** (1–2) 74–88. [https://doi.org/10.1016/S0022-1694\(03\)00087-8](https://doi.org/10.1016/S0022-1694(03)00087-8)

- COSMA C, MOLDOVAN M, DICU T and KOVACS T (2008) Radon in water from Transylvania (Romania). *Radiat. Meas.* **43** (8) 1423–1428. <https://doi.org/10.1016/j.radmeas.2008.05.001>
- DAVIS SN, THOMPSON GM, BENTLEY HW and STILES G (1980) Ground-water tracers – a short review. *Groundwater*. **18** (1) 14–23.
- DURRIDGE COMPANY (2012) *RAD7, RAD H20 Accessory Owner's Manual*. Durrige Company, Massachusetts.
- ELLINS KK, ROMAN-MAS A and LEE R (1990) Using <sup>222</sup>Rn to examine groundwater/surface discharge interaction in the Rio Grande de Manati, Puerto Rico. *J. Hydrol.* **115** (1–4) 319–341. [https://doi.org/10.1016/0022-1694\(90\)90212-G](https://doi.org/10.1016/0022-1694(90)90212-G)
- GIBSON JJ, EDWARDS TWD, BIRKS SJ, ST AMOUR NA, BUHAY WM, MCEACHERN P, WOLFE BB and PETERS DL (2005) Progress in isotope tracer hydrology in Canada. *Hydrol. Process.* **19** (1) 303–327. <https://doi.org/10.1002/hyp.5766>
- HALL FR, DONAHUE PM and ELDRIDGE AL (1985) Radon gas in ground water of New Hampshire. In: *Eastern Regional Ground Water Conference*, 16–18 July 1985, Portland, Maine (USA).
- HARRINGTON GA, COOK PG and HERCZEG AL (2002) Spatial and temporal variability of ground water recharge in central Australia: a tracer approach. *Groundwater*. **40** (5) 518–527.
- HOEHN E and VON GUNTEN HR (1989) Radon in groundwater: A tool to assess infiltration from surface waters to aquifers. *Water Resour. Res.* **25** (8) 1795–1803. <https://doi.org/10.1029/WR025i008p01795>
- HUNT RJ, COPLEN TB, HAAS NL, SAAD DA and BORCHARDT MA (2005) Investigating surface water–well interaction using stable isotope ratios of water. *J. Hydrol.* **302** (1–4) 154–172. <https://doi.org/10.1016/j.jhydrol.2004.07.010>
- KAFRI U (2001) Radon in groundwater as a tracer to assess flow velocities: two test cases from Israel. *Environ. Geol.* **40** (3) 392–398. <https://doi.org/10.1007/s002540000164>
- KALBUS E, REINSTORF F and SCHIRMER M (2006) Measuring methods for groundwater and surface water interactions: a review. *Hydrol. Earth Syst. Sci.* **10** (6) 873–887. <https://doi.org/10.5194/hess-10-873-2006>
- KING C-Y, KING B-S, EVANS WC and ZHANG W (1996) Spatial radon anomalies on active faults in California. *Appl. Geochem.* **11** (4) 497–510.
- LOOMIS DP (1987) Radon-222 Concentration and aquifer lithology in North Carolina. *Groundwater Monit. Remediation*. **7** (2) 33–39.
- MASEVHE L, MAVUNDA R and CONNELL S (2017) A general survey of radon concentration in water from rivers in Gauteng, South Africa using a solid-state  $\alpha$ -detector. *J. Environ. Anal. Toxicol.* **07** (04). <https://doi.org/10.4172/2161-0525.1000472>
- MIDGLEY JJ and SCOTT DF (1994) Use of stable isotopes of water (d and o-18) in hydrological studies in the Jonkershoek valley. *Water SA.* **20** (2) 151–154.
- MORENO V, BACH J, BAIXERAS C and FONT L (2014) Radon levels in groundwaters and natural radioactivity in soils of the volcanic region of La Garrotxa, Spain. *J. Environ. Radioact.* **128** 1–8. <https://doi.org/10.1016/j.jenvrad.2013.10.021>
- MORENO V, BACH J, ZARROCAM, FONT L, ROQUÉ C and LINARES R (2018) Characterization of radon levels in soil and groundwater in the North Maladeta Fault area (Central Pyrenees) and their effects on indoor radon concentration in a thermal spa. *J. Environ. Radioact.* **189** 1–13. <https://doi.org/10.1016/j.jenvrad.2018.03.001>
- MUCINA L and RUTHERFORD MC (2006) *The Vegetation of South Africa, Lesotho and Swaziland*. South African National Biodiversity Institute, Pretoria.
- MULLINGER NJ, BINLEY AM, PATES JM and CROOK NP (2007) Radon in chalk streams: Spatial and temporal variation of groundwater sources in the Pang and Lambourn catchments, UK. *J. Hydrol.* **339** (3–4) 172–182.
- PRAAMSMA T, NOVAKOWSKI K, KYSER K and HALL K (2009) Using stable isotopes and hydraulic head data to investigate groundwater recharge and discharge in a fractured rock aquifer. *J. Hydrol.* **366** (1–4) 35–45. <https://doi.org/10.1016/j.jhydrol.2008.12.011>
- SCANLON BR (2000) Uncertainties in estimating water fluxes and residence times using environmental tracers in an arid unsaturated zone. *Water Resour. Res.* **36** (2) 395–409. <https://doi.org/10.1029/1999WR900240>
- SCHUBERT M, SCHMIDT A, PASCHKE A, LOPEZ A and BALCÁZAR M (2008) In situ determination of radon in surface water bodies by means of a hydrophobic membrane tubing. *Radiat. Meas.* **43** (1) 111–120. <https://doi.org/10.1016/j.radmeas.2007.12.017>
- SCHUBERT M, BRUEGGEMANN L, KNOELLER K and SCHIRMER M (2011) Using radon as an environmental tracer for estimating groundwater flow velocities in single-well tests: RADON AS TRACER FOR GROUNDWATER FLOW ASSESSMENT. *Water Resour. Res.* **47** (3). <https://doi.org/10.1029/2010WR009572>
- SINGH B, KANT K, GARG M, SINGH A, SAHOO BK and SAPRA BK (2018) A comparative study of radon levels in underground and surface water samples of Faridabad district of Southern Haryana, India. *J. Radioanal. Nucl. Chem.* <https://doi.org/10.1007/s10967-018-6384-1>
- SOPHOCLEOUS M (2002) Interactions between groundwater and surface water: the state of the science. *Hydrogeol. J.* **10** (1) 52–67. <https://doi.org/10.1007/s10040-001-0170-8>
- SRINIVASAMOORTHY K, PONNUMANI G, PRAKASH R, GOPINATH S, SARAVANAN K and VINNARASI F (2018) Tracing groundwater inputs to Bay of Bengal from Sankarabarani River Basin, Pondicherry, India, using continuous radon monitoring. *Int. J. Environ. Sci. Technol.* <https://doi.org/10.1007/s13762-018-1938-x>
- THAMM AG and JOHNSON MR (2006) The cape supergroup. In: JOHNSON MR, ANHAEUSSER CR and THOMAS RJ (eds.) *The Geology of South Africa*. Council for Geoscience, Pretoria, South Africa.
- THOMAS J and ROSE T (2003) Environmental isotopes in hydrogeology. *Environ. Geol.* **43** (5) 532–532.
- VOGIANNIS E, NIKOLOPOULOS D, LOUIZI A and HALVADAKIS CP (2004) Radon variations during treatment in thermal spas of Lesvos Island (Greece). *J. Environ. Radioact.* **76** (3) 283–294.
- WEAVER JMC, CAVÉ L and TALMA AS (2007) *Groundwater Sampling* (2<sup>nd</sup> edn.). WRC Report No. TT 303/07. Water Research Commission, Pretoria.
- WU Y, WEN X and ZHANG Y (2004) Analysis of the exchange of groundwater and river water by using Radon-222 in the middle Heihe Basin of northwestern China. *Environ. Geol.* **45** (5) 647–653. <https://doi.org/10.1007/s00254-003-0914-y>

## APPENDIX

**Table A1.** A summary of Rn<sup>222</sup> concentrations, EC and pH in surface water samples. # represents the point at which the Molenaars River flows over the Waterkloof Fault

Sample code	GPS coordinates		River	Season	Rn <sup>222</sup> (Bq·L <sup>-1</sup> )	EC (μS·cm <sup>-1</sup> )	pH
GR_1	19° 14.719' E	33° 43.145' S	Gevonden	Dry	3.3	24	5.5
				Wet	104.93	27.4	6.53
GR_2*	19° 14.713' E	33° 43.184' S	Gevonden	Dry	99.2	24	5.5
GR_3*	19° 14.712' E	33° 43.181' S	Gevonden	Dry	4.6	24	5.6
GR_4	19° 14.714' E	33° 43.186' S	Gevonden	Dry	97.8	24	5.6
				Wet	103.57	24.4	6.7
GR_5	19° 14.723' E	33° 43.112' S	Gevonden	Dry	1	23	5.5
				Wet	96.41	24.95	5.69
GR_6*	19° 14.722' E	33° 43.103' S	Gevonden	Dry	118.8	27	5.6
GR_7	19° 14.728' E	33° 43.075' S	Gevonden	Dry	2.3	24	5.3
				Wet	86.01	25.5	7.37
GR_8*	19° 14.726' E	33° 43.067' S	Gevonden	Dry	103.9	27	5.6
GR_9	19° 14.730' E	33° 43.058' S	Gevonden	Dry	94.4	26	5.7
				Wet	85.65	24.77	5.85
GR_10	19° 14.753' E	33° 43.024' S	Gevonden	Dry	3.9	24	5.6
				Wet	74.54	24.7	5.48
GR_11	19° 14.760' E	33° 42.992' S	Gevonden	Dry	85.4	26	5.7
				Wet	59.85	16.8	5.61
GR_12*	19° 14.755' E	33° 42.983' S	Gevonden	Dry	76.4	24	5.5
GR_13*	19° 14.759' E	33° 42.969' S	Gevonden	Dry	76	26	5.2
GR_21*	19° 14.778' E	33° 42.932' S	Gevonden	Wet	54.89	17	5.78
GR_22*	19° 14.809' E	33° 42.885' S	Gevonden	Wet	50.19	25.63	5.93
GR_23*	19° 14.917' E	33° 42.847' S	Gevonden	Wet	47.30	27	6.07
GR_24*	19° 14.934' E	33° 42.847' S	Gevonden	Wet	46.15	27.85	5.89
GR_25*	19° 14.994' E	33° 42.861' S	Gevonden	Wet	33.46	28.1	6.28
MR_26*	19° 9.021' E	33° 43.351' S	Molenaars	Dry	36	33	6.9
				Wet	29.58	32.03	7.7
MR_27	19° 14.052' E	33° 42.323' S	Molenaars	Dry	39	32	6.3
				Wet	40.47	25.5	7.72
MR_28	19° 14.656' E	33° 42.601' S	Molenaars	Dry	38	32	6.3
				Wet	41.52	24.5	8.01
MR_29#	19° 15.152' E	33° 42.776' S	Molenaars	Dry	43	31	6.2
				Wet	39.82	25.4	9.07
MR_30	19° 10.179' E	33° 43.425' S	Molenaars	Wet	32.98	24.7	7.99

EC – electrical conductivity, \*Does not have a corresponding seasonal sample