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ORIGINAL PAPER

Method development for rapid quantification of Rn-222 in surface water and groundwater

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Abstract Understanding the risks of a developing unconventional hydrocarbons industry, including shale gas, to the chemical quality of surface water and groundwater involves firstly establishing baseline compositions against which any future changes can be assessed. Contaminants of geogenic origin are of particular interest and radon has been identified as one potential contaminant from shale sources. Robust measurement and monitoring of radon in water at environmental concentrations is essential for ensuring protection of water sources and maintaining public confidence. Traditional techniques for Rn-222 determination in water, such as inference by gamma spectrometry and direct alpha counting, are impractical for direct field measurement, and the relatively short half-life of Rn-222 (~ 3.82 days) means that longer analytical protocols from field to the laboratory may result in greater uncertainty for Rn-222 activity. Therefore, a rapid and low-cost method would be beneficial. We have developed and refined a laboratory procedure for Rn-222 monitoring using liquid scintillation counting (LSC). The accuracy of Rn-222 activities obtained via this procedure was evaluated by the analysis of almost 200 water samples collected from streams and boreholes as part of a detailed baseline investigation in the Vale of Pickering, Yorkshire, one potential location for future shale gas exploration. LSC was preferred for measurement of Rn-222 and had comparable accuracy to gamma spectrometry and direct alpha counting. The methodology provided a rapid, portable and low-maintenance option relative to the two established techniques and is shown to be a favourable choice for the measurement of radon in surface water and groundwater at environmental concentrations.

Keywords Radon · Groundwater · Health · Radioactivity · NORM · Liquid scintillation counting · Triathler

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Introduction

Rn-222 is a naturally occurring radioactive material (NORM) and product of the U-238 decay series,

forming directly from the α-particle decay of radium (Ra-226). It is one of the three naturally occurring isotopes of radon and is the most abundant isotope. As Rn-222 is a gas, it can enter households easily through emanation from underlying soil, or dissolved in water (Hopke et al. 2000). Inhalation and ingestion of Rn-222 are the two main exposure pathways (World Health Organization 2016), and domestic water supplies can give exposure through both pathways (drinking and release to indoor air via domestic washing/showering and sanitation). In the UK, the parametric value for radon in drinking water is 100 Bq L^{-1} (DWI 2018). Accurate monitoring of Rn-222 in drinking water is therefore critical to assessing an individual's risk of exposure.

Rn-222 can be released to groundwater by radioactive decay of radium and ultimately uranium in the aquifer minerals and so groundwater from uranium-rich hosts such as granite and shale is more at risk of accumulating elevated concentrations. The Vale of Pickering in Yorkshire is one area where exploration for shale gas has been proposed, and so obtaining a better understanding of the baseline concentrations of radon and other NORMs in regional surface water and groundwater has been a necessary step should any future exploration activity take place there (Kibble et al. 2014). This analytical method would not be limited to the UK, becoming a valuable technique for rapidly measuring Rn-222 activity in groundwater.

Hydraulic fracturing used for the extraction of natural gas from shale rock has the potential to exacerbate Rn-222 release from underground. This is done by pumping liquid chemicals (predominantly water based) down a borehole at a rate that will trigger the formation of fractures within reservoir rock to release natural gases for domestic and industrial consumption from shale rock (Gandossi and Von Estorff 2015). Radon is often trapped in brine held between layers of rock and sediment (e.g. shale), which can be released by fracturing. Baseline monitoring of groundwater Rn-222 concentrations is essential to generate knowledge of natural processes that release Rn-222 into groundwater, which can be compared to data collected from further monitoring following the commencement of hydraulic fracking in order to evaluate the potential geochemical impact of shale gas exploration (Appleton 2013; Mitchell 2013).

To quantify the Rn-222 activity in water, a liquid scintillation counting (LSC) technique was used to measure the activity of alpha- and beta-emitting radionuclides, such as Rn-222 in aqueous samples (Leaney and Herczeg 2006; Abesser 2009). Radon's greater solubility in organic solvents than in water results in a highly efficient transfer of Rn-222 from the water sample into the cocktail, optimising the number of decay events detected (Al-Azmi et al. 2004).

The aim of this research was to determine the feasibility of using a HIDEX Triathler liquid scintillation counter (Oy, Finland) for direct and rapid Rn-222 quantification in the laboratory. Although LSC is commonly employed in radiochemistry, there have been few studies publishing the use of the HIDEX Triathler for determination of radon (Celaya González et al. 2018). To assess this, Rn-222 measurements were obtained for 200 groundwater and 29 surface water samples collected over a two-year period (Smedley et al. 2017). The scope of investigations into the characterisation of the environmental baseline in the Vale of Pickering (water quality, air quality, seismicity and ground motion, soil gas) is detailed in Ward et al. (2016, 2017) and Smedley et al. (2017), along with the survey design and sampling sites used in this study. Further detail on the geology and hydrogeology of the Vale of Pickering can be found in Ford et al. (2015) and Bearcock et al. (2016).

Materials and methods

Sample collection

Water samples were collected for a series of analytical techniques to characterise the groundwater composition as well as determining radon activity by the principle and validation methods. Samples for LSC analysis were collected using 60 mL HDPE, wide-mouthed bottles that were submerged in water, allowing no headspace. The same method was followed for sample splits collected for gamma spectrometry (using 320 mL PMP pots) and direct alpha counting (using 2-L brown glass bottles). Filtered samples were also collected from each site and contained in 30 mL Nalgene^{TM} bottles preserved with 1% v/v HNO_3 for determination of a wide range of analytes by ICP-MS. Samples were then stored in cool boxes and delivered by courier or field worker to the British Geological Survey's Inorganic Geochemistry Laboratories (Nottingham, UK). Samples for

LSC were transferred into glass measurement vials upon receipt in the laboratory, 1–2 days after collection, and were measured within 1 day of receipt.

Analytical methodology

The Triathler is a single-sample LSC counter that possesses a multi-channel analyser, enabling the alpha and beta energy distribution of each isotope across several energy channels to be recorded. The alpha and beta counts are differentiated by pulse height discrimination, which aids the removal of interferences from other beta-emitting radionuclides and thus makes the subsequent calculation of Rn-222 activity more accurate (HIDEX 2015; Pates et al. 1993).

Groundwater Rn-222 activity was measured through mixing with a scintillation cocktail comprising PerkinElmer's Ultima Gold™ scintillators: Ultima Gold™ F (UGF) and Ultima Gold™ uLLT (UGuLLT). UGuLLT is optimised for ultra-low level LSC applications with a good sample uptake and is recommended for environmental monitoring applications. UGF is optimised for small sample volumes and can be combined with UGuLLT to provide an α/β optimised organic cocktail (Online Resource 1: Scintillator Ratio Trial). The benefits of mixing scintillants have also been discussed by Nikolova et al. (2018).

Ultima Gold™ F (6.7 mL) and Ultima Gold™ uLLT (3.3 mL) were combined in a 20-mL glass scintillation vial in a 2:1 ratio. After three inversions to mix the scintillants, 10 mL of sample water was injected below the scintillant, to minimise Rn-222 degassing, capped and inverted for a further 5 min, enabling the Rn-222 to diffuse into the scintillant across a large surface area.

Following from this, the vials were equilibrated at ambient temperature for 3 h, allowing other alpha-emitting radon daughter ingrowth. Each vial was then counted using the Triathler for 1 h. Rn-222 activity is calculated using Eq. 1 (Gallannaugh et al. 2016) which accounts for the presence of other alpha-emitting radionuclides in the sample:

Activity calibration

$$\text{Activity}\,(\text{Bq}\,\text{L}^{-1}) = \frac{(\text{Total}\,\alpha\text{CPM} - \text{Background}\,\alpha\text{CPM})}{(E \times V_{\text{water}} \times 60)} \quad (1)$$

where the αCPM represents the alpha counts per minute, E is the counting efficiency and V is the volume of the water sample (L). For this investigation, E was empirically calculated to be 1.96 for the Ultima Gold™ scintillants (Gallannaugh et al. 2016).

The method measures total alpha activity as the detector is unable to resolve the emission attributable to Rn-222 (5.489 MeV) from its daughters Po-218 (6.002 MeV) and Po-214 (7.687 MeV). This effect is accounted for by the counting efficiency factor used in Eq. 1. The potential for interference from Ra-226 alpha activity (4.784 MeV) is mitigated in this work as the samples contained negligible radium concentrations. Samples re-analysed 28 days after collection showed that radon levels had dropped, suggesting that no radium was present in the groundwater (Gallannaugh et al. 2016). If radon levels had remained unchanged, radium daughters would still have been being produced suggesting a persistent source of radon.

This refined procedure was established from prior experimental tests for determining the influence of scintillant brand, scintillant ratios (Ultima Gold™ F: Ultima Gold™ uLLT) and analytical run time on Rn-222 values obtained by the Triathler, to determine how to maximise the accuracy of the technique (Gallannaugh et al. 2016) (Online Resources 1 and 2: Scintillator Ratio Trial; Analytical Run Time Trial).

Schubert et al. (2012) found that partitioning of radon between air and water was affected by water salinity and temperature. The influence of sample salinity (comparing chloride concentrations of 10 mg L^{-1} and 100 mg L^{-1} against a control sample), alkalinity (bicarbonate concentration 100 mg L^{-1} and 500 mg L^{-1}) and sample temperature (4 °C and 30 °C) on Rn-222 measurements generated by the Triathler were tested for robustness across a range of environmental, geological and seasonal influences. Mean activities for all tests are generally within mean ± 2 standard deviations of the reference sample (Online Resources 3 and 4: salinity and alkalinity; temperature influence results) suggesting that variation in sample salinity and alkalinity and sample temperature in the ranges investigated have negligible effect on determination or radon activity.

Rn-222 activities for these water samples were also measured by two widely accepted techniques for Rn-222 quantification: direct alpha counting (Saphymo

AlphaGUARD^{TM}) and gamma spectrometry (Canberra BEGe; Rn-222 inference using Pb-214 and Bi-214 daughter nuclide activities), to assess the accuracy of the Triathler measurements (Canet and Jacquemin 1990; Schubert et al. 2006). The direct alpha counting method was carried out by bubbling air though 200 mL of sample in an enclosed system and the output monitored for 40–60 min to identify the maximum activity. The gamma spectrometry method counted the 270 mL sample for a period of 24 h.

Quality control

Method bias was assessed using an IAEA proficiency test QC sample with documented Ra-226 activity (IAEA-TEL-2014-03, (IAEA 2014)). A sample was prepared using this water sample and allowed to stand for 25 days to allow ingrowth of Rn-222. The documented and measured activity values for the IAEA QC sample are summarised in Table 1 with the calculated recovery of this method for aqueous sample measurement. The method was shown to have a mean recovery of 90%.

A Ra-226 standard with known activity was diluted and used as a QC; precision was calculated from ten replicate analyses over three runs (relative standard deviation of 5.4%). Multiple blank measurements were obtained using the Triathler to ascertain background values and calculate a working limit of detection: 0.5 Bq L^{-1} (Online Resource 5: Limit of detection) which is significantly lower than the recommended detection limit for monitoring requirements for radon in drinking water of 10 Bq L^{-1} (DWI 2018). Uncertainties associated with each of three techniques tested are summarised in Online Resource 6: Method comparison.

Statistical analysis

Differences in median Rn-222 activity between samples were assessed using nonparametric Mann–Whitney–Wilcoxon test, owing to the non-normal distribution of data for Rn–222 activity. Differences between the three analytical methods were assessed using Pearson correlation analysis. Statistical analyses were performed with the Minitab statistics package (v.17).

Results and discussion

Liquid scintillation method validation

An inter-method comparison was conducted using Rn-222 activity data collected by LSC with the Triathler (n = 113), direct alpha counting with the AlphaGUARD (n = 93) and through inference from daughter nuclide activity (Pb-214 and Bi-214) using gamma spectrometry (n = 46). A strong positive correlation (Fig. 1 and Online Resource 6: Method comparison) was found between the Rn-222 data obtained by the Triathler and data obtained by gamma spectrometry ($r = 0.829, p < 0.001$) and AlphaGUARD ($r = 0.855$, $p < 0.001$). It should be noted that the apparent underestimation by the Triathler compared to gamma spectrometry (slope of 1.2) is comparable with the observed recovery from the IAEA sample of 86–92% (Table 1).

The calculated measurement uncertainties for the Triathler LSC data are typically in the range 8–17%, for gamma spectrometry 5–21% and for direct alpha counting 11–44% (individual measurement uncertainties are listed in Online Resource 6: Method comparison). The precision of the Triathler method is comparable with that achieved by gamma spectrometry and significantly better than can be achieved when using the direct alpha counting method used here.

Table 1 IAEA spiked water QC sample documented and measured activity (Triathler) (IAEA 2014)

	Documented Ra-226 activity (Bq kg^{-1})	Measured Rn-222 activity (Bq kg^{-1})	Recovery (%)
Neat	17.9 ± 0.1	16.5	92
Diluted × 2	8.95 ± 0.05	7.72	86
Diluted × 4	4.48 ± 0.025	4.12	92

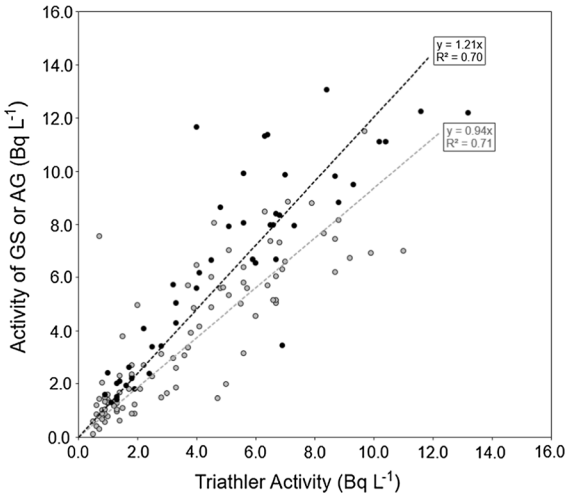

Fig. 1 Rn-222 activity values obtained from the Triathler for each site relative to the Rn-222 activity values obtained with gamma spectrometry (black circles) and direct alpha counting (grey circles)

There are many other benefits associated with the Triathler relative to these two techniques. Firstly, the Triathler produces reliable Rn-222 activity measurements in 1 h compared to a typical measurement time of 24 h for gamma spectrometry, essential given the Rn-222 relatively short half-life of 3.8232 days (Nucléide LARA 2017). Secondly, the identification and interpretation of the Pb-214 and Bi-214 peaks on the resulting spectra from gamma spectrometry requires experienced analytical skill and knowledge, making this technique less accessible to field monitoring staff. As the gamma spectrometry castles are immobile, expensive and require regular cryogenic replenishment, this technique is only suitable for a laboratory environment, which may be some distance from the field site. Thirdly, whilst the AlphaGUARD is portable like the Triathler and capable of providing reliable data in the same duration of time (Schubert et al. 2006), this technique required constant monitoring to determine when the peak in Rn-222 activity occurs, which will vary by sample. The Triathler does not require constant attention, finishing its analysis after the desired run time duration without supervision, allowing the field or laboratory operator to conduct other activities. It can be concluded therefore that the Triathler is a reliable and fit-for-purpose technique for Rn-222 quantification. It should be noted that the apparent underestimation (slope of 1.2) is comparable to gamma spectrometry aligns with the observed recovery of 86–92% (Table 1).

Comparison of Rn-222 activities by water source

Median concentrations for Rn-222 were 3.20 Bq L^{-1} and 0.93 Bq L^{-1} for groundwater (n = 200) and surface water (n = 29), respectively (Fig. 2, Online Resource 7: Geological and water source). Median groundwater Rn-222 activities (Mann–Whitney $W = 25059$, $p < 0.001$) were significantly higher than surface water activities, as expected from loss from surface water by degassing (Cosma et al. 2008; Jobbágy et al. 2017).

Comparison of Rn-222 activities by aquifer

Groundwater is abstracted from two distinct aquifers in the study area: superficial deposits here defined as Quaternary fluviolacustrine sediments together with uppermost sections of the Jurassic Kimmeridge Clay, and the regionally important Jurassic Corallian Limestone (Bearcock et al. 2016; Smedley et al. 2017). The median Rn-222 activities were significantly greater for groundwater samples collected from the Corallian limestone than those from superficial deposits (Mann–Whitney $W = 8114$, $p < 0.001$). Median Rn-222 concentrations (Fig. 3) for groundwater samples collected from the Corallian limestone (n = 65) and superficial deposit geology types (n = 135) were 6.01 Bq L^{-1} and 1.91 Bq L^{-1}, respectively (Online Resource 7:

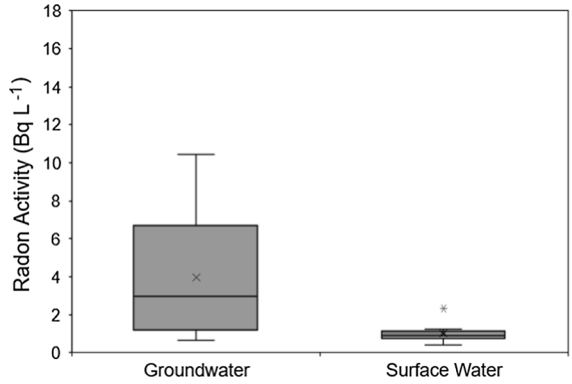

Fig. 2 Mean (×), median and interquartile range (IQR) and full range of Rn-222 activity values obtained for the two water types collected from the Vale of Pickering (* indicates an outlier more than 1.5 times the IQR greater than the third quartile)

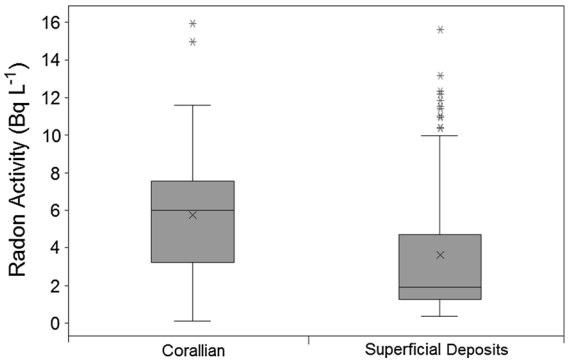

Fig. 3 Mean (×), median and interquartile range (IQR) and full range of Rn-222 activity values obtained for groundwater samples collected from the two aquifers for the Vale of Pickering: Corallian Limestone and Superficial Deposits (* indicates outliers more than 1.5 times the IQR greater than the third quartile)

Geology and water source). This is directly in line with the findings from the PHE/BGS indicative atlas of radon (Miles et al. 2007). The maximum measured activity was 16 Bq L^{-1}, an order of magnitude lower than to the specified value for monitoring requirements for radon in drinking water of 100 Bq L^{-1} (DWI 2018).

The Rn-222 source for these two lithologies is considered to be uranium-bearing minerals including phosphatic horizons in the Corallian (Bearcock et al. 2016; Hemingway 1974; Skeppström and Olofsson 2007). Geochemical affinities govern the distribution of precursor radionuclides in rocks, from which radon will pass into groundwater, which can be drastically changed by secondary geological processes, e.g. groundwater migration or diagenetic changes, thereby causing unexpectedly high radon concentrations in otherwise unsuspicious areas, e.g. in outcrops of mineralised fault and fracture zones and clay-rich residual soils derived from carbonates (Kemski et al. 1996). A full understanding of the causes of the differences in radon concentrations between the two aquifers would require further investigation and is beyond the scope of this study.

Conclusion

The Triathler liquid scintillation counter provides fit-for-purpose quantification of Rn-222 activity in the laboratory for routine surface and ground water monitoring, demonstrated by the analysis and comparison of over 200 water samples, to the other known Rn-222 quantification techniques. Laboratory experimentation supported the development and refinement of the Rn-222 quantification method, enhancing the practicality and data quality of the procedure.

The application of LSC for this study enabled a background Rn-222 activity to be established for several sites in one study area. This provides a valuable baseline for comparing with new Rn-222 data acquired should any shale gas exploration take place. This technique could be used to determine baseline Rn-222 activities in settings with limited laboratory infrastructure, or for monitoring environmental radioactivity hazards in remote locations.

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Compliance with ethical standards

Conflict of interest It is declared by all authors that this study involves no conflict of interest.

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